

Radiative perturbation theory: a review

Michael A. Box *

School of Physics, University of New South Wales, Sydney, NSW 2052, Australia

Abstract

Radiative perturbation theory is a computational technique which can greatly ease the burden of repeated solution of the radiative transfer equation for model atmospheres which differ from one another by only relatively small changes in some of the optical parameters. It requires the solution of both the radiative transfer equation, and its adjoint, followed by some usually straightforward integrations of these solutions. In this work we review the theoretical structure of the technique, and discuss a series of applications which have already demonstrated its utility. Future developments of the theory, and new applications currently under consideration, are also discussed. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Many environmental modelling problems call for the repeated solution of the radiative transfer equation, often for situations where the optical/radiative properties of the medium do not differ greatly from one case to the next. Some examples include global climate and numerical weather prediction modelling, where solutions are required at each time step and grid point; computation of photochemical reaction rates or u.v. indices, which require the integration over wavelength of the product of the relevant radiation quantities and the appropriate reaction cross sections; and the inversion of multiangular (or multispectral) radiance measurements, taken either at the ground or from space, to retrieve atmospheric geophysical data, which often calls for iterative solution methods.

One approach to this challenge is to turn to faster methods of solution, such as the delta-four-stream or two-stream methods. However, the gain in execution time may well come at the expense of necessary accuracy of solution. Another approach to this dilemma is radiative perturbation theory, a technique borrowed from the mathematically similar field of neutron transport theory (Bell and Glasstone, 1970; Marchuk and Lebedev, 1986: in fact the idea seems to go back at least

as far as Wigner, 1945). With this approach, one first performs two accurate computations for a certain ‘base case’ atmospheric optical model, and then perturbs this base model by making small changes to the optical parameters. Results for this perturbed case may then be obtained by a very simple integration over the solution of the base case.

In Section 2, we introduce the operator notation which is essential for the efficient formulation of perturbation theory, and also define the adjoint operator. First order perturbation theory is outlined in Section 3, while Section 4 discusses a number of approaches to extend beyond the first order. The perturbation technique may be applied to a very wide variety of problems in atmospheric radiation. Three such examples are discussed in detail in Section 5. In Section 6 we look at some of the work in progress across the atmospheric radiation community, as well as future applications which are on the drawing board. Finally, in Section 7 some brief comments are made to assist readers who may wish to modify their codes to incorporate perturbation theory calculations.

2. Operator notation

2.1. The transport operator

In a plane-parallel scattering and absorbing atmosphere, the equation of radiative transfer may be written

* Tel.: +612-9385-4545; fax: +612-9385-6060.

E-mail address: m.box@unsw.edu.au (M.A. Box).

$$\mu \frac{\partial I(z, \Omega)}{\partial z} = \sigma_s(z) \int_{4\pi} p(z, \Omega' \rightarrow \Omega) I(z, \Omega') d\Omega' - \sigma_t(z) I(z, \Omega) + Q(z, \Omega) \quad (1)$$

Here $I(z, \Omega)$ is the radiance at altitude z , travelling in the direction $\Omega = (\theta, \phi)$ (where θ is the zenith angle and ϕ is the azimuth angle); $\sigma_s(z)$ and $\sigma_t(z)$ are respectively the scattering and total cross-sections per unit volume; p is the normalized phase function for scattering from one direction to another; and Q represents all sources of radiation, including incoming solar illumination from the direction Ω_0 ,

$$Q = \mu F_0 \delta(z - z_T) \delta(\Omega - \Omega_0) \quad (1a)$$

(where $\mu = \cos\theta$), and atmospheric thermal emission,

$$Q = (1 - \omega_0) \sigma_t(z) B(T(z)) \quad (1b)$$

where B is the Planck function for temperature T . In practice, the vertical coordinate is usually converted to optical depth. However, for reasons which will become apparent below, we find it more convenient to retain the physical coordinate in our formalism.

In order to simplify notation in what follows, and to help guide the reader's thinking, we re-write this equation in an operator notation as follows

$$LI(z, \Omega') = Q(z, \Omega) \quad (2)$$

where the transport operator, L , is clearly given by

$$L \equiv \mu \frac{\partial}{\partial z} + \sigma_t(z) - \sigma_s(z) \int_{4\pi} d\Omega' p(z, \Omega' \rightarrow \Omega) \circ \quad (3)$$

and the \circ notation is used to indicate that the final term is an integral operator, and not a definite integral. (Note that technically there should also be a $\delta(\Omega' - \Omega)$ term multiplying the first two terms of this definition: we omit this for convenience.)

2.2. Adjoint operator

We now need to introduce the adjoint to this operator. Consider a set of functions $\{I(z, \Omega)\}$ subject only to a set of boundary conditions which will be discussed below. Now we introduce a second set of functions $\{I^+(z, \Omega)\}$, subject to their own boundary conditions. Then for a given operator L , its adjoint, L^+ , is defined by requiring that

$$\langle I^+, LI \rangle = \langle L^+ I^+, I \rangle \quad (4)$$

for all I belonging to the set $\{I\}$, and all I^+ belonging to the set $\{I^+\}$. Here we have introduced the notation that angular brackets indicate the inner product; that is integration over the free, or 'phase space' variables:

$$\langle f_1, f_2 \rangle \equiv \int dz \int d\Omega f_1(z, \Omega) f_2(z, \Omega) \quad (5)$$

These integrals are to be understood as spanning the full range of the corresponding variables: from 0 to z_T (the top of the atmosphere); and 4π .

The actual form of the adjoint operator, for a given operator L , will depend on the boundary conditions imposed on both sets of functions. This provides some freedom of choice. It is shown by Marchuk (1964), Bell and Glasstone (1970) and Box et al. (1988a), that if we impose the standard vacuum boundary conditions on the set $\{I\}$, namely

$$I(z_T, \mu, \phi) = 0 \text{ for } -1 \leq \mu < 0$$

and

$$I(0, \mu, \phi) = 0 \text{ for } 0 < \mu \leq 1 \quad (6)$$

(that is, no incoming radiance, apart from Q), and impose complementary boundary conditions of no outgoing adjoint radiance on the set $\{I^+\}$, that is

$$I^+(z_T, \mu, \phi) = 0 \text{ for } 0 < \mu \leq 1$$

and

$$I^+(0, \mu, \phi) = 0 \text{ for } -1 \leq \mu < 0 \quad (7)$$

then the adjoint operator is given by

$$L^+ = -\mu \frac{\partial}{\partial z} + \sigma_t(z) - \sigma_s(z) \int d\Omega' p(z, \Omega \rightarrow \Omega') \circ \quad (8)$$

(Proof of this result is quite straightforward, requiring only one integration by parts, and use of the fact that the phase function depends only on the scattering angle. Surface reflection may also be included: Marchuk, 1964.)

Now that we have the adjoint transport operator, we may write down an adjoint transport equation, which must take the general form

$$L^+ I^+(z, \Omega') = Q^+(z, \Omega) \quad (9)$$

Initially Q^+ may be regarded as a completely arbitrary adjoint source function.

It may seem that solving this adjoint equation would require the development of a totally new computer code. However, this is not the case. Consider solving the transport equation

$$L\Psi(z, \Omega') = Q^+(z, -\Omega) \quad (10)$$

Then it has been shown by Bell and Glasstone (1970) and Box et al. (1988a) that

$$I^+(z, \Omega) = \Psi(z, -\Omega) \quad (11)$$

That is to say, in order to solve the adjoint transport equation for a given (adjoint) source, one first solves the normal (or forward) transport equation for an angle-reversed source, and then reverses all directions on the solution. (Ψ is referred to as the pseudo-radiance.)

2.3. Radiative effects

When one solves the radiative transfer equation the solution is, at least in principle, the complete radiance distribution function, $I(z, \Omega)$. (Some solution techniques provide less than this, of course, and some computer codes only output a subset of this information, even though they generate a representation of the full radiance.) In general, the user is interested in only a small subset of this information — maybe even a single number, such as the flux at the ground. We will refer to such specific pieces of information (single numbers) as radiative effects, E .

Any radiative effect must be able to be extracted from the full radiance distribution. Formally, we may perform this extraction using a suitable response operator (or response function), R , and the following functional relation:

$$E = \langle R, I \rangle \quad (12)$$

For example, the response function for the net flux at an altitude z_0 is clearly

$$R = \mu \delta(z - z_0) \quad (13a)$$

whereas for the actinic flux (average intensity) at that level

$$R = \delta(z - z_0) \quad (13b)$$

Thus, in order to compute say, the flux at the ground due to solar illumination at a certain zenith angle, one firstly solves the radiative transfer equation for this specific source, and then extracts the relevant information by suitably integrating the radiance at ground level. Of course, many computer codes are constructed in such a way that these operations are performed as a matter of course. (Note that the radiance, I , used in Eq. (12) is the full radiance — both the diffuse (scattered) component, and the direct solar beam. Many codes separate these two components: they must be re-combined at some point.)

However, using the adjoint formulation, there is another way to this destination. Consider what happens if we choose to use the response function, R , as an adjoint source, Q^+ . The adjoint transport equation becomes

$$L^+ I^+ = R \quad (14)$$

If we now take the inner product of this equation with I , and use Eq. (4), we obtain

$$\langle R, I \rangle = \langle L^+ I^+, I \rangle = \langle I^+, L I \rangle = \langle I^+, Q \rangle \quad (15)$$

where we have also used Eq. (2). Thus we see that we have two paths to the effect, E :

$$E = \langle R, I \rangle = \langle I^+, Q \rangle \quad (16)$$

What this result is telling us is that we may obtain E

either by starting with the source Q , solving the radiative transfer equation to obtain I , and extracting E using the response function R (the standard approach): or starting with the response function R as adjoint source, solving the adjoint transport equation, and extracting E using the original source Q . In a photon flow picture, these are time-reversed processes, although the radiative transfer equation is, of course, time independent.

Which approach is better? The answer to that question depends on the task at hand. If one is seeking multiple effects from a single source (for example, the flux at a series of levels, for a fixed solar zenith angle), then the normal (or forward) approach is better. However, if one is seeking a single effect, but for multiple sources (for example, surface flux as a function of solar zenith angle), then the adjoint approach is far more efficient, as only a single solution to the transport equation is needed (Gerstl, 1982). In addition, the response functions used as adjoint sources are usually much smoother than the collimated solar beam, which can simplify the computational task.

It should also be pointed out that adjoint techniques have been employed in a number of environmental areas (Marchuk, 1995), including variational assimilation of meteorological (and other) data, and in climate modelling: see for example Hall et al. (1982).

3. Perturbation theory

3.1. Formulation

Suppose now that we have solved the (forward) radiative transfer equation, *and* its adjoint, for a certain base model of the atmosphere, which we may characterize by its transport operator, L_0 , and adjoint transport operator, L_0^+ , and assume (of course) that we have chosen a certain radiative effect, characterized by the response function R . That is, we have solved the two equations

$$L_0 I_0 = Q \quad (17)$$

and

$$L_0^+ I_0^+ = R \quad (18)$$

The solutions to these two equations are referred to as the base case radiance, and base case adjoint radiance, respectively, as indicated by the subscript 0 on each. From either of these solutions we may obtain the base case value of the effect, E_0 .

Suppose now that we wish to find the new value of this effect, corresponding to a new atmospheric optical model (but for the same source, for example the same solar zenith angle), with its own transport operator, L , and its adjoint, L^+ . Can we use the information we currently have, in order to obtain this new value, or at least some approximation to it?

Perturbation theory provides a positive answer to this question. It is straightforward to show that (Marchuk, 1964; Bell and Glasstone, 1970; Box et al., 1989)

$$E = E_0 - \langle I_0^+, \Delta LI_0 \rangle + \langle \Delta I^+, L \Delta I \rangle \quad (19)$$

where

$$\Delta L = L - L_0 \quad (20a)$$

$$\Delta I = I - I_0 \quad (20b)$$

and

$$\Delta I^+ = I^+ - I_0^+ \quad (20c)$$

Since we don't know either I or I^+ (as we haven't solved the corresponding transport equations), we cannot evaluate the second term in Eq. (19). However, if the difference in the two optical models (that is, the difference between the two transport operators) is sufficiently small, both of these components should also be small. Hence this term contains two small components ('second order of smallness'), while the first term contains only one small component. Thus, as a first approximation, we will ignore the second term in Eq. (19), and write

$$E \cong E_0 - \langle I_0^+, \Delta LI_0 \rangle \quad (21)$$

This is the standard form of first order perturbation theory. (In Section 4 we discuss ways to go beyond this first order term.)

Marchuk (1964) has actually presented a more general form of Eq. (21) for the case where the source, Q , may also be considered variable — for example, thermal emission. In our notation, and with the above assumptions, his result may be written

$$E \cong E_0 + \langle I_0^+, \Delta Q - \Delta LI_0 \rangle \quad (21a)$$

This idea has recently been re-addressed by Ustinov (1990, 1991).

The transport operator (or the optical model of the atmosphere) may be characterized by a (possibly large) number of parameters — for example, the Rayleigh scattering optical thickness, the ozone absorption optical thickness, the aerosol optical thickness, single scattering albedo, and the coefficients of the Legendre expansion of the phase function, defined by

$$p(z, \Omega' \rightarrow \Omega) = \sum_n (2n+1) \chi_n(z) P_n(\Omega' \rightarrow \Omega) / 4\pi \quad (22)$$

where P_n is the Legendre polynomial of order n . (In practice, the atmosphere will be divided into a series of layers, in each of which parameters such as χ_n will be held fixed.)

Many applications of perturbation theory may be regarded as the variation of one of these parameters (or the parallel variation of several parameters). In such cases, it is convenient to re-write Eq. (21) in the form

$$E \cong E_0 - \delta \langle I_0^+, \Delta LI_0 \rangle \quad (21b)$$

where δ is a 'scale parameter' indicating the magnitude of the perturbation. Eq. (21b) has the form of the first term of a Taylor series, so that we may regard the actual perturbation term (the term in angular brackets) as a partial derivative in 'parameter space'.

3.2. The perturbation integral

The inner product in Eq. (21b) is a multiple integral, as can be seen from Eqs. (5) and (3). Evaluation of such perturbation integrals depends on the nature of the problem under investigation. By making use of the addition theorem for Legendre polynomials (Arfken, 1970, section 12.8) to expand the phase function, we may express the difference transport operator, ΔL , in the form (Box et al., 1989):

$$\begin{aligned} \Delta L = & \Delta \sigma_t(z) - \int_{-1}^1 d\mu' \int_0^{2\pi} d\phi' \sum_{n=0}^{\infty} (2n+1) \\ & \times \Delta \eta_n(z) / 4\pi \sum_{m=0}^n (2 - \delta_{0m}) \\ & \times \frac{(n-m)!}{(n+m)!} P_n^m(\mu) P_n^m(\mu') \cos m(\phi - \phi') \end{aligned} \quad (23)$$

where

$$\Delta \sigma_t(z) = \sigma_t(z) - \sigma_t^0(z) \quad (24a)$$

$$\Delta \eta_n(z) = \eta_n(z) - \eta_n^0(z) \quad (24b)$$

and we have defined

$$\eta_n^0(z) = \sigma_s^0(z) \chi_n^0(z) \quad (25)$$

with a similar expression for the perturbed case. (Note that χ_0 is unity, to ensure the correct normalization of the phase function.) Note that the derivative term in the transport operator is not present in the difference operator, due to our choice of physical depth, rather than optical depth, as vertical coordinate. (For this reason, the adjoint difference operator is identical to the forward difference operator.)

In those cases where the effect being considered is independent of azimuth — for example flux, actinic flux, or heating rates — the adjoint intensity will also be independent of azimuth. In such cases, the perturbation integral reduces to (Box et al., 1989):

$$\begin{aligned} \Delta E = & 2\pi \int dz \left\{ \Delta \sigma_t(z) \Xi(z) - \right. \\ & \left. \frac{1}{2} \sum_n (2n+1) \Delta \eta_n(z) \xi_n^+(z) \xi_n^-(z) \right\} \end{aligned} \quad (26)$$

where

$$\xi_n(z) = \int_{-1}^1 d\mu I_0(z, \mu) P_n(\mu) \quad (27a)$$

$$\xi_n^+(z) = \int_{-1}^1 d\mu I_0^+(z, \mu) P_n(\mu) \quad (27b)$$

$$\Xi(z) = \int_{-1}^1 d\mu I_0^+(z, \mu) I_0(z, \mu) \quad (27c)$$

The far more complex situation where the effect in question is the radiance in a specific direction (such as might be measured by a ground-based or space-based radiometer) will be discussed below, as will the case where the polarized form of the radiative transfer equation needs to be employed.

4. Beyond first order

Eq. (21b) shows clearly that, once we have decided to neglect the second term in Eq. (19), the formalism we have is linear — the first Taylor series term. Thus, its accuracy will be limited to perturbations which are small, in some sense (in parameter space). Before we present a series of applications of the perturbation technique, we will discuss three methods of extending the procedure to allow for more accurate calculations in those cases where we do not wish to be quite so restricted.

4.1. Higher order terms

As Eq. (21b) represents the first term in a Taylor series, it is natural to ask about the higher order terms. To derive them we must first introduce the Green's function, which is the general solution of the transport equation for a point source (Bell and Glasstone, 1970):

$$LG(\mathbf{r}_0, \Omega_0 \rightarrow \mathbf{r}, \Omega) = \delta(\mathbf{r} - \mathbf{r}_0) \delta(\Omega - \Omega_0) \quad (28)$$

with a similar definition for the adjoint Green's function. Thus, G can be said to represent the solution to *all* radiative transfer problems (for that model atmosphere). For a specific source, Q , the intensity distribution is then given by

$$I = \langle G, Q \rangle \quad (29)$$

and any desired effect may be obtained via

$$E = \langle R, GQ \rangle \quad (30)$$

Box et al. (1988b) showed that the Green's function, G , for an atmospheric transport operator L , is related to the Green's function, G_0 , for a second transport operator L_0 , via the equation

$$G = G_0 - G \Delta L G_0 \quad (31)$$

This is essentially a Dyson equation (Ziman, 1969), and may be solved by successive substitution or (possibly) by a matrix inversion. In the former case, we may use Eq. (30) to obtain the following 'series expansion' for the effect, E :

$$E = E_0 - \langle I_0^+, \Delta L I_0 \rangle + \langle I_0^+, \Delta L G_0 \Delta L I_0 \rangle - \langle I_0^+, \Delta L G_0 \Delta L G_0 \Delta L I_0 \rangle + \dots \quad (32)$$

Note that if the perturbation can be expressed in terms of a scale parameter, δ , as was done in Eq. (21b), then this equation is a power series (essentially a Taylor series) in that parameter.

Keevers (1986) showed how to express each term in this series as a combination of integrals and summations by first Fourier-expanding G_0 , and showed that successive terms could be obtained in an iterative manner. Croke (1988), Trautmann and Box (1993) and Box and Trautmann (1993) have attempted to compute the Green's function for an aerosol atmosphere, and evaluate the terms in this series, with mixed success. It is clearly necessary to obtain G_0 with sufficient accuracy, or else the errors compound with each higher term. We hope to return to this challenge in the near future, by using a potentially more accurate radiative transfer code to obtain G_0 .

4.2. Multiple base cases

If the intention is to be in a position to obtain a particular effect as one atmospheric model parameter is varied over a range which may be too large for the first order term to be sufficiently accurate, then two (or more) base cases may be used, at two (or more) different values of the relevant parameter (for example, aerosol optical thickness). At each of these values, we have both the value of E (from the radiative transfer calculation) and its derivative (from the perturbation integral). These may then be 'connected' using spline techniques for example (Trautmann and Box, 1995). More base cases may be required if perturbation in several 'directions' in parameter space is desired.

4.3. Alternate formulations

Gerstl and Stacey (1973) used the Schwinger variational principle to obtain an alternative formulation of perturbation theory. Their result

$$E \cong \frac{E_0}{1 + \langle I_0^+, \Delta L I_0 \rangle / E_0} \quad (33)$$

is identical to Eqs. (20a, 20b) and (20c) to first order, and hence for small perturbations. However, for larger perturbations, the Schwinger version makes specific pre-

dictions as to the form of the higher order terms in the perturbation series.

A third, essentially ad hoc formulation is based on the realization that the radiative transfer equation, being a first order differential equation, ought to have solutions with an essentially exponential behaviour. Hence Box et al. (1996) suggested the expression

$$E = E_0 \exp[-\langle I_0^+, \Delta LI_0 \rangle / E_0] \quad (34)$$

which again agrees with the linear formulation for small perturbations, but makes its own predictions as to the form of the higher order terms.

Douriaguine and Box (1999) have investigated the applicability of all three formulations in the case of large perturbations in cloud optical thickness. They found that in a pure scattering atmosphere, the Schwinger formulation provided results which were useful over a wide perturbation range, whereas in an atmosphere with even a small amount of absorption the exponential formulation was clearly superior. (In all cases, the linear formulation gave negative results for moderate perturbations, something the other two formulations cannot do.) Investigations are continuing to find the cross-over point between these two situations.

5. Applications

Over the years, my group, and others, have applied perturbation theory to a number of problems in atmospheric radiation. In this section we concentrate on three main application areas in order to illustrate the utility of this technique.

5.1. Aerosol atmospheres

Atmospheric aerosols are one of the most variable components of our atmosphere, and affect its radiative properties in several ways. It is now realized that aerosols, and especially anthropogenic aerosols, play a key role in the Earth's radiative balance (Houghton et al., 1995). Some simple questions can be asked, and answered quite nicely using perturbation theory. For example, what is the forcing (change in net flux at the top of the atmosphere) caused by the addition of a sulphate aerosol layer of variable optical thickness? The answer is already there in Eq. (21b). Perform a base case computation using a background (natural) aerosol, and treat the sulphate layer as the perturbation (Box and Trautmann, 1994). Note that the great power of perturbation theory is that it gives the forcing for a variable sulphate loading with a single computation.

There are some situations where a small change in atmospheric composition can lead to quite large radiative changes. One interesting example is the impact of a stratospheric aerosol layer on radiation with wavelengths

on the edge of the ozone u.v. absorption band (Michelangeli et al., 1989). If that layer is at or above the bulk of the ozone, and the solar zenith angle is large, then it may actually scatter sunlight more vertically downwards, providing the photons with a shorter path through the ozone layer (Davies, 1993), leading to significant increases in surface radiation. We addressed this problem using perturbation theory (Box, 1995), and found that, although the perturbation in atmospheric optical properties was small, the perturbation in surface flux could be very large. Despite this, perturbation theory gave reliable answers.

A challenge of a different kind is provided by the need to include aerosol radiative properties in both global climate models, and numerical weather prediction models (Werner, 2000). Here it is their wide variability which poses its own problems for modellers, such as how often is it necessary to re-solve the transport equation; and how many different aerosol types should be included. Perturbation theory provides at least one answer to this dilemma. Choose a 'standard' aerosol model as the base case, and perform an accurate calculation (possibly off line) of both the forward and adjoint intensity fields. Then select a set of aerosol models — or model components — and perform perturbation integrals for each. In fact, since one can conceive of any of the aerosol components being varied within any of the computational layers of the model atmosphere, a separate perturbation integral may be performed for each component for each layer (again off-line). Any actual aerosol situation which occurs in the model may then be treated as a perturbation on the base model, and the change in any given effect obtained as an obvious extension of Eqs. (20a, 20b) and (20c) (Trautmann et al., 1992; Box et al., 1996):

$$E = E_0 - \sum_{k=1}^K \sum_{j=1}^J \delta_{jk} \Delta E_{jk} \quad (35)$$

where δ_{jk} is the perturbation scale factor for component j in layer k , and ΔE_{jk} is the corresponding perturbation integral.

In contrast to this off-line approach, Gabriel et al. (1998, 2000) have used the two-stream method, which is fully analytic. They have shown that the adjoint solution is also analytic, although some care is needed in its manipulation, as growing exponentials appear, rather than decaying exponentials. Perturbation integrals may also be performed analytically.

5.2. Spectral integration

There are a number of situations where it is necessary to perform a careful integration over wavelength of a particular radiation quantity multiplied by some response quantity. Examples include photolysis rates, where the radiation quantity is the actinic flux and the response

quantity is the reaction cross section; and u.v. indices, where the radiation quantity is the net flux, and the response quantity is the relevant action spectrum. As the principles are essentially the same, we will concentrate on the latter.

The ‘dose rate’ may be expressed in the form

$$F_{\text{dose}} = \int d\lambda AS(\lambda)F^{\downarrow}(\lambda) = \int d\lambda AS(\lambda)F_0(\lambda)T(\lambda) \quad (36)$$

Here AS is the action spectrum for erythema (McKinley and Diffey, 1987), and F^{\downarrow} is the downward flux at the ground, which we have chosen to factorize into the extraterrestrial solar flux, F_0 , times the atmospheric transmission, T . As indicated, all quantities are wavelength dependent. The range of the integration will be over all wavelengths for which the integrand is not negligible: say 290–330 nm.

Of the three spectral quantities in Eq. (36), two of them — the action spectrum and the extraterrestrial flux — are known with sufficient precision. The transmission, however, is not, as it varies markedly with the total ozone column (and, to a lesser extent, with its vertical distribution). However, it is precisely this quantity that is amenable to the perturbation theory approach. Choose a base case that corresponds to the wavelength at the maximum of the integrand — say 310 nm — and perform a full radiative transfer computation for each desired solar zenith angle, as well as a single adjoint transfer computation (as there is only one effect: surface flux). To obtain the transmission at any other wavelength, we need to perturb both the ozone absorption optical thickness, and the Rayleigh scattering optical thickness. (We may assume that the aerosol optical properties are constant over such a small wavelength range, although these, too, could be perturbed.) Thus, we actually need to evaluate two perturbation integrals, one for ozone absorption and one for Rayleigh scattering. As the variation in ozone optical thickness over this spectral range is quite large (13.6 to almost zero), the exponential formulation of perturbation theory will be needed:

$$T(\lambda) = T_0 \exp\{-[\delta_{\text{ab}}\Delta T_{\text{ab}} + \delta_{\text{sc}}\Delta T_{\text{sc}}]/T_0\} \quad (37)$$

where δ_{ab} is the wavelength dependent scale factor for ozone absorption, δ_{sc} is the scale factor for Rayleigh scattering, and ΔT_{ab} and ΔT_{sc} are the corresponding perturbation integrals. As this approximation is ‘pushed’ further from the base case (that is, to longer or shorter wavelengths), its accuracy will decrease. However, the contribution to the total integral from such wavelengths is also small, so that the overall accuracy of the final result for the dose rate will remain high (Box et al., 1997). If necessary, a second base case may be required.

In fact, it is possible to include an additional perturbation, namely the total ozone column, simply by reinterpreting δ_{ab} . Box et al. (1997) compared the accuracy

of perturbation theory against a full radiative transfer calculation. They chose 2.5 nm wavelength steps between 290 and 330 nm (17 calculations), for 16 different ozone columns between zero and double the base case — a total of 272 calculations, compared to just one for perturbation theory. With the exception of very low sun angles, the results of the two methods were all but indistinguishable.

Loughlin and Box (1998) actually chose two base wavelengths, 310 and 360 nm, in order to accurately include the tail of the erythema spectrum. They then present a set of tables of the various quantities in Eq. (36) for a total of 10 solar zenith angles. With these, users can obtain T at any wavelength, for any ozone column (but for a set vertical profile), and thus perform the integral in Eq. (35).

If more accurate results are required, then perturbation of the actual ozone profile is also possible, and probably desirable. (Perturbation of the aerosol properties from the assumed base case might also be required.) In this case, users would need access in some form to the quantities defined in Eqs. (26, 27a, 27b) and (27c). Note from Eq. (26) that the only mathematical step which would remain is the vertical integral, so that a highly efficient, and flexible, code could be easily produced.

5.3. Remote sensing inversion

One method for obtaining geophysical data on aerosol optical properties is to make multiangular measurements of the radiation exiting the atmosphere at either ground level (using a Cimel radiometer — Holben et al., 1998) or from space (for example using the POLDER instrument (Herman et al., 1997) or the MISR instrument (Diner et al., 1998)). It is then necessary to invert such data sets, which effectively means inverting the radiative transfer equation. There are two standard approaches to such a problem — table look-up, and iteration — each with its own advantages and disadvantages.

Perturbation theory can be applied to this process, as it can significantly aid iterative inversions. However, as we are dealing with radiances and not fluxes we need the full intensity field as a function of azimuth angle — for example as a Fourier series — and thus far more work is required for each computation. Box and Sendra (1995) showed that perturbation theory may be used to predict with reasonable accuracy how the intensity in any direction will change if any of the parameters of the atmospheric optical model are changed. Formally, we may write this in the form (Box and Sendra, 1999)

$$\Delta \mathbf{E} = \mathbf{A} \Delta \boldsymbol{\eta} \quad (38)$$

This time we interpret \mathbf{E} as a vector of effects (the intensity in different directions), $\Delta \mathbf{E}$ as a vector of changes in these effects, $\boldsymbol{\eta}$ as a vector of parameters (for example optical thickness, single scattering albedo, and the Leg-

endre expansion coefficients), $\Delta\eta$ as a vector of perturbations in these parameters, and \mathbf{A} as a matrix of perturbation integrals which connect the parameters to the radiances (in effect they are a set of partial derivatives).

Can this result be turned into an inversion algorithm? Box and Sendra (1999) suggested the following procedure. Make an initial guess (base case) as to the composition and optical properties of the atmosphere. Solve the radiative transfer equation (and its adjoint) for this base case, and from this predict what the measured results should be. In general, these predictions will differ from the actual measurements. Next we use the radiance and adjoint radiance functions to compute all required elements of the \mathbf{A} matrix. (This is a very much simpler task than actually solving the radiative transfer equation.) Then we construct a difference vector, $\Delta\mathbf{E}$, of the differences between the actual and the predicted measurements, and invert Eq. (38) to obtain the difference between the true optical parameters, and the first guess parameters:

$$\Delta\eta = \mathbf{B}\Delta\mathbf{E} \quad (39)$$

Here \mathbf{B} is the generalized inverse of \mathbf{A} , which will need to include some form of regularization (Box and Sendra, 1999).

$$\mathbf{B} = (\mathbf{A}^T\mathbf{A} + \gamma\mathbf{I})^{-1}\mathbf{A}^T \quad (40)$$

Here \mathbf{I} is the identity matrix, γ is a smoothing parameter, and the superscript T denotes a matrix transpose. (Note that \mathbf{A} need not be a square matrix.) If this inversion process shows that the initial guess was too far from the truth, a second iteration may be required (Box and Sendra, 1999).

Box and Sendra (1999) showed that in the case of ground-based observations, where one may assume that the total optical thickness is also measured, it is possible to retrieve the single scattering albedo (or equivalently the scattering optical depth) and a number of Legendre expansion coefficients [the exact number will depend on the angular spread of the measurements, and the noise level (Sendra and Box, 1999)]. In the case of space-based observations, where the total optical thickness is usually not known, Sendra and Box (2000) showed that it should be possible to retrieve the scattering optical depth and the most important of the Legendre coefficients. Note that these are some of the key input parameters required to ascertain the radiative impact of an aerosol layer, and also that this technique makes no assumptions as to the sphericity of the particles — Mie theory is not assumed. (However, since we usually don't have direct information on the total optical thickness, we similarly lack information on absorption.)

Polonsky and Box (2001) suggested a different approach, which may be operationally more efficient. Assume that the aerosol in a certain region is composed of a number of well defined components, but in unknown

concentrations. Make a first guess as to the actual composition (perhaps from inversion of the previous pixel), and treat variations in this composition as perturbations. In this case, it is necessary to compute the perturbation integrals for each possible component (possibly in several distinct layers) as in Eq. (35). This again leads to an equation formally identical to Eq. (38), except that η should now be interpreted as a vector of aerosol components.

Satellite observation of thermal emission to determine profiles of temperature and water vapour is one of the oldest examples of atmospheric remote sensing. If one assumes that scattering is negligible at the wavelengths most often used, the radiative transfer equation simplifies considerably. In fact, both it and its adjoint may be obtained analytically, so that perturbation integrals may also be performed analytically. Marchuk (1964) proposed an inversion procedure which is similar in principle to that just described. He suggested using a standard, or climatological, temperature profile as the base case, and inverting for the difference between this and the true profile. Recently, Ustinov (2000, 2001) has revisited this idea, and extended it to the case where scattering cannot be neglected.

6. Current and future developments

There are a number of areas where we, and others, intend to apply perturbation theory to radiation problems which may be amenable to its powers. These are currently being addressed, or will be addressed as soon as time and other resources are available.

6.1. Polarization

Measurements of the degree and direction of polarized radiances exiting the atmosphere may contain much valuable information of aerosols and other atmospheric constituents, and are available from the ground-based Cimel radiometer (Holben et al., 1998) or the POLDER space-borne instrument (Herman et al., 1997). However, interpreting such data is quite a challenge as it essentially involves the inversion of the vector form of the radiative transfer equation (Coulson, 1988). Again we believe that perturbation theory has the potential to contribute to such tasks.

Perturbation theory has recently been extended to the vector form of radiative transfer by Tian (Tian, 2000; Tian and Box, 2002). In order to achieve this he needed to consistently define the vector transport operator and its adjoint, which includes the 4×4 Stokes matrix (transposed in the adjoint case). It was then necessary to define appropriate response functions in order to extract (the elements of) the effect vector. With these definitions he was able to construct the complete appar-

atus for the perturbation of any effect within the framework of polarized radiative transfer.

The first application we have considered is to polarized surface fluxes, which probably do not contain much additional information. However it was felt that this should be the first test of the formalism. Results will be submitted for publication shortly. Measurements of the degree and direction of polarization, as well as intensity, effectively tripple the amount of information available on aerosol scattering properties. However, the Stokes matrix contains 16 times as much content as the scalar phase function. Thus an inversion along the lines proposed by Box and Sendra (1999) is unlikely to succeed. By contrast, the new approach of Polonsky and Box (2001) is readily adaptable to the polarized case.

6.2. Non plane parallel media

One of the biggest current challenges in radiative transfer computation is its application to non plane parallel media, such as broken cloud fields. In such cases the radiation field becomes a function of five variables (three spatial and two directional), instead of three, and the transfer equation becomes equally complex. While the Monte Carlo technique is capable of handling any medium, it is very slow and inefficient. More recently, several new solution methods have been introduced, such as the Fourier–Riccati Method of Gabriel et al. (1993) and the spherical harmonics discrete ordinate method of Evans (1993). Depending on the actual information required of the solution, the perturbation technique has the potential to also address this situation.

Assume that the information which we seek is spatially averaged in some way: for example, the average surface flux underneath a broken cloud field, or the radiance measured by a satellite sensor from a cloud-contaminated pixel. The perturbation approach to such problems would be to start with a plane parallel base case (presumably with spatially averaged optical properties), and treat the horizontal inhomogeneities as the perturbation. In this case, the transport operator for the base case will be the standard plane parallel operator we have used so far, while the perturbed operator will involve all three spatial derivatives, and have optical properties which are functions of y and z . The difference operator will then be given by

$$\Delta L = \Omega \cdot \nabla - \mu \frac{\partial}{\partial z} + \Delta \sigma_s(\mathbf{r}) - \Delta \sigma_s(\mathbf{r}) \int d\Omega' p(z, \Omega' \rightarrow \Omega) \quad (41)$$

Note that when we evaluate the relevant inner product integrals, the phase space variables will now include y and z .

Consider now evaluating the first order perturbation

term, $\langle I_0^+, \Delta L I_0 \rangle$. Because we have defined the base case to be the spatial average of the perturbed case, this term integrates to zero. (As the base case radiance is independent of y and z , the derivative terms in ΔL do not come into play.) Thus we have no alternative but to turn to the second (and higher) order terms, which will require the computation of the full Green's function.

Turn now to the second order term: see Eq. (32). Keeters (1986) has shown that in the case of azimuthally independent effects such as surface flux, this term may be reduced to

$$\Delta E_2 = 2\pi \int d\mathbf{r} \int d\mu \int d\mathbf{r}' \int d\mu' G_0(\mathbf{r}, \mu \rightarrow \mathbf{r}', \mu') Z(\mathbf{r}, \mu) Z(\mathbf{r}', \mu')^+ \quad (42)$$

where

$$Z(\mathbf{r}, \mu) \equiv \Delta \sigma_t(\mathbf{r}) I_0(z, \mu) - 0.5 \sum (2n+1) \Delta \eta_n(\mathbf{r}) P_n(\mu) \xi_n(z) \quad (43)$$

Computation of the full, three dimensional Green's function would appear to be almost as great a challenge as the original problem we started with. However, there are some key simplifications, including the fact that we probably only need to perform such calculations a few times, for carefully selected base cases.

The case of satellite remote sensing is, in general, not azimuth independent, except for the important case of nadir viewing sensors. (In this case the effect is azimuth independent, although it is probably not the same as the flux — nevertheless, Eq. (42) is still valid.) For all other cases, and especially POLDER and MISR, we will need to extend this formalism to the full Fourier series, as indicated in Section 5.3. This will certainly be a challenge, although in view of the importance of the correct interpretation of such observations, this is a challenge which we hope to take up.

6.3. Ozone retrievals

The retrieval of vertical ozone distributions from backscattered ultraviolet light from the Earth's atmosphere involves the linearization of the radiative transfer equation. Operational retrieval algorithms should possess the following properties: (i) accurate modeling of radiance measurements and its linearization with respect to ozone profile characteristics, and (ii) fast numerical performance of the forward modeling as an essential part of the retrieval process.

Recently, Hasekamp and Landgraf (2001) successfully demonstrated the feasibility of ozone profile retrievals from GOME nadir radiance observations. Perturbation theory provides a sound basis for the computation of the functional derivatives of the radiance field with respect to ozone density as a function of altitude. An operational ozone retrieval algorithm has been

developed in combination with an efficient solution technique for the computation of the forward and adjoint radiance fields. At present the performance of this algorithm is validated with a representative set of ozone sonde measurements covering the full globe. The results of this investigation will be reported elsewhere.

Future space-borne spectrometers, such as SCIAMACHY (Scanning Imaging Absorption SpectroMeter for Atmospheric CHartrography, to be launched on ENVISAT-1 in 2001) and OSIRIS (Optical Spectrograph and InfraRed Imaging System, to be launched on ODIN in 2001) employ limb observations for determining the vertical ozone distribution in fine vertical resolution. For the interpretation of these measurements an extension of the plane-parallel perturbation theory to include sphericity effects of the Earth's atmosphere is necessary. A first step towards a linearized spherical radiative transfer code is the pseudo-spherical approximation (Trautmann et al., 2000). Work is in progress to extend perturbation theory to include such sphericity effects in both the direct beam as well as the single-scattered diffuse radiation.

A third development concerns the fact that, in principle, the atmosphere responds nonlinearly to changes in ozone. This means that for the ozone profile retrieval an iterative scheme has to be employed, which in each iteration step requires a new computation of the functional derivatives. Another promising approach to treat this nonlinearity is to compute the Green's function of the plane-parallel radiative transfer equation as described in Trautmann and Box (1993). This approach provides an efficient means to compute functional derivatives for arbitrary states of the atmosphere from the Green's function determined for a climatologically representative state of the atmosphere.

7. Software considerations

Because radiative perturbation theory is a technique which can be applied to a wide variety of atmospheric radiation problems, it is clearly not possible to provide potential users with a computer code for their work. We can, however, provide some insight into what changes may be required in users' own codes.

In order to perform the phase space integrals indicated in Eqs. (20a, 20b) and (20c), we need both the intensity, and the adjoint intensity, as functions of height and direction. As indicated previously, obtaining the adjoint intensity is really no more difficult than obtaining the normal intensity function. However, the adjoint is 'generated' from the adjoint source, which may relate to the flux at an arbitrary height for example. Thus, users will need to modify their codes to incorporate additional potential sources (Box et al., 1988a; Qin et al., 2002). The case of surface flux is perhaps the easiest to handle,

as some codes already allow this as a simple way to handle Lambertian surface reflection.

Once all appropriate adjoint sources have been included, it is necessary to extract the full details of the solution. Some codes such as DISORT (Stamnes and Conklin, 1984) usually only provide the user with selected output, although in most cases all the required information is stored internally. (One exception is the doubling/adding technique, which only contains information at the layer boundaries. While this may be interpolated with reasonable accuracy for thin layers, this is certainly not the case with optically thick layers, which are usually the object of this technique.) Our original investigations using perturbation theory were based on the Gauss-Seidel code (Herman and Browning, 1965; Dave and Gazdag, 1970), which is suitable for optically thin atmospheres (up to an optical thickness around 1.0) but not for clouds.

We have recently been able to extract all the necessary information from the DISORT code, in the form of exponential series (Douriaguine and Box, 1999; Qin et al., 2002), which has allowed us to begin an investigation of the use of perturbation theory in cloudy atmospheres. The work by Qin et al. (2002) is particularly valuable, as they have been able to perform calculations for a number of sources simultaneously, with only a moderate increase in computation time. Thus, both the forward and adjoint cases may be obtained in a single run. Polonsky (work in progress) has developed a spherical harmonics code which is also capable of providing the inputs for perturbation theory.

Once the radiance and adjoint radiance distributions are available, performing the perturbation integrals is usually straightforward. However, it needs to be appreciated that the direct solar beam needs to be incorporated into I_0 : it is usually regarded as being a delta function in direction, and so needs some care. One situation which proved especially challenging for us was the satellite remote sensing application (Section 5.3), where the adjoint intensity also contains a 'direct beam' term. The interaction between these two beams requires considerable care (Sendra and Box, 2000).

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