Status of the Solar and Infrared Radiation Submodels in the LLNL 1-D and 2-D Chemical-Transport Models

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Introduction

The LLNL 1-D and 2-D chemical-transport models of the troposphere and stratosphere are useful diagnostic and prognostic tools for studying chemical, dynamical, and radiative processes and interactions in the atmosphere. These uses include analysis of the temporal and spatial variation of trace gases in the troposphere and stratosphere (and, as appropriate, the mesosphere). Direct comparison of model concentration estimates can be made for species for which satellite and other observations are available. Via model chemistry and physics, concentrations of unmeasured species can be estimated consistent with concentrations of observed species.

Incorporating accurate radiative transfer submodels is an important step in developing such chemical-transport models. Calculation of new temperatures based on feedback from changes in species concentrations has long been an important capability of the LLNL 1-D model (Luther et al., 1976). Currently, improvements in both radiative transfer algorithms and computer speed prompt implementing more accurate calculations of radiative heating rates in both the LLNL 1-D and 2-D models.

The radiation submodel development described in this report has already substantially upgraded our capabilities and paves the way for future improvements. In our most recent solar submodel, we have combined our former accurate computation of absorption coefficients with more accurate and general two-stream solution techniques. This development provides us the capability of treating clouds and aerosols in addition to clear sky cases. We have added absorption by H_2O , CO_2 and O_2 in the near infrared, and now use a state-of-the-art infrared parameterization for absorption and emission by CO_2 , O_3 and H_2O .

An immediate motivation for developing new radiation transport submodels for the LLNL models is the computation of accurate net radiative heating rates within each model layer. This is particularly true for the 2-D model, in which these heating rates are in turn used to calculate vertical and meridional wind fields, thereby affecting both the transport of species and the temperature field. These latter changes in turn, affect the chemistry and concentration of species, thereby feeding back into the calculated heating rates as well as directly changing rates of photochemical reactions.

This particular feedback cycle represents an intermediate state in the evolution of model treatment of interactions between dynamics and radiant heating. In the recent past, transport and temperatures in the 2-D model were both either fixed or parameterized. Currently, temperatures are still prescribed, but transport is now dependent on radiant and latent heating. The technique used to derive the residual meridional and vertical wind fields is similar to that of Murgatroyd and Singleton (1961) and Dunkerton (1978). In the near future, we intend to extend the LLNL 2-D model by calculating the residual mean

circulation through a primitive equation dynamical submodel that will calculate both the wind and temperature fields, consistent with the calculated heating rates.

Requirements for the radiation codes used in the LLNL models are accuracy and speed of computation. Highly accurate methods for obtaining benchmark solutions are available in the literature for both the solar and terrestrial spectral regions. For ultraviolet and visible wavelengths, numerous computational techniques have been developed to solve accurately the radiation transport equation for the directionally dependent radiance or full Stokes matrix (Lenoble, 1985; Hansen and Travis, 1974). These calculations include the effects of scattering and absorption by molecules, aerosols, and clouds, and can be made accurate to the extent that the optical properties of these constituents can be determined by observations and theory. In the infrared, calculations integrated line-by-line (LBL) through the gaseous absorption spectra constitute the current state-of-the-art reference standards against which the accuracy of parameterized calculations are customarily judged (WMO, 1984, 1985). In both spectral regions, the very precise and detailed treatment which these calculations represent makes them far too expensive for use in chemical-transport or general circulation models. They do, however, play a very practical role as calibration standards for less detailed radiation models actually used. A number of these simplified schemes are reviewed by Stephens (1984), and King and Harshvardhan (1986).

The LLNL Chemical-Transport Models

The current LLNL 2-D chemical-transport model uses a grid with 16 latitudes and 18 vertical layers. The vertical layers are uniformly spaced in log-pressure between the surface and 0.56 mb, corresponding approximately to a layer thickness of 3 km between the surface and 54 km. The 1-D model has 44 vertical layers between the surface and 56.25 km, with approximate layer depths of 1 km up to 35 km, and depths of 2.5 km above 35 km. The geometry of the radiation transport problem for a single model layer is shown in Figure 1.

Pressure, temperature, and concentrations of over thirty species within each model layer and latitude zone (2-D version only) are among the state variables of the LLNL models, therefore directly available as input to the radiation transport submodels. Radiatively active species of particular interest include O_2 , O_3 , NO_2 , H_2O , CO_2 , CH_4 , N_2O , and several chlorofluorocarbons. The diurnally averaged versions of the models calculate noontime values of the solar zenith angle as a function of latitude and season. In addition to these internal model variables, a number of climatologically based parameterizations have been included. As described in Appendix A, seasonally and latitudinally dependent surface-albedo, surface-temperature, and fractional cloud cover parameterizations were developed. Surface albedo and fractional cloud cover are input required by the solar radiation submodel. Surface temperature and fractional cloud cover are required by the terrestrial radiation submodel. The column-total release of latent heat in the troposphere was parameterized from latitude-dependent monthly rainfall data (Jaeger, 1976; Potter and Gates, 1984). The algorithm used to vertically distribute the column-total latent heat release among model layers is discussed in Appendix B. The latent heating is added to the solar heating and infrared cooling to obtain the net heating rates.

Absorption and Scattering of Solar Radiation

The LLNL chemical-transport models use three separate radiation submodels to calculate solar heating rates. The majority of solar heating is calculated by a submodel treating absorption by O_2 , O_3 , and NO_2 in the UV and visible spectral regions (133.3 nm to 735 nm). A second submodel is used to calculate heating from absorption by H_2O in the near infrared. The final solar submodel calculates near infrared heating due to absorption by CO_2 and O_2 .

The LLNL models currently treat shortwave absorption by O_2 , O_3 and NO_2 based on the 148 wavelength bins and the corresponding absorption coefficients previously used in models for the LLNL models for photodissociation calculations (Connell and Wuebbles, 1983). While this degree of explicit wavelength detail is needed for photodissociation calculations, this may not be necessary for calculation of solar heating rates and we may reduce this number of bins in the near future.

For part of the interim calculations and comparisons reported here, a multiple-scattering scheme previously developed by Luther (1981) was used (Figure 2). This scheme, which accounts only for Rayleigh scattering, explicitly follows five orders of scattering, assuming in each case that the flux scattered from a given layer is scattered equally in the upwards and downwards directions. However, all future calculations with the 1-D and 2-D models will use the much more accurate and complete multiple-scattering routine described below.

To enable the LLNL models to treat the effects of both clouds and aerosols, and to provide a more accurate dependence of heating rates on solar zenith angle, a new two-stream model has been developed and is currently being tested. The O_3 heating rates calculated with this new model for a clear sky atmosphere are compared in Figure 2 with heating rates for the same conditions calculated by the old LLNL model and the O_3 model of Harshvardhan et al. (1987). On completion of testing, this algorithm will supersede the algorithm based on Luther (1981). The new model uses the Sagan and Pollack (1967) algorithm within each layer to obtain reflection and transmission fractions for the diffuse flux incident on the layer. The solar zenith angle dependent fractional sources of upwards and downwards diffuse radiation due to scattering from the direct beam are obtained by use of the delta-Eddington approximation (Joseph et al., 1976; King and Harshvardhan, 1986). This is done for each separate layer, in each case assuming a unit direct-beam flux incident at the top of the current layer. The direct-beam transmission of the layer is also calculated at this point.

For each layer, reflection and transmission fractions, upwards and downwards source fractions, and the direct beam transmission are calculated both for a cloud-free layer and for a totally cloudy layer. Cloud optical properties are parameterized as functions of model layer and wavelength. Climatological cloudiness fractions are used, as discussed in Appendix A. Cloud overlap is treated by linearly averaging each reflection, transmission, source, or beam transmission operator proportionally to the fractional cloudiness assigned to the layer. This technique is similar to the method suggested by Morcrette and Fouquart (1986) and Fouquart and Bonnel (1980). It is an approximation to the random cloud-overlap assumption, with the advantage of decreasing the number of column flux solutions from sixteen down to one, for the four layers of clouds being used.

Using the direct beam transmission calculated for each layer (after averaging for fractional cloudiness), the direct beam flux transmitted from the top of the atmosphere down to the current layer is calculated. The upwards and downwards fluxes at each level are then calculated from the diffuse reflection, diffuse transmission and source (diffuse radiation scattered from the direct beam) operators using a layer adding algorithm. This algorithm is described in detail by Davies (1982) and more briefly by Harshvardhan et al. (1987). It is a simplification for two-stream flux calculations of the more general algorithm of Grant and Hunt (1969). The resultant fluxes include the effects of multiple scattering within the atmosphere as well as those of separate surface albedos for diffuse and direct-beam incident radiation. The direct-beam flux is included in the downward flux. Finally, layer heating rates are obtained from the vertical divergence of the net downward flux (i.e., downward flux minus upward flux). Although aerosols are not included in the version of the model currently being tested, future inclusion only requires appropriately modifying the layer optical depths, asymmetry factors, and single-scattering albedos used for the cloudy and cloud-free operator calculations described above.

For the treatment of near-infrared absorption by H_2O , the model described by Harshvardhan et al. (1987) and Davies (1982) has been adapted to the LLNL model grids. The clear-sky solution is obtained via the absorption parameterization of Lacis and Hansen (1974). For the cloudy-sky case, the model uses a five-term exponential sum to calculate absorption and scattering. Except for the treatment of cloud properties and overlap, the solution technique is identical with that described immediately above. At present, this submodel does not account for fractional cloud overlap directly. The cloud optical depth is instead scaled linearly by cloud amount. Future modifications include making the treatment of clouds consistent with that in the model for the UV and visible regions described above.

An approximate treatment of absorption by CO_2 and O_2 in the near infrared was obtained from Ellingson (1986). For the pertinent absorption bands, absorption from the direct solar beam is calculated, including a correction for overlap of CO_2 absorption with H_2O absorption. Effects of multiple scattering in the near IR are ignored. Since, for purposes of calculating heating rates, this near infrared absorption itself acts as a correction to the absorption of O_3 , O_2 , and NO_2 in the solar visible, the errors introduced by ignoring multiple scattering for CO_2 and O_2 in the near infrared can be considered to be of second order.

In all of these solar radiation models, diurnal averaging is included by use of approximation number four of Cogley and Borucki (1976). Using the Lacis and Hansen (1974) parameterization for absorption by O_3 , Cogley and Borucki found the ratio of the average from their approximation relative to the exact diurnal average to vary smoothly as a function of altitude, latitude, and solar declination. They observed the approximate diurnal averages to have a maximum error of about 4%. The coefficients obtained from this approximation are an effective solar zenith angle and an effective daylight coefficient, the latter multiplying the direct beam flux incident at the top of the atmosphere. The equations used to calculate these coefficients are obtained by matching properties of the approximation (such as area or moments) with those of the original integrals over solar

day, avoiding empirical corrections. The resulting coefficients are not the same as the average solar zenith angle during daylight hours and the fraction of the day that the sun is above the horizon, which together constitute a less accurate approximation to the diurnal average.

Absorption of Terrestrial Radiation

The model developed by Harshvardhan and Corsetti (1984) and Harshvardhan et al. (1987), adapted to the LLNL model grid, is currently being used to calculate the longwave emission and absorption by CO_2 , O_3 , and H_2O . For CO_2 absorption, the model uses the broadband parameterization of Chou and Peng (1983), for water vapor that of Chou (1984), and for O_3 the method of Rodgers (1968). In treating absorption by CO_2 and H_2O along optical paths through a vertically inhomogeneous atmosphere, use is made of the far-wing scaling approximation discussed by Chou and Arking (1980). This scaling assumes that the pressure and temperature dependence of the absorption coefficients are separable from the wavelength dependence. For each separate absorption band, the wavelength dependent factor is calculated once for a reference temperature and pressure chosen to be typical of the altitude of greatest absorbance for that band. The pressure dependence is approximated by the power-law scaling correct for the far wings of lines. The temperature dependence is interpolated from tabulated values. For each band, the coefficients used to parameterize the temperature and pressure dependence were determined by fits to line-by-line calculations. The effects of Doppler broadening on the CO_2 parameterization have been modeled by holding the pressure scaling of absorber amount constant above an empirically chosen critical altitude (1 mb in Chou and Peng). The effects of Doppler broadening of O_3 absorption lines in the upper stratosphere have been included using the strategy of Fels (1979).

This longwave model represents a clear step forward from the use of Newtonian relaxation calculations to obtain the longwave cooling, as has been done in several 2-D models and general circulation models. Its results (heating rates, outgoing fluxes at the top of the atmosphere, surface fluxes) compare favorably with results from the body of current longwave parameterizations which participated in the ICRCCM analysis (WMO, 1984). Nevertheless, our initial analyses indicated that substantial errors remained in the heating rates calculated for the upper stratosphere (O(30%) above 2 mb), by increasing the critical pressure for Doppler broadening from 1 mb to 2 mb in the CO_2 parameterization of Chou and Peng (1983) and by extending the atmosphere used for IR Calculations above that used by the LLNL 2-D model, substantially better agreement with LBL calculations was obtained. Above the critical pressure, absorption by CO_2 is assumed to be independent of pressure for a given absorber amount. These comparison are shown in Figure 3, depicting vertical profiles of infrared cooling rates from both the original and modified IR parameterizations and for LBL calculations from Schwarzkopf and Fels (1987). Ongoing consideration is being given to methods to further improve the IR parameterizations used in the LLNL 2-D model.

Conclusions

We have implemented a series of state-of-the-art radiation transport submodels in our 1-D and 2-D chemical-transport models of the troposphere and stratosphere. These submodels provide us the capability of calculating accurate solar and infrared heating

rates. They are a firm basis for further radiation submodel development as well as for studying interactions between radiation and model dynamics under varying conditions of clear sky, clouds, and aerosols.

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Figure 1. Geometry of the radiative transfer problem for a single model layer. Layers are numbered sequentially from the bottom of the atmosphere to the top. Levels inherit the number of the layer immediately above them. Radiation components for the solar scattering problem are shown. The emergent diffuse radiation depends on both the incident diffuse radiation and the radiation scattered out of the direct beam.

Intercomparison of LLNL Shortwave Models

Figure 2. Intercomparison of O₃ heating rates from the new LLNL shortwave model, the old LLNL model, and the O₃ model of Harshvardhan et al. (1987) adapted to the LLNL grid. Clear sky conditions with the mid-latitude summer profiles of McClatchey et al. (1971) are assumed.

Figure 3. Comparison of vertical profiles of infrared cooling rates from the original and modified LLNL parameterized models with cooling rates from line-by-line calculations by Schwarzkopf and Fels.

Appendix A: Description of Cloud Distribution, Surface Albedo, and Surface Temperature as Used in the LLNL 2-D Model

Cloud Distribution

Clouds are given as zonal mean monthly fractional amounts with tops and bases defined by the 2-D model layers. The vertical and seasonal distribution of the clouds by latitude was taken from London (1957) for the Northern Hemisphere and Sasamori et al. (1972) for the Southern Hemisphere. The vertical and seasonal distribution of the cloud amount was proportionally scaled so that the randomly correlated total cloud amount would be identical to the monthly mean zonal total cloud amount of Berlyand et al. (1980).

Surface Albedo

Monthly and zonal mean surface albedos were taken from Kukla and Robinson (1980). These albedos were estimated from a number of sources of data for the time period 1 April 1974 to 31 March 1975. Kukla and Robinson consider them to be approximately representative of average conditions in the last decade.

Surface Temperature

Monthly and zonal mean surface air temperatures were taken from Oort (1983). As shown by Oort, the surface air temperatures are a good approximation in the mean to the ocean surface temperatures (within 2°C). However, we know that land surface temperatures can differ substantial from the surface air temperature, particularly during a cloudless diurnal cycle.

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Appendix B: Crude Estimates of The Vertical Distribution of Latent Heat Release for Use in Climate Models

Our aim is to estimate how much latent heat is released at various altitudes in the atmosphere, based on the local precipitation rate, surface temperature, and a simple, conceptual model. The simplifying assumptions needed to formulate the model are not unreasonable but are clearly crude approximations. The model is based on the idea that in clouds that produce precipitation, saturated air is lifted pseudoadiabatically from near the earth's surface to great heights where very little water vapor remains. The water vapor that condenses is assumed to fall out as precipitation. The amount of water vapor in the near surface air is approximately the amount of precipitation available to a cloud as that parcel of air ascends through the atmosphere. In our current model the surface air is assumed to be saturated at the observed temperature (Oort, 1983). Calculation of the pseudoadiabatic lapse rate provides information on what fraction of the vapor is removed at each altitude. The total latent heat released is proportional to the local precipitation rate which we obtain from observations (Jaeger, 1976; Potter and Gates, 1984). Thus the model gives the fraction of available water vapor condensed and removed at each altitude, and the observed precipitation rate determines the absolute amount removed. The latent heat released is proportional to the condensation rate.

This model may crudely mimic individual cumulus towers, although in naturally occurring convective clouds, the latent heat is carried upward away from its altitude of release by the motion within the cloud. The effects of entrainment on the condensation rate within the cloud might also modify the result significantly, as could the fact that the lifting condensation level is not often found right at the surface but some ways above it. It should be remembered, however, that the latent heat release estimate we will derive here is for use in global scale climate models, so that the formula we derive need only be valid on average, not for an individual cloud. In frontal cloud systems, the convection may take place slantwise along the frontal surface, but again averaging over several such systems in space and time will yield approximately the same relationship between condensation rate and altitude.

From the prognostic equation representing the conservation of water vapor, the following expression can be derived relating the rate of condensation in a cloud to the change in the saturation mixing ratio in a rising parcel of air:

$$S = \rho \omega \frac{\partial \mu_s}{\partial p} \quad (1)$$

where S is the rate of condensation ($\text{kg/m}^3/\text{s}$), ρ the air density, μ_s the saturation mixing ratio, $\omega = dp/dt$ the pressure coordinate analog to vertical velocity, and p the pressure. We have assumed here that the rate of change of mixing ratio following an air parcel is equal to the rate at which the saturation mixing ratio changes within the air parcel (i.e., we assume the air parcel is saturated). The saturation mixing ratio depends on pressure and temperature, but if we assume that nonadiabatic processes other than latent heat release are negligible, then the saturation mixing ratio can be expressed as a function of

pressure alone (given that the air parcel is initially saturated at some known pressure and temperature).

In our model we assume that air parcels that precipitate originate at the earth's surface where they are saturated at a zonally averaged temperature T . The temperature following the saturated air parcel can be found by solving the following two equations:

$$\mu_s = \frac{\epsilon e_o}{\rho} \exp \left[\frac{EL}{R} \left(\frac{1}{T_o} - \frac{1}{T} \right) \right] \quad (2)$$

$$\frac{\partial T}{\partial p} = \frac{RT}{C_p \bar{p}} \left[\frac{1 + \frac{L\mu_s}{RT}}{1 + \frac{EL^2\mu_s}{RT^2}} \right] \quad (3)$$

where e_o is the partial pressure of water vapor at reference pressure T_o , T the temperature, R the gas constant for dry air, L the latent heat of evaporation, ϵ the ratio of the molecular weight of water vapor to the molecular weight of dry air, and C_p the specific heat of air at constant pressure.

The first equation above comes from the Clausius-Clapeyron equation and relates the saturation mixing ratio to the temperature and pressure. The second equation is the well known expression for the saturated pseudoadiabatic lapse rate (see, for example, Hess (1959)), and it provides a relationship between temperature, saturation mixing ratio and pressure. In this set of equations, temperature and saturation mixing ratio can be considered the dependent variables. Given the surface temperature and pressure (currently obtained from observations processed by Oort, 1983), we can determine these variables at any other pressure, although it is not possible to find explicit analytical expressions, so solutions must be obtained numerically.

After determining the saturation mixing ratio as a function of pressure, we can proceed to calculate the condensation rate. At this point we make another simplifying assumption that in the rising cloud parcel ω is constant (i.e., the flow is nondivergent). If we can determine what this velocity is, then (1) can be used to calculate the condensation rate. (Note that the density is known from the ideal gas law.) Without a complete dynamical model of cloud processes it is impossible to find ω from first principles. We do, however, have some climatological observations for zonally averaged and monthly mean precipitation rates (Jaeger, 1976; Potter and Gates, 1984). We can use these to find the proportionality constant ω in (1). By integrating (1) through the depth of the troposphere (assuming the precipitating clouds extend to the tropopause), we find that

$$\omega = \frac{-\rho_w g P_w}{\mu_{ss} - \mu_{st}} \quad (4)$$

where P_w is the rate of precipitation (m/s), ρ_w the density of liquid water, g the acceleration of gravity, μ_{ss} and μ_{st} the saturation mixing ratio in the cloud parcel at the surface and when it arrives at the tropopause, respectively.

The final step is to calculate the atmospheric heating rate due to the latent heat release. Clearly the heating rate must be proportional to the condensation rate and it is easy to show that

$$Q = - \frac{LS}{\rho C_P}$$

where Q is the rate of heating (K/s).

In summary, S is found from (1), using the observed precipitation rate and (4) to give ω and using the observed or model calculated surface temperature and pressure with (2) and (3) to obtain $\partial\mu_s/\partial p$. The heating rate as a function of pressure is then calculated from (5).

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