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# Retrieval of the phase function and scattering optical thickness of aerosols: a radiative perturbation theory application

Claudia Sendra, Michael A. Box\*

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

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## Abstract

In a previous paper we showed that radiative perturbation theory offers a suitable platform to solve the problem of retrieving key phase function details from measurements of the exiting radiances at the top or bottom of the atmosphere. In this paper we extend the formalism to also account for uncertainty in the total optical thickness while predicting the changes in the emerging radiation. We examine the sensitivity of the predictions and demonstrate that inverting the new extended direct problem allows us to accurately retrieve the scattering coefficient and the key phase function parameters. © 1999 Elsevier Science Ltd. All rights reserved.

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## 1. Introduction

In a previous paper [1] we used radiative perturbation theory [2] to develop a retrieval technique that let us determine the radiative properties of a scattering atmosphere. We showed that it is possible to establish a system of equations represented by a kernel matrix, such that its inversion leads to a stable solution. In a follow-up paper [3] we showed how one could examine the information content of different observation sets.

In those papers, we addressed a restricted problem, in that we assumed that the total optical thickness was a known quantity (a valid assumption in the case of ground-based observations), the surface albedo could be ignored (a not unreasonable assumption in many situations), and that the atmosphere was vertically homogeneous (a reasonable assumption in the near IR, where molecular scattering is negligible). In the present paper we extend the perturbation formalism to include the change in the exiting radiation caused by a perturbation

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\* Corresponding author. Tel.: 00-612-9385-4545; fax: 00-612-9385-6060.

E-mail address: [mab@newt.phys.unsw.edu.au](mailto:mab@newt.phys.unsw.edu.au) (M.A. Box)

(or uncertainty) in the extinction optical thickness, which may then be regarded as an additional unknown to be retrieved.

The analytical expression relating the variation of the exiting radiances to the perturbation in the aerosol characteristics is derived in Sections 2 and 3. The numerical inversion scheme is discussed in Section 4, while in Section 5 we present some numerical case studies to show the retrieval of the atmospheric parameters. These results demonstrate that our technique is capable of retrieving the scattering optical thickness (but not the total optical thickness, or single scattering albedo), and three coefficients of the Legendre expansion of the phase function. While this data does not comprise the full set of parameters needed to determine the radiative/climatic effect of the aerosol, it does represent a valuable potential contribution to our knowledge of aerosol optical properties.

## 2. Problem formulation via radiative perturbation theory

Radiative perturbation theory is a tool which may be used to predict how radiative quantities (or “effects”), such as fluxes and radiances, vary when some (or all) of the parameters which define the optical model of the atmosphere are perturbed. Like all forms of (first order) perturbation theory, its predictive accuracy generally decreases steadily as the magnitude of such perturbation increases. In order to understand the formalism, we start by writing the radiative transfer equation in operator notation [4] as

$$LI = Q,$$

where  $I$  is the radiance distribution,  $Q$  the source of radiation (the extraterrestrial solar beam in this case), and  $L$  the transport operator [4], which may be expressed as

$$L \equiv \mu \frac{\partial}{\partial z} + \sigma_t - \sigma_{sc}(z) \int_{4\pi} p(z, \Omega' \rightarrow \Omega) d\Omega'. \quad (1)$$

Here  $\sigma_t(z)$ ,  $\sigma_{sc}(z)$  and  $p(z, \Omega \rightarrow \Omega')$  are the extinction cross section, the scattering cross section, and the phase function, respectively.

If we alter (perturb) any of these parameters, we obtain a new optical model, with a new transport operator, and consequently a new radiance distribution. We now ask the question: what change does this induce in a specific radiative effect,  $E$ , such as a flux or (exiting) radiance? The answer (to first order) is [2]

$$\Delta E = - \langle I^+, \Delta L I \rangle, \quad (2)$$

where  $I^+$  and  $I$  are the adjoint [4] and normal radiance distributions calculated from the initial model,  $\Delta L$  is the difference transport operator, and angular brackets indicate integration over all “phase space” variables, in this case height and direction. (The adjoint radiance is obtained by solving the adjoint transport equation “backwards” from the effect as its source.)

The original applications of radiative perturbation theory were to azimuth-independent effects such as fluxes and heating rates [5–8], including a very successful application to UV indices [9], which involve an additional integration over wavelength. More recently, we have turned our attention to calculating exiting radiances, with the aim of producing an inversion procedure for

obtaining information about atmospheric optical properties from either ground- or spaced-based observations.

As a first step, we were able to show [10] the prediction capability for changes in the exiting radiances, caused by changes in the phase function, and single-scattering albedo. (Our working restrictions during that analysis were that the total optical thickness was to be held fixed, and ground reflection was ignored.) We start by expanding the phase function in terms of Legendre polynomials:

$$p(\theta) = \sum_{\ell=0}^L \chi_{\ell} P_{\ell}(\theta)/4\pi.$$

We may then define a set of parameters,  $\eta$ , by

$$\eta_{\ell} = \omega_0 \chi_{\ell} \quad \ell = 0, \dots, L,$$

where  $\omega_0$  is the single scattering albedo.

Thus we have a vector of optical parameters,  $\eta$ , which we may perturb, and a set of (one or more) exiting radiances, which can also be treated as a vector,  $E$ . Then we may formally write the connection between the changes in these radiances, and the perturbations in the optical parameters, as a matrix equation:

$$\Delta E = A \Delta \eta,$$

where  $A$  is a matrix of coefficients computed from the initial radiance and adjoint radiance distributions by an algorithm based on Eq. (2) (see below).

Having established this predictive (or forward) capability, we then turned our attention to the inverse problem (but with the same restrictions). While the reader should consult our two papers [1,3] for full details, the basic concept of using perturbation theory as an inverse tool is as follows:

1. Obtain a set of measurements of exiting radiances, either above or below the atmosphere.
2. Choose a ‘first guess’ optical model of the atmosphere. (In line with our restrictions, this model was required to include the correct optical thickness, but the single-scattering albedo, and phase function details, are arbitrary.)
3. Compute the ‘first guess’ exiting radiances. In general, they will differ from the actual measurements.
4. Use the ‘first guess’ radiance distribution, and the corresponding adjoint radiance distribution, to compute the coefficient matrix connecting any perturbations in the optical parameters with changes in the exiting radiances as indicated by the previous matrix equation.
5. Invert this set of equations to obtain corrections in the optical parameters in terms of the differences between measured and computed radiances:

$$\Delta \eta = B \Delta E,$$

where  $B$  is the inverse (or generalized inverse) of the matrix  $A$ . (Full details of this procedure are given in Section 4.) When these changes are added to the original optical parameters, the result should be an optical model which is closer to the true optical state of the atmosphere. If necessary, the process may be iterated until a satisfactory convergence is achieved. (Convergence can best be

judged by the agreement between the radiances calculated using the progressively improving optical model, and the measurements.)

### 3. Extension to variable optical thickness

In the present paper we release the more restrictive of our previous assumptions, namely that the total optical thickness was known (and hence fixed). To make the calculations easier to understand we divide the perturbation of the transport operator, described in Eq. (1), into two terms

$$\Delta L = \Delta L_1 - \Delta L_2 \tag{3}$$

with

$$L_1 = \sigma_t(z) \tag{4}$$

(this term accounts for the extinction process), and

$$L_2 = \sigma_{sc}(z) \int d\Omega' p(z, \Omega' \rightarrow \Omega). \tag{5}$$

(this term computes the scattering process).

Using this notation, the perturbation of the intensity described in Eq. (2) may be expressed as

$$\Delta E_1 = \int dz \int_{-1}^1 d\mu \int_0^{2\pi} d\phi I_o^+(z, \mu, \phi) \Delta\sigma_t I_o(z, \mu, \phi) \tag{6}$$

and

$$\begin{aligned} \Delta E_2 = & - (4\pi)^{-1} \sum_{i=0}^{\infty} \Delta(\sigma_{sc} \chi_i) \int dz \int_{-1}^1 d\mu \int_0^{2\pi} d\phi I_o^+(z, \mu, \phi) \\ & \times \left\{ \sum_{m=0}^{\ell} (2 - \delta_{0m}) \frac{(l-m)!}{(l+m)!} \int_{-1}^1 d\mu' \int_0^{2\pi} d\phi' P_l^m(\mu) P_l^m(\mu') \cos m(\phi - \phi') I_o(z, \mu', \phi') \right\}. \tag{7} \end{aligned}$$

In order to carry out the various integrations required to compute these terms, we expand both  $I_o$  and  $I_o^+$  in Fourier Legendre series as follows:

$$I_o(z, \mu', \phi'; \mu_s) = F_o \sum_l \sum_i B_{li}(z) P_i(\mu') \cos l\phi' + F_o \pi f(z) \delta(\mu' - \mu_s) \delta(\phi'), \tag{8}$$

$$I_o^+(z, \mu, \phi; \mu_o) = \sum_k \sum_j A_{kj}(z) P_j(\mu) \cos k(\phi - \phi_o) + \pi f^+(z) \delta(\mu - \mu_o) \delta(\phi - \phi_o), \tag{9}$$

where  $\mu_s, \mu_o$  are the solar and observational zenith cosines, respectively,  $F_o$  is the solar flux,  $\phi_o$  is the difference between the solar and observation azimuth angles, while  $f(z)$  and  $f^+(z)$  are defined as follows:

$$f(z) = \exp[-\tau(z)/\mu_s], \tag{10}$$

$$f^+(z) = \exp[-\tau(z)/\mu_o], \tag{11}$$

if we are concerned with radiances exiting at the top of the atmosphere. However, the definition of  $f^+(z)$  changes for radiances at the bottom of the atmosphere:

$$f^+(z) = \exp\{-[\tau_{\text{top}} - \tau(z)]/\mu_0\}. \tag{12}$$

In the above expression,  $\tau$  and  $\tau_{\text{top}}$  are the optical depth to position  $z$ , and to the top of the atmosphere, respectively. Inserting these expansions into Eqs. (6) and (7) and after some manipulations (which are outlined in the appendix) we obtain

$$\Delta E = F_0 \pi \left\{ \sum_m \cos m\phi_0 [Z_m^v + Z_m] + Z^{v22} \right\}, \tag{13}$$

where

$$Z_m^v = Z_m^{v11} + Z_m^{v12} + Z_m^{v21}, \tag{14}$$

$$Z_m = Z_m^{11} + Z_m^{12} + Z_m^{21} + Z_m^{22}. \tag{15}$$

Here

$$Z_m^{v11} = \frac{1}{2} (1 + \delta_{0m}) \int dz \Delta\sigma_t(z) \sum_j A_{mj}(z) B_{mj}(z) \frac{1}{2j + 1}, \tag{16}$$

$$Z_m^{11} = \frac{1}{2} (1 + \delta_{0m}) \sum_l \int dz \Delta\eta_l(z) \sum_j A_{mj}(z) I_{jl}^m \sum_i B_{mi}(z) I_{il}^m, \tag{17}$$

$$Z_m^{v12} = \int dz \Delta\sigma_t(z) f^+(z) \sum_i B_{mi}(z) P_i(\mu_0), \tag{18}$$

$$Z_m^{12} = \frac{1}{2} \sum_l \int dz \Delta\eta_l(z) f^+(z) \sum_i B_{mi}(z) I_{il}^m P_l^m(\mu_0) \left\{ \frac{(l - m)!}{(l + m)!} \right\}^{1/2}, \tag{19}$$

$$Z_m^{v21} = \int dz \Delta\sigma_t(z) f(z) \sum_j A_{mj}(z) P_j(\mu_s), \tag{20}$$

$$Z_m^{21} = \frac{1}{2} \sum_l \int dz \Delta\eta_l(z) f(z) \sum_j A_{mj}(z) I_{jl}^m P_l^m(\mu_s) \left\{ \frac{(l - m)!}{(l + m)!} \right\}^{1/2}, \tag{21}$$

$$Z_m^{22} = \frac{1}{4} (2 - \delta_{0m}) \sum_l \int dz \Delta\eta_l(z) f^+(z) f(z) P_l^m(\mu_s) P_l^m(\mu_0) \frac{(l - m)!}{(l + m)!} \tag{22}$$

and

$$Z^{v22} = \int dz \Delta\sigma_t(z) f^+(z) f(z) \tag{23}$$

with

$$I_{jl}^m = \left\{ \frac{(l - m)!}{(l + m)!} \right\}^{1/2} \int_{-1}^1 d\mu P_j(\mu) P_l^m(\mu). \tag{24}$$

As we did in our previous work [1], we will make the simplifying assumption that  $\Delta(\sigma_{sc}\chi_l)$  and  $\Delta\sigma_t$  are independent of  $z$ , so that we may re-write  $\Delta E$  as

$$\Delta E = F_o\pi \sum_{m=0} \cos m\phi_o \left\{ \sum_{l=m} \Delta(\sigma_{sc}\chi_l)Z_{l_m} \right\} + F_o\pi \left\{ \sum_{m=0} \cos m\phi_o \Delta\sigma_t Z_m^v + \Delta\sigma_t Z^{v22} \right\}. \tag{25}$$

Inverting the order of the summation we obtain

$$\Delta E = \sum_{l=0} \Delta(\sigma_{sc}\chi_l) \left\{ \sum_m F_o\pi \cos m\phi_o Z_{l_m} \right\} + \Delta\sigma_t \left\{ \sum_m F_o\pi \cos m\phi_o Z_m^v + F_o\pi Z^{v22} \right\} \tag{26}$$

$$= \sum_l \Delta(\sigma_{sc}\chi_l)A_l + \Delta\sigma_t A_{-1} \tag{27}$$

$$= \sum_l \Delta(\eta_l)A_l + \Delta\eta_{-1}A_{-1}. \tag{28}$$

We may express this equation in compact form as

$$\Delta E = \sum_{l=-1}^{\infty} \Delta(\eta_l)A_l, \tag{29}$$

where  $A_l$  is solar and observational angular dependent, and the variables that describe the model,  $\eta_l$ , are defined as

$$\eta_l = \begin{cases} \sigma_t & \text{if } l = -1, \\ \sigma_{sc}\chi_l & \text{if } l \geq 0 \end{cases} \tag{30}$$

and

$$A_l = \sum_m \pi F_o Z_{lm} \cos m\phi_o, \tag{31}$$

$$A_{-1} = \sum_m \pi F_o Z_{lm}^v \cos m\phi_o + \pi F_o Z^{v22}. \tag{32}$$

#### 4. Retrieval of the optical parameters

The description of our direct problem is given by the linear relation shown by Eq. (29). It establishes the relationship between perturbation or changes in the atmospheric optical model and the resulting perturbation in the exiting radiances. To explicitly show this connection, and note the angular dependency, we may rewrite Eq. (29) as

$$\begin{aligned} \Delta E(\Omega_{\text{obs}}, \Omega_{\text{solar}}) &\equiv E^{\text{pc}}(\Omega_{\text{obs}}, \Omega_{\text{solar}}) - E^{\text{bc}}(\Omega_{\text{obs}}, \Omega_{\text{solar}}) \\ &= \sum_{l \geq -1} A_l(\theta_{\text{obs}}, \theta_{\text{solar}}, \varphi_{\text{obs}}) \Delta\eta_l, \end{aligned} \tag{33}$$

where  $\Delta\eta_l = \eta_l^{\text{pc}} - \eta_l^{\text{bc}}$  is the perturbation in the  $l$ th coefficient  $\eta_l$ , and the superscripts pc and bc refer to perturbed and base case models respectively.  $\Omega_{\text{obs}} = (\theta_{\text{obs}}, \varphi_{\text{obs}})$  and  $\Omega_{\text{solar}} = (\theta_{\text{solar}}, \varphi_{\text{solar}})$

are the angular positions of sensor and sun, respectively, while  $\varphi_o$  denotes the difference between solar and observational azimuth angles.  $E^{pc}$  and  $E^{bc}$  are the exiting intensities corresponding to the perturbed and base model, respectively. These intensities may be obtained at the top or bottom of the atmosphere. We are interested in inverting this relation with respect to  $\Delta\eta_l$ , and thus obtaining an expression for these coefficients as a function of the measured intensity. We will give the measured intensity the role of “perturbed intensity”. This intensity will be associated with the unknown profile, which we are interested to retrieve, while the base case intensity will be computed from an assumed “first guess” profile.

In a real application we have a set of measurements where the observational coordinates change from one datum to another. However for all of them the sun position is almost the same. Therefore, we may assume that the sun position is fixed while the measurements are performed at the top or bottom of the atmosphere. The coefficients  $A_l$ , which are independent of any atmospheric perturbation, are computed from the base model, and are a function of the observational,  $\Omega_{obs}$ , and solar coordinates,  $\Omega_{solar}$ . (They will also vary depending in whether we are interested in intensities at the top or bottom of the atmosphere.) Computing Eqs. (31) and (32) for  $K$  different observational directions, we obtain a system of  $K$  linear equations, each equation with  $L$  terms where  $L - 2$  is the maximum order used in the Legendre expansion of the phase function. We may now express Eq. (29) in matrix form as

$$\Delta E = A\Delta\eta, \tag{34}$$

where  $A$  is the kernel matrix of dimension  $(K \times L)$  with elements  $A_{lk} \equiv A_l(\Omega_k)$ , relating the change of the observed intensity in direction  $\Omega_k$ , and the perturbation suffered by the  $l$ th coefficient,  $\eta_l$ .  $\Delta E$  is now a vector of dimension  $K$ , where each component represents the difference between the observed data,  $E$ , and the computed value,  $E^{bc}$ , (in this specific direction) from our initial guess.  $\Delta\eta$  is a vector of dimension  $L$ , being the difference between the parameter corresponding to the new (unknown) model and the initial guess model. Inverting Eq. (34) we obtain (in the case  $K = L$ )

$$\Delta\eta = A^{-1}\Delta E \tag{35}$$

and then

$$\eta = A^{-1}\Delta E + \eta^{bc}. \tag{36}$$

This equation offers us the possibility of acquiring information about the unknown profile  $\eta$  from a base model  $\eta^{bc}$  knowing the difference of the exiting intensities between both models. We will call  $\eta^{retr}$  the “retrieved coefficients” obtained using Eq. (36). The first retrieved coefficient accounts for the perturbation of the extinction cross section:

$$\Delta\eta_{-1}^{retr} = \sigma_t^{retr} = -\sigma_t^{bc}. \tag{37}$$

The second retrieved coefficient corresponds to the scattering cross section:

$$\Delta\eta_0^{retr} = \Delta\sigma_s^{retr} = \sigma_s^{retr} - \sigma_s^{bc}. \tag{38}$$

The third and successive retrieved coefficients are related to the phase function expansion coefficients:

$$\Delta\eta_l^{retr} = \sigma_s^{retr} \chi_l^{retr} - \sigma_s^{bc} \chi_l^{bc}. \tag{39}$$

Replacing the scattering cross section retrieved in Eq. (38) into Eq. (39) we obtain an expression for the coefficients  $\chi_i^{\text{retr}}$  of the phase function:

$$\chi_i^{\text{retr}} = (\Delta\eta_i^{\text{retr}} + \eta_i^{\text{bc}}) / \sigma_s^{\text{retr}}. \quad (40)$$

As discussed in some detail in our previous paper [1], we know that the fidelity of the inversions depends critically on the  $L$  dimension of the matrix, in order to avoid magnification of the measurement errors. The second dimension,  $K$ , which is associated with the number of measurements, should also be determined based on our knowledge about the number of independent pieces of information that are present [3]. The same selection criteria have been used in this work to solve the matrix inversion problem using the generalized inverse matrix,  $B$  defined by

$$B = [A^T A + \gamma I_{\text{id}}]^{-1} A^T, \quad (41)$$

where  $I_{\text{id}}$  is the identity matrix of dimension  $L \times L$  and  $A^T$  denotes the transpose of matrix  $A$ . Selection of both  $\gamma$  and the  $L$ -dimension of  $A$  have been selected as dictated by the associated singular values of  $B$  and  $A$ , respectively [2,3].

## 5. Application to satellite observations

In order to test the capability of our inversion procedure, we have applied it to nine case studies (synthetic data sets). We decided to use the same base case (first guess) for all nine case studies, as this meant that we would only need to compute the  $A$  coefficient matrix once. This is in line with what would be the most likely operational procedure, for the same reason. (Alternatively, one might compute a small number of base cases, and then choose the one, which most closely matched the measurements.)

The base case selected for these case studies corresponded to a total optical thickness of 0.2, a single-scattering albedo of 0.9, and the Henyey–Greenstein phase function, with an asymmetry factor of 0.75. The solar zenith angle was set at  $59.5^\circ$ , and a total of 32 measurements in the solar plane was assumed. (Note that we are continuing to assume that the atmosphere is vertically homogeneous, which is a reasonable approximation for the near IR wavelengths employed by a number of satellite sensors. We plan to remove this restriction as the next phase of this on-going project.) An examination of the singular values of the  $A$  matrix suggested that somewhere between 4 and 8 coefficients could be retrieved. We have therefore chosen to present results for  $L$  values of 4 and 7.

Our results are presented in Tables 1–9, corresponding to our 9 case studies. In each of these, we have used a different combination of total optical thickness and single-scattering albedo. (The total optical thickness, for example, ranged from 0.134 to 0.260, representing a perturbation of 30%.) However, we have used the same phase function as in our previous studies, namely the Lenoble Haze L model [11].

When we examine the results in Tables 1–9, we see that the retrieval of the first Legendre coefficient (essentially the asymmetry factor) is, in most cases, extremely good, with the second and third coefficients also reasonably accurate. (These three coefficients are sufficient to define a two-term Henyey–Greenstein phase function, for example, which is more than required for any radiative forcing calculation.) In the majority of cases, we were also able to retrieve the scattering

Table 1  
Retrieved coefficients for case 1

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_t$	0.160	0.200	0.242	0.251
$\tau_{sc}$	0.144	0.180	0.148	0.143
1	2.412	2.250	2.399	2.430
2	3.230	2.812	3.299	3.313
3	3.372	2.953	—	3.545
4	3.230	2.847	—	3.491
5	2.892	2.610	—	3.256

Table 2  
Retrieved coefficients for case 2

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_t$	0.134	0.200	0.242	0.127
$\tau_{sc}$	0.114	0.180	0.136	0.120
1	2.412	2.250	2.374	2.458
2	3.230	2.812	3.377	3.428
3	3.372	2.953	—	3.800
4	3.230	2.847	—	3.880
5	2.892	2.610	—	3.727

Table 3  
Retrieved coefficients for case 3

Order	True	1st guess	Retrieval for $L = 4$	Retrieval for $L = 7$
$\tau_t$	0.180	0.200	0.217	0.222
$\tau_{sc}$	0.144	0.180	0.150	0.147
1	2.412	2.250	2.396	2.421
2	3.230	2.812	3.271	3.283
3	3.372	2.953	—	3.499
4	3.230	2.847	—	3.433
5	2.892	2.610	—	3.179

optical thickness with more than acceptable accuracy. (For reasons we have been unable to explain, in the three cases in which the “true” optical thickness was greater than the first guess, the results for this parameter were less satisfactory.) We also note that, as expected, cases where the difference between true and first guess values were comparatively small gave the most accurate retrievals. This clearly implies that a second iteration in cases where the differences were larger would have enabled us to obtain even better results.

By contrast, our retrievals of total optical thickness, and hence single-scattering albedo, are less than satisfactory. This clearly reflects the fact that the photons we are measuring are

Table 4  
Retrieved coefficients for case 4

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_t$	0.180	0.200	0.232	0.229
$\tau_{sc}$	0.162	0.180	0.156	0.158
1	2.412	2.250	2.408	2.411
2	3.230	2.812	3.240	3.242
3	3.372	2.953	—	3.399
4	3.230	2.847	—	3.289
5	2.892	2.610	—	2.997

Table 5  
Retrieved coefficients for case 5

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_l$	0.180	0.200	0.201	0.217
$\tau_{ac}$	0.126	0.180	0.145	0.142
1	2.412	2.250	2.383	2.219
2	3.230	2.812	3.298	3.344
3	3.372	2.953	—	3.279
4	3.230	2.847	—	3.566
5	2.892	2.610	—	3.135

Table 6  
Retrieved coefficients for case 6

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_l$	0.260	0.200	0.168	0.169
$\tau_{ac}$	0.162	0.180	0.158	0.158
1	2.412	2.250	2.379	2.386
2	3.230	2.812	3.180	3.183
3	3.372	2.953	—	3.349
4	3.230	2.847	—	3.247
5	2.892	2.610	—	2.977

overwhelmingly singly scattered photons. Hence, we are quite sensitive to the scattering optical thickness, and the phase function structure, but not the absorption part of the optical thickness.

For Cases 6 and 8, we repeated the above procedure, but with the assumption that the solar zenith angle was approximately  $4^\circ$ . In this case, measurements in either half of the solar plane will give essentially the same information, so that we expect to retrieve slightly fewer coefficients. In both cases, however, the results were less than satisfactory. We believe that one additional factor which must be considered is the reduced range of scattering angles observed in this situation [3].

Table 7  
Retrieved coefficients for case 7

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_l$	0.260	0.200	0.179	0.165
$\tau_{ac}$	0.192	0.180	0.169	0.176
1	2.412	2.250	2.390	2.366
2	3.230	2.812	3.115	3.110
3	3.372	2.953	—	3.192
4	3.230	2.846	—	3.028
5	2.892	2.610	—	2.709

Table 8  
Retrieved coefficients for case 8

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_l$	0.220	0.200	0.208	0.188
$\tau_{ac}$	0.192	0.180	0.170	0.181
1	2.412	2.250	2.409	2.378
2	3.230	2.812	3.136	3.131
3	3.372	2.953	—	3.194
4	3.230	2.847	—	3.015
5	2.892	2.610	—	2.669

Table 9  
Retrieved coefficients for case 9

Order	True	1st guess	Retrievals for $L = 4$	Retrievals for $L = 7$
$\tau_l$	0.240	0.200	0.192	0.175
$\tau_{ac}$	0.192	0.180	0.170	0.179
1	2.412	2.250	2.399	2.372
2	3.230	2.812	3.124	3.119
3	3.372	2.953	—	3.191
4	3.230	2.847	—	3.019
5	2.892	2.610	—	2.684

If measurements are obtained with a ground-based instrument, the (total) optical thickness would be included in the data set, so that the more restricted inversion task discussed in our first paper [1] would be invoked. Nevertheless, we have employed the extended inversion presented in this paper to all 9 data sets, for measurements at the bottom of the atmosphere. In almost all cases, the results were generally inferior to the top of the atmosphere measurement results. This is hardly a drawback for the radiative perturbation procedure, as the results presented in our original paper show that, if we know the optical thickness, then satisfactory retrievals can usually be obtained.

## 6. Discussion

Most methods of obtaining information about the optical properties of atmospheric aerosols from satellite observations use a combination of observations at a series of wavelengths to select from a given handful of precomputed aerosol models. By contrast, the technique presented in this paper is radically different. Firstly, it works on a set of multidirectional observations at a single wavelength. Consequently, it provides the aerosol properties at each such wavelength for which a suitable set of measurements is available. Such multiangular data sets are, or soon will be, available from spaced-borne instruments such as POLDER and MISR.

We see from the results of the previous section, that provided that the first guess is not too far from the true values, our radiative perturbation technique is capable of retrieving the scattering optical thickness and the first few parameters in the Legendre expansion of the aerosol phase function. We should point out that at no time do we make any assumptions about the aerosols being spherical — that is, Mie theory is not used at any stage in the procedure.

It is unlikely that our technique would supplant techniques currently in practice. For a start, it is unable to return the total optical thickness, and/or the single-scattering albedo. Nevertheless, it is clearly capable of providing information unavailable from any other technique. This is especially significant when it is realized that no model assumptions have been included at any stage in the procedure.

Finally, we should point out that we are still working within the restriction of a zero ground reflectivity, so that the current formulation is restricted to retrievals over ocean. It would be a relatively straightforward task to add a Lambertian surface of unknown albedo, and add it to the list of parameters to be retrieved (presumably at the expense of one of the phase function parameters). However, the assumption of a Lambertian surface is difficult to justify over many terrestrial surface types. In fact, it appears that multiangular satellite observations are primarily sensitive to the surface BRDF, with the aerosol phase function being of only secondary importance. It is clearly impossible to retrieve both from the same set of observations.

## Appendix. Calculation of the perturbation of emerging radiances

In this appendix we outline the steps involved in the derivation of Eq. (13). As a first step, we consider the equations that describe the perturbation of the total optical thickness. The perturbation operator  $\Delta L_1$  may be written in the form:

$$\Delta L_1 = \int_{-1}^1 d\mu' \int_0^{2\pi} d\phi' \delta(\mu - \mu') \delta(\phi - \phi') \Delta \sigma_t. \quad (\text{A.1})$$

The following function names will be used in this appendix.  $I_d$  and  $I_s$  represent the diffuse and direct components of the forward intensity while  $I_d^+$  and  $I_s^+$  are the diffuse and direct adjoint intensities. The Legendre–Fourier expansions of these functions are:

$$I_d(z, \mu', \phi'; \mu_{\text{sol}}) = F_o \sum_l \sum_i B_{li}(z) P_i(\mu') \cos l\phi', \quad (\text{A.2})$$

$$I_s(z, \mu', \varphi'; \mu_{\text{sol}}) = F_o \pi f(z) \delta(\mu' - \mu_{\text{sol}}) \delta(\varphi'), \quad (\text{A.3})$$

$$I_d^+(z, \mu, \varphi; \mu_{\text{obs}}, \varphi_{\text{obs}}) = \sum_k \sum_j A_{kj}(z) P_j(\mu) \cos k(\varphi - \varphi_{\text{obs}}), \quad (\text{A.4})$$

$$I_s^+(z, \mu, \varphi; \mu_{\text{obs}}, \varphi_{\text{obs}}) = \pi f^+(z) \delta(\mu - \mu_{\text{obs}}) \delta(\varphi - \varphi_{\text{obs}}). \quad (\text{A.5})$$

Then the diffuse–diffuse term (or term 11) can be written

$$\Delta E_1^{11} = \langle I_d^+, \Delta L_1 I_d \rangle \quad (\text{A.6})$$

$$= \left\langle I_d^+, F_o \int_{-1}^1 d\mu' \int_0^{2\pi} d\varphi' \delta(\mu - \mu') \delta(\varphi - \varphi') \Delta \sigma_t(z) \sum_l \sum_i B_{li}(z) P_i(\mu') \cos l\varphi' \right\rangle \quad (\text{A.7})$$

$$= F_o \int_0^{z_t} dz \int_{-1}^1 d\mu \int_0^{2\pi} d\varphi \sum_k \sum_j A_{kj}(z) P_j(\mu) \cos k(\varphi - \varphi_{\text{obs}}) \Delta \sigma_t(z) \times \sum_l \sum_i B_{li}(z) P_i(\mu) \cos l\varphi \, d\varphi. \quad (\text{A.8})$$

If we make use of the integral expression

$$\int_0^{2\pi} \cos k(\varphi - \varphi_{\text{obs}}) \cos l\varphi \, d\varphi = \pi(1 + \delta_{ok}) \delta_{kl} \cos k\varphi_{\text{obs}} \quad (\text{A.9})$$

and the orthogonality property between Legendre polynomials:

$$\int_{-1}^1 P_i(\mu) P_j(\mu) \, d\mu = \frac{2}{2j+1} \delta_{ij}. \quad (\text{A.10})$$

Eq. (A.8) can be rearranged as

$$\Delta E_1^{11} = \pi F_o \sum_k \cos k\varphi_{\text{obs}} \frac{1}{2} (1 + \delta_{ok}) \int_0^{z_t} dz \Delta \sigma_t(z) \sum_j A_{kj}(z) B_{kj}(z) \frac{1}{2j+1}. \quad (\text{A.11})$$

Expanding now the diffuse–direct term (or term 21) we obtain

$$\Delta E_1^{21} = \langle I_d^+, \Delta L_1 I_s \rangle \quad (\text{A.12})$$

$$= \left\langle I_d^+, \pi F_o \int_{-1}^1 d\mu' \int_0^{2\pi} d\varphi' \delta(\mu - \mu') \delta(\varphi - \varphi') \Delta \sigma_t f(z) \delta(\mu_{\text{obs}} - \mu') \delta(\varphi') \right\rangle \quad (\text{A.13})$$

$$= \langle I_d^+, \pi F_o \Delta \sigma_t(z) f(z) \delta(\mu_{\text{obs}} - \mu) \delta(\varphi) \rangle \quad (\text{A.14})$$

$$= \pi F_o \int_0^{z_t} \sum_k \sum_j A_{kj}(z) \Delta \sigma_t(z) f(z) \int_{-1}^1 d\mu P_j(\mu) \delta(\mu_{\text{obs}} - \mu) \int_0^{2\pi} d\varphi \cos k(\varphi - \varphi_{\text{obs}}) \quad (\text{A.15})$$

$$= \pi F_o \sum_k \sum_j P_j(\mu_{\text{obs}}) \cos k\varphi_{\text{obs}} \int_0^{z_t} A_{kj}(z) \Delta \sigma_t(z) f(z). \quad (\text{A.16})$$

The derivation of the direct–diffuse (or 12) term is, of course, quite similar to the diffuse–direct (or 21) term, so we will not repeat the derivation for that term.

Turning finally to the direct–direct (or 22) term:

$$\Delta E_1^{22} = \langle I_s^+, \Delta L_1 I_s \rangle \quad (\text{A.17})$$

$$= \pi^2 F_o \int_0^z dz f(z) f^+(z) \Delta \sigma_t(z) \int_{-1}^1 d\mu \delta(\mu - \mu_{\text{obs}}) \quad (\text{A.18})$$

$$= \pi^2 F_o \int_0^{z_i} dz f(z) f^+(z) \Delta \sigma_t(z) \int_{-1}^1 \delta(\mu - \mu_{\text{obs}}) \delta(\mu - \mu_{\text{sol}}) d\mu \\ \times \int_0^{2\pi} \delta(\varphi) \delta(\varphi - \varphi_{\text{obs}}) d\varphi \quad (\text{A.19})$$

$$= \pi^2 F_o \delta(\mu_{\text{sol}} - \mu_{\text{obs}}) \delta(\varphi_{\text{obs}}) \int_0^{z_i} dz f(z) f^+(z) \Delta \sigma_t(z). \quad (\text{A.20})$$

Next, we outline the steps involved in the derivation of the expressions that describe the perturbation due to the scattering phase function and albedo. The diffuse–diffuse (or 11) term may be written in the form:

$$\Delta E_2^{11} = \langle I_d^+, \Delta L_2 I_d \rangle \quad (\text{A.21})$$

$$= \left\langle I_d^+, F_o (4\pi)^{-1} \sum_n \int_0^{z_i} dz \Delta(\sigma_s \chi_n) \left( \sum_m (2 - \delta_{0m}) \frac{(n-m)!}{(n+m)!} \int_{-1}^1 d\mu' \right. \right. \\ \left. \left. \times \int_0^{2\pi} d\varphi' P_n^m(\mu) P_n^m(\mu') \cos m(\varphi - \varphi') \sum_l \sum_i B_{li}(z) P_i(\mu') \cos l\varphi' \right) \right\rangle \quad (\text{A.22})$$

$$= F_o (4\pi)^{-1} \sum_n \int_0^{z_i} \Delta(\sigma_s \chi_n) \sum_l \sum_i \sum_k \sum_j A_{kj}(z) B_{li}(z) \sum_m (2 - \delta_{0m}) \frac{(n-m)!}{(n+m)!} \\ \times \int_{-1}^1 d\mu P_j(\mu) P_n^m(\mu) \int_{-1}^1 d\mu' P_i(\mu') P_n^m(\mu') \\ \times \int_0^{2\pi} d\varphi \int_0^{2\pi} d\varphi' \cos m(\varphi - \varphi') \cos l\varphi' \cos k(\varphi - \varphi_{\text{obs}}) \quad (\text{A.23})$$

$$= F_o (4\pi)^{-1} \sum_n \int_0^{z_i} \Delta(\sigma_s \chi_n) \sum_l \sum_i \sum_k \sum_j A_{kj}(z) B_{li}(z) \\ \times \sum_m (2 - \delta_{0m}) \cos \varphi_{\text{obs}} [\pi(1 + \delta_{0m})]^2 \delta_{ml} \delta_{mk} I_{jn}^m I_{in}^m, \quad (\text{A.24})$$

where

$$I_{jn}^m \equiv \left\{ \frac{(n-m)!}{(n+m)!} \right\}^{1/2} \int_{-1}^1 d\mu P_j(\mu) P_n^m(\mu). \quad (\text{A.25})$$

The evaluation of this integral will be discussed later on. Therefore,

$$\Delta E_2^{11} = F_o \frac{\pi}{2} \sum_2 \cos m \varphi_{\text{obs}} (1 + \delta_{0m}) \sum_n \int dz \Delta(\sigma_s \chi_1) \sum_j A_{mj}(z) I_{jn}^m \sum_i B_{mi}(z) I_{in}^m. \quad (\text{A.26})$$

We now address the limits of these summations. In Eqs. (A.2) and (A.4), the  $l$  and  $k$  summations are essentially open ended, whereas those over  $i$  and  $j$  are constrained by the quadrature order  $L$ . In Eq. (A.22), the summation over  $n$  is again open ended, while that over  $m$  runs from 0 to  $n$ . We have now reversed the order of these two summations, with the result that  $m$  now runs from 0 to infinity, and  $n$  from  $m$  to infinity.

Now we turn to the diffuse–direct (12) term:

$$\Delta E_2^{12} = \langle I_d^+, \Delta L_2 I_s \rangle \quad (\text{A.27})$$

$$\begin{aligned} &= \left\langle I_d^+, F_o (4\pi)^{-1} \sum_n \int_0^{z_i} dz \Delta(\sigma_s \chi_n) \sum_m (2 - \delta_{0m}) \frac{(n-m)!}{(n+m)!} \int_{-1}^1 d\mu' \right. \\ &\quad \left. \times \int_0^{2\pi} d\varphi' P_n^m(\mu) P_n^m(\mu') \cos m(\varphi - \varphi') \sum_l \sum_i B_{li}(z) P_i(\mu') \cos l\varphi' \right\rangle \end{aligned} \quad (\text{A.28})$$

$$\begin{aligned} &= F_o (4\pi)^{-1} \sum_n \int_0^{z_i} dz \Delta(\sigma_s \chi_n) \pi f^+(z) \sum_l \sum_i B_{li}(z) \sum_m (2 - \delta_{0m}) \frac{(n-m)!}{(n+m)!} \\ &\quad \times \int_{-1}^1 d\mu \delta(\mu - \mu_{\text{obs}}) P_n^m(\mu) \int_{-1}^1 d\mu' P_i(\mu') P_n^m(\mu') \\ &\quad \times \int_0^{2\pi} d\varphi \int_0^{2\pi} d\varphi' \cos m(\varphi - \varphi') \cos l\varphi' \delta(\varphi - \varphi_{\text{obs}}) \end{aligned} \quad (\text{A.29})$$

$$\begin{aligned} &= F_o \frac{1}{4} \sum_n \int_0^{z_i} dz \Delta(\sigma_s \chi_n) f^+(z) \sum_l \sum_i B_{li}(z) \\ &\quad \times \sum_m (2 - \delta_{0m}) \frac{(n-m)!}{(n+m)!} \cos m \varphi_{\text{obs}} \pi (1 + \delta_{0m}) \delta_{0m} P_n^m(\mu_{\text{obs}}) I_{in}^m \end{aligned} \quad (\text{A.30})$$

$$= F_o \frac{\pi}{2} \sum_m \cos m(\varphi_{\text{obs}}) \sum_n \int_0^{z_i} dz \Delta(\sigma_s \chi_n) f^+(z) \sum_i B_{li}(z) I_{in}^m \left\{ \frac{(n-m)!}{(n+m)!} \right\}^{1/2} P_n^m(\mu_{\text{obs}}). \quad (\text{A.31})$$

The summation limits on  $m, n$  and  $l$  are the same as for the 11 term. The derivation of the diffuse–direct (21) is quite similar to the 12 term. Finally, we examine the direct–direct (22) term:

$$\Delta E_2^{22} = \langle I_s^+, \Delta L_2 I_s \rangle \quad (\text{A.32})$$

$$\begin{aligned} &= \left\langle I_s^+, F_o (4\pi)^{-1} \int_{-1}^1 d\mu' \int_0^{2\pi} d\varphi' \sum_{n=0}^{\infty} \Delta(\sigma_s \chi_n) \sum_{m=0}^n (2 - \delta_{0m}) \right. \\ &\quad \left. \times \frac{(n-m)}{(n+m)} P_n^m(\mu) P_n^m(\mu') \cos m(\varphi - \varphi') \pi f(z) \delta(\mu' - \mu_{\text{sol}}) \delta(\varphi') \right\rangle \end{aligned} \quad (\text{A.33})$$

$$\begin{aligned}
 &= F_o(4\pi)^{-1} \sum_{n=0}^{\infty} \int dz \Delta(\sigma_s \chi_n) \pi f(z) \pi f^+(z) \sum_{m=0}^n (2 - \delta_{0m}) \frac{(n-m)!}{(n+m)!} \\
 &\quad \times \int_{-1}^1 d\mu \delta(\mu - \mu_{\text{obs}}) P_n^m(\mu) \int_{-1}^1 d\mu' \delta(\mu' - \mu_{\text{sol}}) P_n^m(\mu') \\
 &\quad \times \int_0^{2\pi} d\varphi \int_0^{2\pi} d\varphi' \cos m(\varphi - \varphi') \delta(\varphi - \varphi_{\text{obs}}) \delta(\varphi')
 \end{aligned} \tag{A.34}$$

$$= F_o \frac{\pi}{4} \sum_m \cos m\varphi_{\text{obs}} (2 - \delta_{0m}) \sum_{n=0}^{\infty} \int dz \Delta(\sigma_s \chi_n) \pi f(z) \pi f^+(z) \frac{(n-m)!}{(n+m)!} P_n^m(\mu_{\text{obs}}) P_n^m(\mu_{\text{sol}}). \tag{A.35}$$

During our calculations we have made use of the integrals

$$\tilde{I}_{kn}^m \equiv \int_{-1}^1 d\mu P_k(\mu) P_n^m(\mu) \tag{A.36}$$

over one ordinary, and one associated Legendre function. We first note that

$$P_k(\mu) = \sum_{j=0}^k A_j^k \mu^j \tag{A.37}$$

with the constraint that  $k - j$  is always even. The expansion coefficients,  $A_j^k$ , are given explicitly by

$$A_k^j = (-1)^{(k-j)/2} \frac{(k+j-1)!!}{j!(k-j)!!}. \tag{A.38}$$

These coefficients may be constructed by iteration for maximum computational stability.

Next, we note that

$$\begin{aligned}
 P_n^m(\mu) &= (1 - \mu^2)^{m/2} \frac{d^m}{d\mu^m} P_n(\mu) \\
 &= (1 - \mu^2)^{m/2} \sum_{i=m}^n A_i^n \frac{i!}{(i-m)!} \mu^{i-m}.
 \end{aligned} \tag{A.39}$$

Therefore,

$$\begin{aligned}
 \tilde{I}_{kn}^m &= \sum_{j=0}^k \sum_{i=m}^n A_j^k A_i^n \frac{i!}{(i-m)!} \int_{-1}^1 \mu^{j+i-m} (1 - \mu^2)^{m/2} d\mu \\
 &= \sum_{j=0}^k \sum_{i=m}^n A_j^k A_i^n \frac{i!}{(i-m)!} I_{m+1, i+j-m},
 \end{aligned} \tag{A.40}$$

where

$$I_{pq} = \int_0^\pi \sin^p \theta \cos^q \theta d\theta. \tag{A.41}$$

This integral is zero whenever  $q$  is odd. When  $q$  is even,

$$I_{2p,2q} = \pi \frac{(2p-1)!(2q-1)!!}{2^{p+q}(p+q)!}, \quad (\text{A.42})$$

$$I_{2p-1,2q} = \pi \frac{2^p(p-1)!(q-1)!}{(2p+2q-1)!!}. \quad (\text{A.43})$$

These may be again constructed by iteration for maximum stability. After constructing the arrays  $A_j^k$  and  $I_{pq}$ , the final double summation in Eq. (A.40) is done. Note that  $\tilde{I}_{kn}^m$  is non-zero only if  $m$  is not greater than  $n$ , and  $k+n+m$  is even.

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