

GROUND-BASED ATMOSPHERIC PROFILING OF OZONE USING HIGH SPECTRAL RESOLUTION RADIOMETRIC DATA

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1. INTRODUCTION

The need to provide better estimates of temperature and trace gas concentrations in the atmosphere has led to the advent of high spectral resolution instruments yielding high quality, calibrated atmospheric radiance spectra. The improved capability for vertical profiling of atmospheric temperature and trace gases using these data has been demonstrated (Smith et al., 1990, Smith et al., 1991). Presented here is a report on progress to date with a linear retrieval scheme for ozone concentration profiles using high spectral resolution data. Ozone was chosen as the retrieval product due to its existence in measurable quantities in both the troposphere and the stratosphere. An improved knowledge of ozone concentration is a significant variable impacting our knowledge of atmospheric transmittance.

A constrained linear least squares solution scheme has been applied to synthetic data generated from FASCODE. The synthetic spectra were generated at an appropriate spectral resolution to simulate measured atmospheric spectra from the Ground-Based High-resolution Interferometer Sounder (GB-HIS). This paper does not address all the issues affecting ozone profile retrieval but concerns itself primarily with demonstrating the feasibility of ozone profiling and outlines the problem introduced by a non-linear radiometric response.

2. DERIVATION OF LINEAR PERTURBED RTE

The radiative transfer equation (RTE) for downwelling infrared radiation is

$$R_\nu = \int_0^{p_s} B_\nu(T) \frac{d\tau_\nu}{dp} dp \quad (1)$$

where R_ν = downwelling spectral radiance at ν

$B_\nu(T)$ = Planck radiance at temperature T

τ_ν = transmittance from pressure, p , to the surface. By definition, $\tau_\nu(p_s) = 1$

To obtain the perturbed form of the upward looking RTE, an initial condition - the guess state - is defined

$$R_v^o = \int_0^{p_i} B_v(T^o) \frac{d\tau_v^o}{dp} dp \quad (2)$$

and the radiance perturbation is described by

$$\delta R_v = R_v - R_v^o \quad (3)$$

so from (3) the perturbed radiative transfer equation (PRTE) is formed

$$\begin{aligned} \delta R_v &= \int_0^{p_i} B_v(T) \frac{d\tau_v}{dp} dp - \int_0^{p_i} B_v(T^o) \frac{d\tau_v^o}{dp} dp \\ &= \int_0^{p_i} \delta B_v(T) \frac{d\tau_v^o}{dp} dp - \int_0^{p_i} \delta\tau_v \frac{dB_v(T)}{dp} dp - [B_v(T)\delta\tau_v]_0^{p_i} \end{aligned} \quad (4)$$

An expression must be found for $\delta\tau_v$ in terms of the mixing ratio perturbation δq_i where i labels a particular absorbing constituent. A positive perturbation in mixing ratio at any level will affect the transmittance value at every level *above* the perturbed level due to the increased opacity of that layer, whereas it will remain unchanged *below* the perturbed level. It is therefore necessary to define a change in transmittance at pressure p due to a change in mixing ratio at some other pressure p' where the integration limits identify the region of atmosphere over which the transmittance perturbation is evaluated (i.e. from pressures 0 to p).

The transmittance perturbation, as a first order approximation, may be written as

$$\begin{aligned} \delta\tau_v(p) &= \tau_v^o \delta \ln \tau_v \\ &= -\frac{\tau_v^o}{g} \int_p^{p_i} \delta q_i(p') k_{\nu} dp' \end{aligned} \quad (5)$$

so that, as the integration proceeds, the transmittance perturbation at p is zero below the perturbed level at p' and modified above p' . This is based on the assumption that the transmittance at the surface is unity.

Since at $p = 0$, $T \rightarrow 0$, so $B_v(T) \rightarrow 0$; and from (5) $\delta\tau_v = 0$ at $p = p_i$, then (4) becomes

$$\delta R_v = \int_0^{p_i} \delta B_v(T) \frac{d\tau_v^o}{dp} dp - \int_0^{p_i} \delta\tau_v \frac{dB_v(T)}{dp} dp \quad (6)$$

The first part of (6) can be treated as in Smith *et al.* (1991) and is concerned with the temperature perturbation, δT , derived from δB . The second term of (6) describes the effect on the radiance perturbation

arising from a mixing ratio perturbation, δq . For the work described here, the temperature and all other constituent profiles were held constant and only the ozone mixing ratio profile was perturbed. Under these conditions, $\delta T = 0$. This approach was adopted to evaluate how well the retrieval scheme performed for ozone concentration profile perturbations only as, in a real atmosphere, δq would modify δT . Also, δT would be determined in a spectral region which does not include ozone, such as the $15\mu\text{m}$ CO_2 region.

Considering just the second term of (6), if a pressure level p_{top} is defined which is equal to some small (but non-zero) value above which $\delta\tau_v$ is constant, assuming that the mixing ratios are constant, then we have

$$\begin{aligned} \int_0^{p_i} \delta\tau_v \frac{dB_v(T)}{dp} dp &= \int_0^{p_{top}} \delta\tau_v \frac{dB_v(T)}{dp} dp + \int_{p_{top}}^{p_i} \delta\tau_v \frac{dB_v(T)}{dp} dp \\ &= \delta\tau_{v,total} B_v(T_{top}) + \int_{p_{top}}^{p_i} \delta\tau_v \frac{dB_v(T)}{dp} dp \end{aligned} \quad (7)$$

where $\delta\tau_{v,total}$ = transmittance perturbation of the entire atmospheric column

Substituting (5) into (7) gives

$$\delta R_v(\delta q_i) = \int_{p_{top}}^{p_i} \frac{dB_v}{dp} \left[\frac{\tau_v^o}{g} \int_p^{p_i} \delta q_i(p') k_{\nu} dp' \right] dp + \frac{B_v(T_{top}) \tau_v^o}{g} \int_{p_{top}}^{p_i} \delta q_i(p') k_{\nu} dp' \quad (8)$$

which can be simplified by integrating by parts so the radiance perturbation due to a mixing ratio perturbation now may be written as

$$\delta R_v(\delta q_i) = \frac{1}{g} \int_{p_{top}}^{p_i} \delta q_i(p') k_{\nu} \left[\int_{p_{top}}^p \tau_v \frac{dB_v}{dp} dp + \tau_{v,total} B_v(T_{top}) \right] dp' \quad (9)$$

which is a linear integral equation. Equation (9) may be more clearly expressed as

$$\delta R_v(\delta q_i) = \int_{p_{top}}^{p_i} \frac{\delta q_i(p')}{q_i^o(p')} W_Q dp' \quad (10)$$

$$W_Q(T^o, \tau_v^o, q_i^o) = \frac{k_{\nu} q_i^o(p')}{g} \left[\int_{p_{top}}^p \tau_v \frac{dB_v}{dp} dp + \tau_{v,total} B_v(T_{top}) \right] \quad (11)$$

where (10) is a Fredholm equation of the first kind with fixed limits. Note that in (11) the quantity k_i is the absorption coefficient for the i^{th} molecule only and τ is the transmittance for *all* absorbing constituents present.

3. SOLUTION SCHEME FOR LINEAR MODEL

The constituent mixing ratio term of the linearised PRTE, (9), may be recast as a matrix equation

$$\mathbf{r} = \mathbf{A} \mathbf{q} \tag{12}$$

where $\mathbf{r} = (N \times 1)$ radiance perturbation vector δR

$\mathbf{q} = (L \times 1)$ mixing ratio perturbation vector $\delta q/q^o$

$\mathbf{A} = (N \times L)$ matrix of layer radiance perturbations $\mathbf{W}_Q dp$

$N =$ no. of spectral channels

$L =$ no. of atmospheric levels to be retrieved

and for the synthetic and GB-HIS data we have $N > L$, i.e. the system is overdetermined.

A solution may be obtained using a constrained linear least squares method (Twomey, 1963, Hayden, 1988).

The solution is then

$$\mathbf{q} = [\mathbf{A}^T \mathbf{A} + \gamma \mathbf{I}]^{-1} \mathbf{A}^T \mathbf{r} \tag{13}$$

where $\gamma =$ Lagrangian multiplier reciprocal

$\mathbf{I} =$ identity matrix

The matrix \mathbf{A} describes the contributing radiance perturbation at each wavenumber (channel) for each pressure level in the system. As the pressure layer thickness, dp , decreases the elements of \mathbf{A} also decrease. FASCODE (Clough et al., 1981, Clough et al., 1986), which was used to calculate the layer radiances, does not allow layers of uniform pressure difference throughout the atmosphere. The consequence of using radiance perturbations sensitive to layer thickness is that the radiance contribution from locally thinner layers is necessarily smaller and the retrievals for these layers are driven towards the guess mixing ratio.

The layer radiance perturbation dependence on layer thickness may, by definition, be removed by dividing out the logarithmic pressure differential, $d \ln p$. This changes (12) to

$$\mathbf{r} = \frac{1}{(d \ln p)} \mathbf{A} \cdot (d \ln p) \mathbf{q} \tag{14}$$

which gives

$$(d \ln p) \mathbf{q} = \left[\frac{\mathbf{A}^T \mathbf{A}}{(d \ln p)^2} + \gamma \mathbf{I} \right]^{-1} \frac{1}{(d \ln p)} \mathbf{A}^T \mathbf{r} \tag{15}$$

This is the same as

$$\mathbf{q} = [\mathbf{A}^T \mathbf{A} + (d \ln p)^2 \gamma \mathbf{I}]^{-1} \mathbf{A}^T \mathbf{r} \tag{16}$$

Thus dividing the layer radiance perturbations by $d\ln p$ is the same as weighting γ by $(d\ln p)^2$. It should be noted that while (15) and (16) are theoretically similar, the results obtained from each may be different due to computational precision. Equation (15) divides matrix elements by $d\ln p$, typically 0.05 - 0.4, magnifying any rounding errors, whereas (16) modifies γ , multiplying by a small number thereby keeping any rounding errors small.

4. LINEAR MODEL RADIANCE PERTURBATION CALCULATION

4.1 Mixing ratio perturbation function

A Gaussian perturbation for the mixing ratio has been used in the linear model to calculate the forward radiance perturbation. The perturbation function used was

$$\frac{\delta q_i(p)}{q_i^o(p)} = \frac{\delta q_i(p_o)}{q_i^o(p_o)} \exp[-a(p - p_o)^2] \quad (17)$$

where p_o = pressure of peak perturbation

a = slope constant of Gaussian curve, $\ln(2)/\alpha^2$

α = perturbation at half maximum

Different halfwidths have been used for the lower and higher pressure sides of the peak to satisfy the condition that above some pressure p_{top} , $\delta\tau$ is constant assuming the q_i 's are constant. The terms α_{lo} and α_{hi} are used to describe the perturbation halfwidths on the low and high pressure sides of the perturbation maximum respectively.

4.2 Tropospheric ozone mixing ratio perturbation

A linear scheme may be used with some success in the troposphere due to the mainly linear radiometric response of atmospheric ozone in this region. Figure 1 shows the calculated radiance difference, ΔR , for FASCODE and the linear model for a relatively large tropospheric ozone concentration perturbation ($p_o=1000\text{mb}$, $\alpha_{lo}=100\text{mb}$, $\delta q_i/q_i^o=2$). The differences between these models (figure 2.) are very close to the noise level ($0.1\text{mW/m}^2.\text{st.cm}^{-1}$)^a but larger than the noise level in two regions near 1043cm^{-1} and $1050-1060\text{cm}^{-1}$. These differences may be attributed to a non-linear component in the tropospheric response. Figure 3 shows the relationship between ΔR and $\delta q_i/q_i^o$ for a selection of wavenumbers in the ozone $9.6\mu\text{m}$ band showing the predominantly linear response. As the overall magnitude of the radiance differences increases, a linear approximation to a non-linear system would begin to break down, as occurs in the $1050-1060\text{cm}^{-1}$ region. The reason for the relatively large difference at $\approx 1043\text{cm}^{-1}$ has yet to be identified, but may be due to the spectroscopic data used in the FASCODE calculation. Here the ozone signal is the weakest and good agreement between the models would be expected.

^a Nominal noise level for GB-HIS system.

Calculated radiance perturbations for a TROPOSPHERIC ozone perturbation

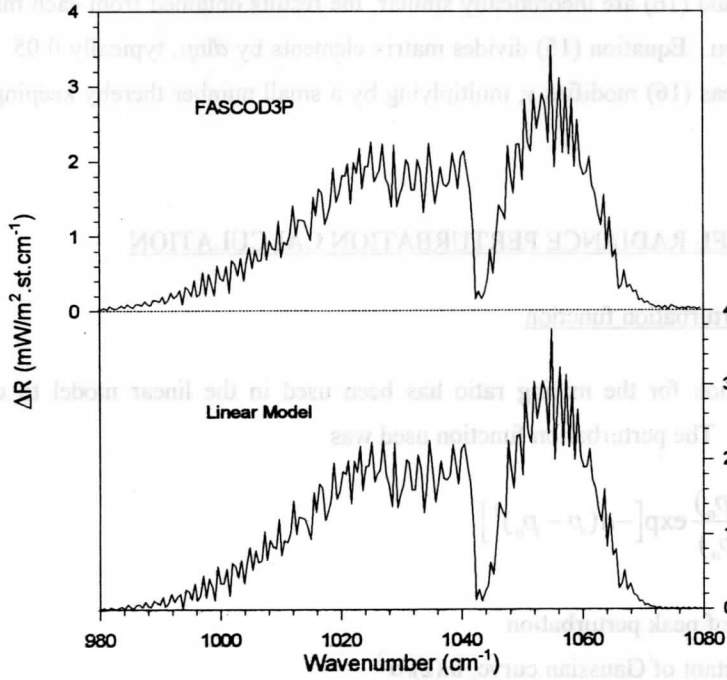


Figure 1.

Difference in FASCOD3P and linear model ΔR for a TROPOSPHERIC ozone perturbation

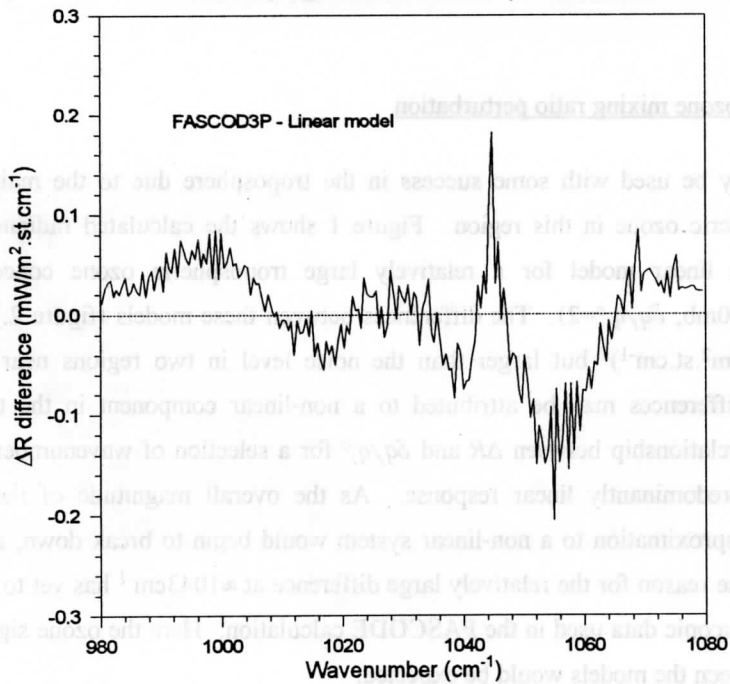


Figure 2.

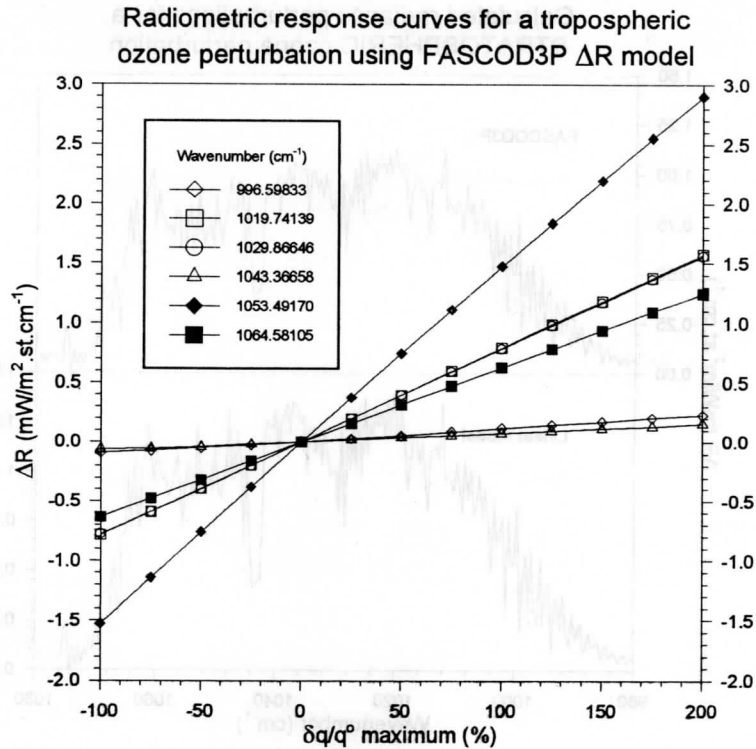
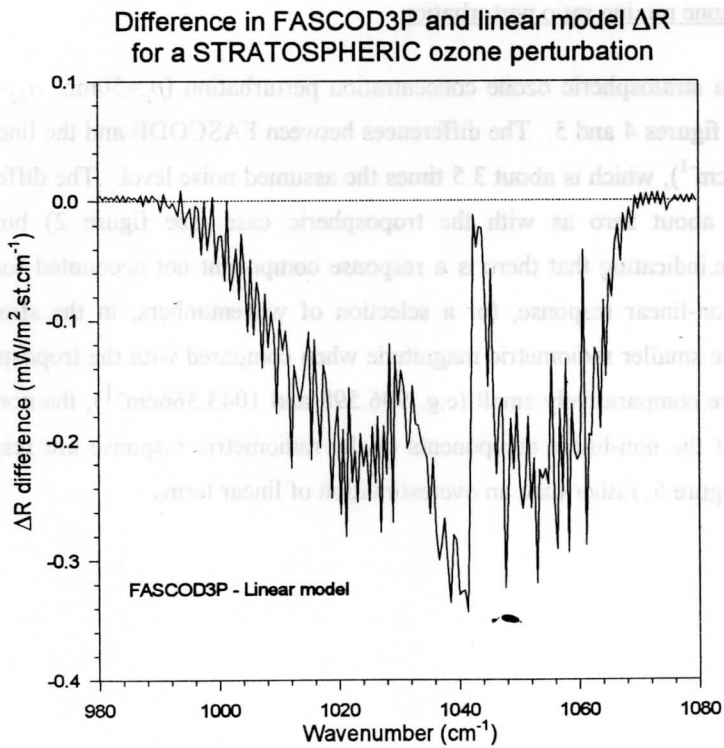
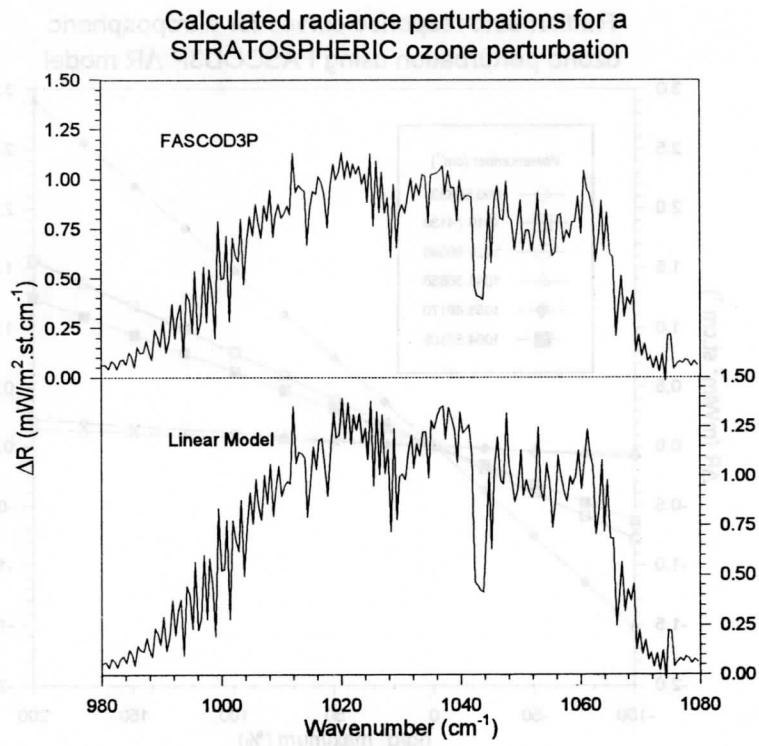


Figure 3.

4.3 Stratospheric ozone mixing ratio perturbation

Similarly, results for a stratospheric ozone concentration perturbation ($p_o=50\text{mb}$, $\alpha_{lo}=10\text{mb}$, $\alpha_{hi}=100\text{mb}$, $\delta q_i/q_i^o=1$) are shown in figures 4 and 5. The differences between FASCODE and the linear model peak around $0.30\text{--}0.35\text{ mW}/(\text{m}^2.\text{st}.\text{cm}^{-1})$, which is about 3.5 times the assumed noise level. The difference shown in figure 5 does not fluctuate about zero as with the tropospheric case (see figure 2) but has a recognisable spectroscopic structure, indicating that there is a response component not accounted for by the linear model. Figure 6 shows the non-linear response, for a selection of wavenumbers, in the stratosphere to an ozone perturbation despite the smaller radiometric magnitude when compared with the troposphere. Even where the radiance differences are comparatively small (e.g. 996.598 and 1043.366cm^{-1}), the non-linearity is apparent. Figure 6 confirms that the non-linear components of the radiometric response are responsible for the large differences shown in figure 5, rather than an overestimation of linear terms.



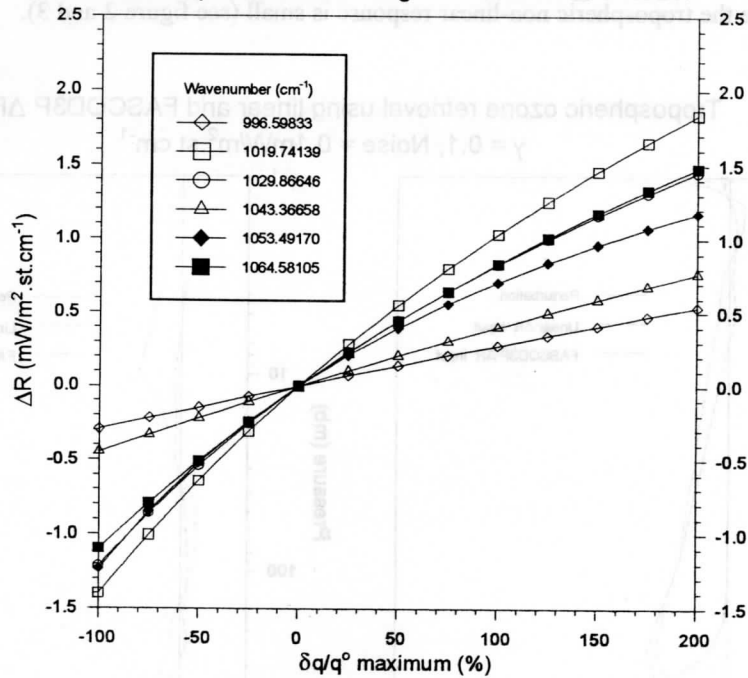
Radiometric response curves for a stratospheric ozone perturbation using FASCOD3P ΔR model

Figure 6.

5. OZONE MIXING RATIO RETRIEVALS FROM SYNTHETIC DATA

The following is a summary of our progress to date with retrieval performance using both the calculated forward linear model ΔR and the FASCODE calculated ΔR . The Lagrangian multiplier reciprocal, γ , used in the solution scheme has been adjusted to provide the sharpest vertical resolution functions while still providing stable, physically acceptable results. However, it should be noted that the system has not been optimised for γ nor has any smoothing been applied to the system matrix A .

5.1 Tropospheric perturbation retrieval

The retrieval of the tropospheric ozone perturbation (as defined in §4.2) shown in figure 7 does not work well because the solution scheme using (13) weights every layer equally, i.e. γ is held constant for all p . The effect of this is to smear out the perturbation feature in the retrieval. The feature at 800-950mb is due to the layering scheme chosen. In this region there are locally "thinner" layers, namely 40mb thick compared with 50mb thick layers surrounding them. Weighting γ by $(d \ln p)^2$ (figure 8) as in (16) permits the retrieval scheme to recover the perturbation feature much better, but major instabilities in these retrievals still occur in the 800-950mb region. The increased sensitivity of the retrieval magnifies these features in figures 8 compared with that of figure 7. Tuning the system by optimising γ may provide more stable solutions than those shown in figure 8 but too large a γ value also will degrade the vertical resolution of the retrieval. The retrievals using a linear forward ΔR perform better than those using a non-linear ΔR . This is expected as the solution scheme is linear

but, once again, optimising γ might also improve the retrievals using a non-linear forward radiance perturbation because the tropospheric non-linear response is small (see figure 2 and 3).

Tropospheric ozone retrieval using linear and FASCOD3P ΔR input
 $\gamma = 0.1$, Noise = $0.1 \text{ mW/m}^2 \cdot \text{st} \cdot \text{cm}^{-1}$

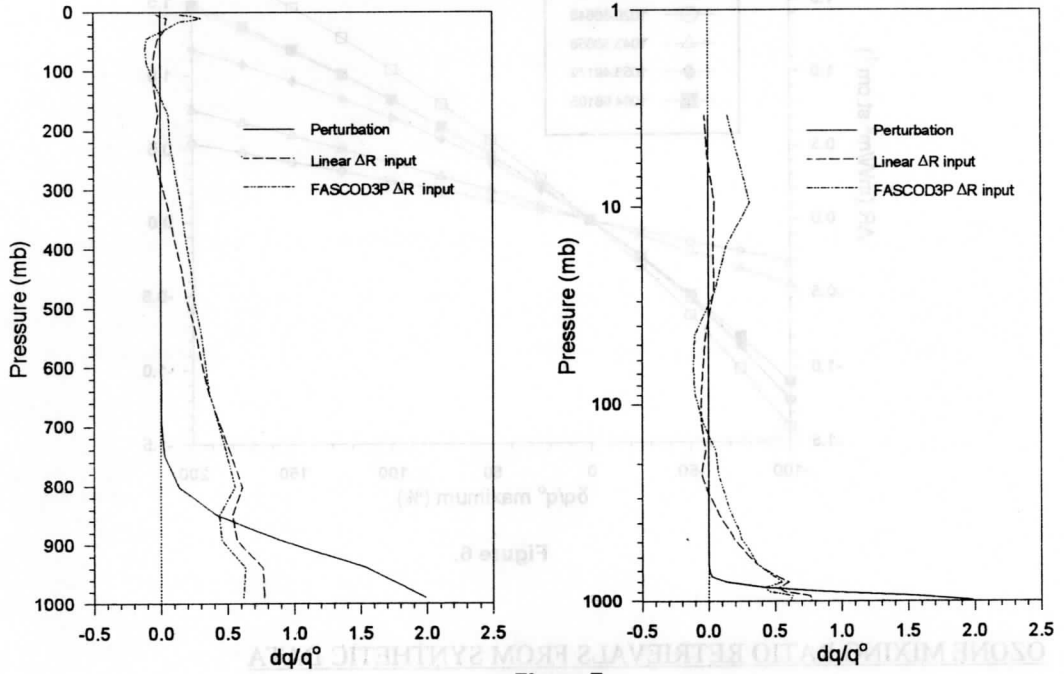


Figure 7.

Tropospheric ozone retrieval using linear and FASCOD3P ΔR input
 γ weighted by $(\ln p)^2$; $\gamma = 1.0$, Noise = $0.1 \text{ mW/m}^2 \cdot \text{st} \cdot \text{cm}^{-1}$

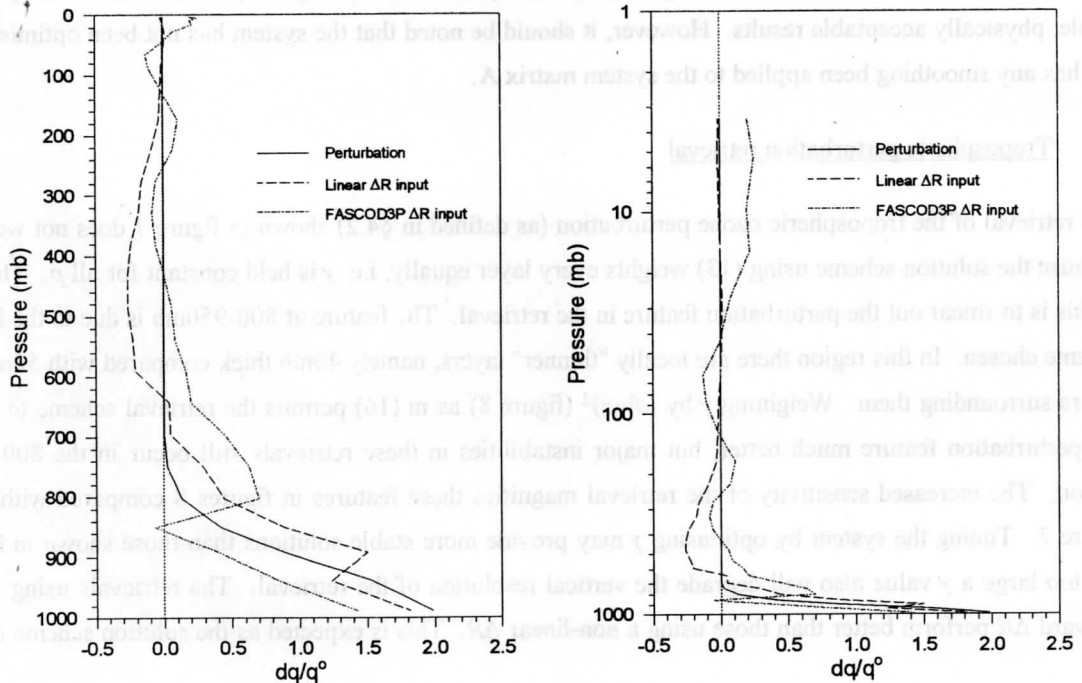


Figure 8.

5.1 Stratospheric perturbation retrieval

In contrast to the tropospheric retrievals, weighting γ by $(d\ln p)^2$ did not have as large an impact. Figure 9 shows the unweighted γ retrieval. The retrieval recovers only 50% of the perturbation, but the stability of the scheme in the troposphere is very good. Figure 9 also suggests that the integrated ozone signal has been retrieved quite well. The $\gamma(d\ln p)^2$ system retrieval (figure 10) locates the perturbation feature somewhat better but there is a significant spread in the retrieved profile because of the large value of γ employed. It was necessary to use a large γ value because the result was extremely unstable for small γ . Figure 10 shows some small instabilities in the solution remain in the troposphere. This result suggests that optimising γ is a priority in this solution scheme. The vertical smearing of the retrieved profiles in the region above the perturbation (figure 9), which is due to the very rapid change in the perturbation function and the sharpness of the weights in the upper stratosphere, also may be minimised by adjusting γ .

The retrieved perturbations to the stratospheric profiles are in good agreement with the input mixing ratio perturbation when one considers the differences in magnitude between the calculated linear and FASCOD3P forward radiance perturbations (see figure 5). However, the accuracy of these retrievals must be questioned until a more representative estimate of the forward radiance perturbation is used in the solution scheme.

Stratospheric ozone retrieval using linear and FASCOD3P ΔR input
 $\gamma = 0.5$, Noise = $0.1 \text{ mW/m}^2 \cdot \text{st} \cdot \text{cm}^{-1}$

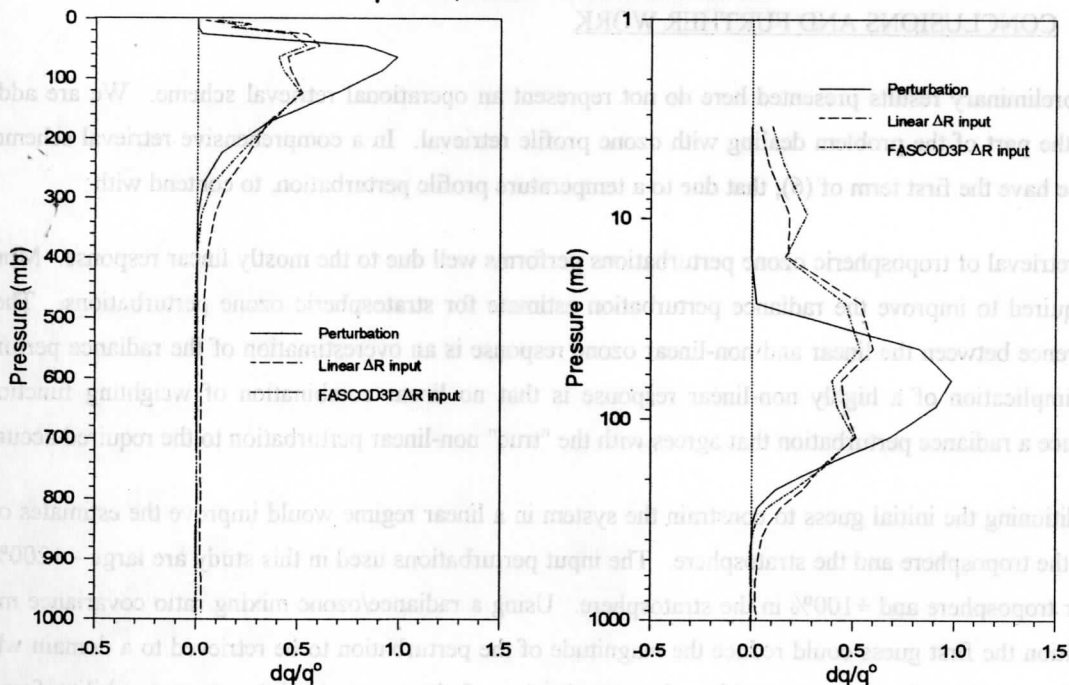


Figure 9.

Stratospheric ozone retrieval using linear and FASCOD3P ΔR input
 γ weighted by $(d\ln p)^2$; $\gamma = 5.0$, Noise = $0.1 \text{ mW/m}^2 \cdot \text{st.cm}^{-1}$

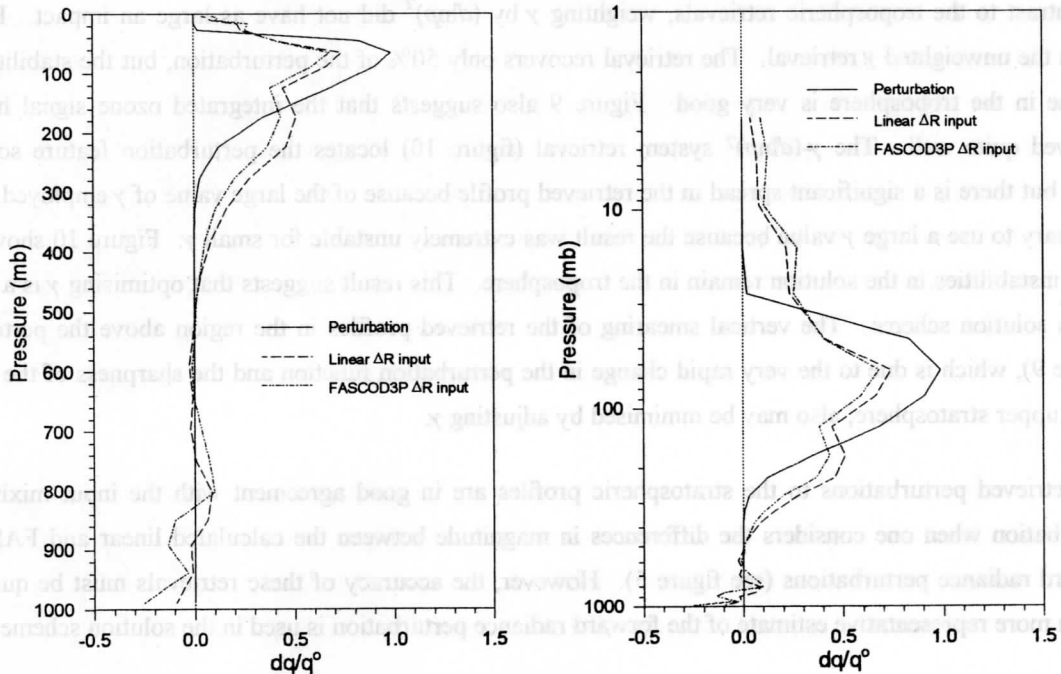


Figure 10.

6. CONCLUSIONS AND FURTHER WORK

The preliminary results presented here do not represent an operational retrieval scheme. We are addressing only the part of the problem dealing with ozone profile retrieval. In a comprehensive retrieval scheme we of course have the first term of (6), that due to a temperature profile perturbation, to contend with.

The retrieval of tropospheric ozone perturbations performs well due to the mostly linear response. More work is required to improve the radiance perturbation estimate for stratospheric ozone perturbations. The major difference between the linear and non-linear ozone response is an overestimation of the radiance perturbation. The implication of a highly non-linear response is that no linear combination of weighting functions will produce a radiance perturbation that agrees with the "true" non-linear perturbation to the required accuracy.

Conditioning the initial guess to constrain the system in a linear regime would improve the estimates of ΔR in both the troposphere and the stratosphere. The input perturbations used in this study are large - +200% in the lower troposphere and +100% in the stratosphere. Using a radiance/ozone mixing ratio covariance matrix to condition the first guess could reduce the magnitude of the perturbation to be retrieved to a domain where the non-linear effects become acceptable. An examination of short-term (daily) ozone variability from ozone sondes taken during FIRE II/SPECTRE suggests that deviations from a guess profile may be large (a few hundred percent) in the tropopause region, so a reliable first guess is essential. Coupled with determining more workable constraints on the linear solution scheme is the task of improving retrieval accuracy by optimising the choice of γ . In this regard we are looking at elements of King (1982) to determine the ideal choice of γ based on both physical and statistical information. Our work is also proceeding to investigate an

iterative linear least squares solution scheme for trace gas retrievals in situations where the non-linear response is a dominant effect in the forward radiance perturbation.

Another aspect of ozone profiling is that, apart from the water vapour continuum, the non-linearities evident in the ozone radiometric response are spectrally isolated with respect to other atmospheric constituents. Application of this method to other trace gas species, methane for example, has not yet been attempted. The effect of other molecules or species, such as water vapour, on the radiative response of trace gases and the subsequent impact on this retrieval scheme has yet to be studied.

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