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Positivity Preserving in Difference Schemes for the 2D Diffusive Transport of Atmospheric Gases

MELITTA FIEBIG-WITTMAACK*

*Departamento de Matemática, Facultad de Ciencias, Universidad de La Serena, Casilla 599, La Serena, Chile

AND

WOLFGANG BÖRSCH-SUPAN

Fachbereich Mathematik, Joh. Gutenberg-Universität, Postfach, 55099 Mainz, Germany

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Modelling the transport of tracer gases in the lower stratosphere leads to a diffusivity matrix with non-zero off-diagonal elements. This in turn implies non-positivity of conventional difference methods. Computations for regions with sharp gradients of mixing ratios therefore may show unrealistic small, even negative values. A modification of the difference scheme is proposed which is able to remove this feature and, in addition, to improve the accuracy of the calculations.

1. INTRODUCTION

In atmospheric models incorporating chemical reactions the global behavior of tracers is often studied by analyzing a two-dimensional zonally averaged model including the troposphere and the stratosphere (i.e., the two lower layers of the atmosphere up to about 60 km) and treating a very detailed chemistry (approximately 70 species and 100 reactions, e.g., see [3–5, 7, 9]). Zonal and monthly time averaging leads to linear transport equations with important eddy diffusion terms [7]. The diffusion is not only anisotropic, but in the stratosphere the diffusivity matrix also has non-zero off-diagonal elements due to the inclination of the main exchange direction (isolines of potential temperature) against the horizontal.

In mathematical terms the model is described by a parabolic system of differential equations given by

$$\frac{\partial X_i}{\partial t} = -\frac{\partial F_z}{\partial z} - \frac{1}{a\cos\varphi}\frac{\partial}{\partial\varphi}(\cos\varphi\, F_\varphi) + P_i - L_i X_i \qquad (1)$$

with

$$F_\varphi = -K_{yy}M\frac{1}{a}\frac{\partial\mu_i}{\partial\varphi} - K_{yz}M\frac{\partial\mu_i}{\partial z} + vX_i$$

$$F_z = -K_{yz}M\frac{1}{a}\frac{\partial\mu_i}{\partial\varphi} - K_{zz}M\frac{\partial\mu_i}{\partial z} + wX_i$$

$$i = 1, 2, 3, \ldots, m,$$

where X_i is the concentration (number of molecules per unit of volume) of the tracer or tracer family numbered by i. M is the air concentration, $\mu_i = X_i/M$ is the mixing ratio, φ is the latitude, z is the altitude, t is the time, a is the Earth's radius, $dy = ad\varphi$, F_φ is the meridional density, F_z is the vertical flux density, v is the averaged meridional advection velocity, w is the averaged vertical advection velocity, K_{yy}, K_{yz}, and K_{zz} are eddy diffusion coefficients, P_i is the number of molecules of the tracer produced by chemical processes (per unit time and volume) and L_i is the portion of the tracer destroyed by chemical processes (per unit time). P_i and L_i may depend on the concentrations, also of other tracers; they also may depend on space and time, e.g., via the sun's position in the sky. The other coefficients are space- and time-dependent.

It is well known that the condition

$$K_{yz}^2 < K_{yy}K_{zz}$$

implies positive definitness and positivity preserving of the diffusional terms (see [6]). This condition is always satisfied in the model; also the advective and the chemical terms preserve positivity of the solution.

Several investigations in air chemistry (e.g. [3, 7, 12]) solve

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Eqs. (1) numerically by applying a splitting method (as described in [11]), which consists in the alternate calculation of changes due to transport and to chemistry within one time step. Whereas for chemistry specially tailored methods are applied; the transport step uses Euler's forward method as time discretization and centered difference methods with staggered grids for concentrations and fluxes as spatial discretization. This rather simple approach is possible since, within the time step of 2 h, changes by transport are small and the diffusion terms ensure numerical stability for mesh sizes of 10° in latitude and approximately 2 km in altitude, even in the presence of moderate advection. The scheme is still in use, since it is economic with respect to computer time and therefore suitable for investigations with many varying parameters which allow the study of different scenarios (see [5, 9]).

Whereas most results produced by this implemented model are satisfactory, under some special circumstances, very low or even negative values of the concentrations of some species show up in areas with sharp gradients in a mixing ratio. We call this unrealistic phenomenon "numerical holes." Since the chemistry procedure is constructed in such a way that changes into negative values cannot occur, the transport routine must be responsible for the violation of positivity.

Positivity preserving and other properties of numerical methods for pure advection have been extensively discussed in the recent literature (see [15, 16, 2]). We found out, however, that this kind of numerical holes is due to the diffusive part of the equation, in particular to the off-diagonal coefficients in the diffusivity matrix. In fact, such a type of phenomenon was already described by Kershaw [10] with diffusion in Lagrangian coordinates, and by Pert [14] with diffusion perpendicular to the gradient in magnetic fields, and also with diffusion in non-orthogonal geometry. In [14] the appearance of negative values is prevented by applying an antidiffusive flux chosen to preserve a monotonicity constraint in the manner of Boris and Book [1]. This, however, reduces the order of accuracy of the implicit scheme. Instead, we propose a modification of the explicit scheme which retains spatial second-order consistency and regains positivity, although the latter under certain conditions only, similar to the method suggested by Gorenflo [8] with the Fokker–Planck equation. As long as advection is not too large, the simple scheme adopted for the advective part does not destroy positivity preserving for the whole scheme. Hence, from this point of view, it is not necessary to introduce the more sophisticated methods for advection with our modified diffusion scheme. But it is evident that such a combination is possible, at least on the basis of a "splitting" treatment.

Our modification is easily implementable within the framework of existing box model routines [4, 5, 7, 9], without notably increasing the computing time. It eliminated the appearance of unrealistic values and even diminished the local error in some important model situations.

2. STUDY OF A SIMPLIFIED CASE

In order to better understand the essential aspects of the hole problem, we consider a simplified model for just one tracer. We presume that the appearance of the hole is not directly linked to the rapid changes in diffusivity at the tropopause (i.e., the lower boundary of the stratosphere), nor to the advection. Thus, we take $\cos\varphi$, M, v, w, and the eddy diffusion coefficients K_{yy}, K_{yz}, and K_{zz} as constants, using cartesian coordinates $y = a\varphi$ and z. Equation (1) then becomes

$$\frac{\partial \mu}{\partial t} = -\frac{\partial f_y}{\partial y} - \frac{\partial f_z}{\partial z} + S, \tag{2}$$

where

$$f_y = -K_{yy}\frac{\partial \mu}{\partial y} - K_{yz}\frac{\partial \mu}{\partial z} + \mu v$$

$$f_z = -K_{yz}\frac{\partial \mu}{\partial y} - K_{zz}\frac{\partial \mu}{\partial z} + \mu w,$$

and the net source term

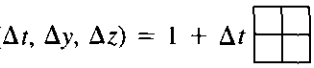
$$S = -\kappa\mu, \quad \kappa \text{ a given constant},$$

describes a very simplified chemistry. We consider a rectangular region representing a neighborhood of the hole and impose as boundary conditions purely vertical flux (i.e., $f_y = 0$) at the lateral boundaries, given vertical flux f_z at the upper boundary, and a Newton's type boundary condition at the lower boundary (i.e., $f_z + \nu\mu = 0$, with a positive constant ν, suitably chosen to prevent backflow from below).

All terms are discretized as in the original model [7] (see description in the Introduction). In order to maintain the accuracy order for the boundary conditions we introduce artificial outer points. For the simplified discrete model additional corner conditions are needed also. We use combinations of the flux conditions from the adjacent edges. In order to satisfy the discrete boundary conditions, for every time step, after calculating the new inner values of μ by an Euler's forward step we must solve a linear algebraic system of equations of essentially tridiagonal structure for the outer point values of μ.

For the sake of simplicity we first restrict ourselves to the purely diffusive part of the time evolution operator without boundary conditions. We represent the corresponding discretization operator $\mathfrak{C}$ which generates the μ distribution of the inner points of our rectangular region after a time step from the distribution before that step (see [13]) by

$$\mathfrak{C}(\Delta t, \Delta y, \Delta z) = 1 + \Delta t$$



with the classical scheme:

$$\boxplus = \bar{K}_{yy}\begin{bmatrix} \cdot & \cdot & \cdot \\ 1 & -2 & 1 \\ \cdot & \cdot & \cdot \end{bmatrix} + \bar{K}_{zz}\begin{bmatrix} \cdot & 1 & \cdot \\ \cdot & -2 & \cdot \\ \cdot & 1 & \cdot \end{bmatrix} + \frac{1}{2}\bar{K}_{yz}\begin{bmatrix} -1 & \cdot & 1 \\ \cdot & \cdot & \cdot \\ 1 & \cdot & -1 \end{bmatrix}, \tag{3}$$

with

$$\bar{K}_{yy} = K_{yy}/(\Delta y)^2, \quad \bar{K}_{zz} = K_{zz}/(\Delta z)^2, \quad \bar{K}_{yz} = K_{yz}/(\Delta y\, \Delta z).$$

Typical values of the diffusion coefficients in the neighborhood of the "hole" are

$$K_{yy} \approx 2.4 \times 10^{10}\ \mathrm{cm^2/s}, \quad K_{yz} \approx -1.3 \times 10^{7}\ \mathrm{cm^2/s},$$
$$K_{zz} \approx 1.1 \times 10^{4}\ \mathrm{cm^2/s}$$

(see [4, 7]).

In order to have simpler values to work with we renormalize the space coordinates such that the mesh spacing becomes $\Delta y = \Delta z = 1$ in the new coordinates. Simultaneously we renormalize the time scale such that the diffusion coefficients become of order 1; using a time unit of $\sim 400\ h = 1.44 \times 10^6$ s, this leads to the renormalized values of diffusion coefficients

$$K_{yy} = 3, \quad K_{yz} = -1, \quad K_{zz} = \tfrac{1}{2} \tag{4}$$

which we select as our reference model values. The stability condition is

$$\Delta t[K_{yy}/(\Delta y)^2 + K_{zz}/(\Delta z)^2] \leq \tfrac{1}{2}$$

(see [13]). With the time step of the original model renormalized to $\Delta t = 0.005$ and space steps $\Delta y = \Delta z = 1$, we are far inside the stability limits. For calculations near a stationary case a larger time step (e.g., $\Delta t = 0.1$) is possible.

We define a scheme to be positive (or positivity preserving), if any nonnegative distribution gives rise to a nonnegative distribution after one time step. If the diagonal diffusion coefficient $K_{yz} \neq 0$, then it follows from (3) that the scheme is not positive in this sense. Certain positive distributions with relatively sharp gradients (hence, considerable differences between neighboring mesh points) may cause negative values afterwards. We want to modify the scheme so that it changes to a positive one. It is easily seen, e.g., by derivation with Taylor's expansion, that consistent nine-point-formulae allow for some freedom even if accuracy order 2 is required (see Eq. (10) below, also [8], where this idea of generating a positive scheme is already applied to a different difference scheme).

We illustrate the procedure for f_y; this flux density is given for the middle of the right or left boundary of a mesh-cell by the following "difference-star" (i.e., coefficient scheme):

$$-\begin{bmatrix} \frac{K_{yz}}{4\Delta z} & \frac{K_{yz}}{4\Delta z} \\ -\frac{K_{yy}}{\Delta y} & \frac{K_{yy}}{\Delta y} \\ -\frac{K_{yz}}{4\Delta z} & -\frac{K_{yz}}{4\Delta z} \end{bmatrix} - \begin{bmatrix} -1 & 1 \\ 2 & -2 \\ -1 & 1 \end{bmatrix}\rho_y. \tag{5}$$

The first term corresponds to the classical discretization, whereas the second one constitutes the freedom left by the conditions of accuracy and is physically artificial with an arbitrarily valued factor ρ_y which we also write $\rho_y^*/\Delta z$. We use the factor to obtain non-negative coefficients for the diagonal values of the difference star (3), for example, by taking

$$\rho_y^* \geq \frac{|K_{yz}|}{4}.$$

Accuracy of order 2 is retained by this modification: Expressing the additional term by central differences and, in turn, differences by derivatives, one obtains

$$\frac{\delta_y \delta_z^2 \rho_y^*}{\Delta z} = \rho_y^*\, \Delta y\, \Delta z \frac{\partial}{\partial y}\frac{\partial^2}{\partial z^2} + \text{higher order terms},$$

which is of second order if ρ_y^* is fixed independent of Δy and Δz. We proceed similarly for the vertical flux density f_z, working with an arbitrarily chosen factor $\rho_z = \rho_y^*/\Delta y$. In this way, for (3) we acquire an additional term

$$\rho\begin{bmatrix} 1 & -2 & 1 \\ -2 & 4 & -2 \\ 1 & -2 & 1 \end{bmatrix}$$

with $\rho = \rho_y/\Delta y + \rho_z/\Delta z = (\rho_y^* + \rho_z^*)/\Delta y\, \Delta z$. We do not know of any physical interpretation of this artificial term which corresponds to the differential operator $\partial^4/\partial y^2\, \partial z^2$ multiplied by $(\rho_y^* + \rho_z^*)\, \Delta y\, \Delta z$, hence going to zero if the mesh-width vanishes.

If we want to get all the coefficients non-negative, ρ must satisfy the conditions

$$|\bar{K}_{yz}| \leq 2\rho \leq \min\{\bar{K}_{yy}, \bar{K}_{zz}\} \tag{6}$$

with

$$1 + \Delta t(-2\bar{K}_{yy} - 2\bar{K}_{zz} + 4\rho) \geq 0. \tag{7}$$

The latter is valid for every stable scheme if $\rho \geq 0$. However, the first two can be satisfied simultaneously only when the diagonal diffusion coefficient K_{yz} is sufficiently small: the matrix of the transformed diffusion coefficients,

$$\begin{bmatrix} \overline{K}_{yy} & \overline{K}_{yz} \\ \overline{K}_{yz} & \overline{K}_{zz} \end{bmatrix},$$

has to satisfy

$$|\overline{K}_{yz}| \leq \overline{K}_{yy}, \quad |\overline{K}_{yz}| \leq \overline{K}_{zz}, \tag{8}$$

which almost correspond to the conditions of weak diagonal dominance in [8] and which we shortly call "conditions of diagonal dominance." These conditions need not be met, even for a positive definite matrix of diffusion coefficients, i.e., with

$$\overline{K}_{yy} > 0, \quad \overline{K}_{zz} > 0, \quad \overline{K}_{yy}\overline{K}_{zz} > \overline{K}_{yz}^2,$$

as can be seen by our reference example (4). Observer, however, that in this case the condition (8) can be achieved by a suitable change in the ratio of the spatial stepsizes. With $\Delta y = 1$, $\frac{1}{3} \leq \Delta z \leq \frac{1}{2}$ we obtain

$$\overline{K}_{yy} = 3, \quad 2 \leq |\overline{K}_{yz}| \leq 3, \quad 2 \leq |\overline{K}_{zz}| \leq \tfrac{9}{2},$$

and

$$\tfrac{2}{3} \leq |\overline{K}_{yz}|/\overline{K}_{zz} \leq 1.$$

Our experience has shown that, even if the diagonal dominance condition (8) is violated and the step sizes are not changed, in many cases the introduction of such an additional ρ-term is useful, e.g., with $2\rho \equiv \Theta|K_{yz}|$, $0 < \Theta \leq 1$, negative values of the solutions disappear.

Our considerations may be extended to include constant advection. We then have additional terms

$$\frac{1}{2}\overline{v}\begin{bmatrix} \cdot & \cdot & \cdot \\ 1 & \cdot & -1 \\ \cdot & \cdot & \cdot \end{bmatrix} + \frac{1}{2}\overline{w}\begin{bmatrix} \cdot & -1 & \cdot \\ \cdot & \cdot & \cdot \\ \cdot & 1 & \cdot \end{bmatrix}$$

in (3), where $\overline{v} = v/\Delta y$, $\overline{w} = w/\Delta z$, and the condition (6) is to be replaced by

$$|\overline{K}_{yz}| \leq 2\rho \leq \min\{\overline{K}_{yy} - \tfrac{1}{2}|\overline{v}|, \overline{K}_{zz} - \tfrac{1}{2}|\overline{w}|\} \tag{9}$$

which further restricts the magnitude of the off-diagonal diffusion coefficient.

Besides the statement on the order of accuracy we cannot give details on the influence of the ρ-term on the discretization error with general μ distributions. However, with some exponential distributions which are important for our problem, we are able to show that the discretization error is even diminished by introducing the ρ-term.

Let us denote the right-hand side of Eq. (2) by G and its discretization by D, both calculated at the center of a mesh-cell. We define the local discretization error of the spatial operator by $R := D - G$. By Taylor's expansion we then obtain

$$\begin{aligned} R = {} & K_{yy}\left[\frac{(\Delta y)^2}{12}\frac{\partial^4\mu}{\partial y^4} + \cdots\right] + K_{zz}\left[\frac{(\Delta z)^2}{12}\frac{\partial^4\mu}{\partial z^4} + \cdots\right] \\ & + K_{yz}\left[\frac{(\Delta y)^2}{3}\frac{\partial^4\mu}{\partial y^3\,\partial z} + \frac{(\Delta z)^2}{3}\frac{\partial^4\mu}{\partial y\,\partial z^3} + \cdots\right] \\ & + \Theta|K_{yz}|\left[\frac{\Delta y\,\Delta z}{2}\frac{\partial^4\mu}{\partial y^2\,\partial z^2} + \cdots\right] \\ & - v\left[\frac{(\Delta y)^2}{6}\frac{\partial^3\mu}{\partial y^3} + \cdots\right] - w\left[\frac{(\Delta z)^2}{6}\frac{\partial^3\mu}{\partial z^3} + \cdots\right], \end{aligned} \tag{10}$$

where our additional ρ-term is given by the third line. Let us consider an exponential distribution $\mu = \mu_0 \exp\{\beta y + \gamma z\}$ with purely vertical diffusion flux. With $\alpha := K_{yz}/K_{yy}$ we obtain $K_2 := K_{zz} - \alpha^2 K_{yy} > 0$ because of the positive definiteness of the diffusion coefficient matrix. One has $K_2 \ll K_{zz}$ in the lower stratosphere, particularly near our observed "numerical hole." Purely vertical diffusion $f_y = 0$ requires

$$K_{yy}(\beta + \alpha\gamma)\mu = 0.$$

By substituting in (10) and neglecting higher order terms, after some calculations we obtain

$$\begin{aligned} R \approx {} & -\gamma^4 K_{yy}\left\{\frac{\alpha^2}{4}(\Delta z - |\alpha|\,\Delta y)^2 + (1 - \Theta)\frac{|\alpha|^3}{2}\Delta y\,\Delta z\right\} \\ & + \gamma^4\frac{K_2(\Delta z)^2}{12} + \frac{\gamma^3}{6}[v(\Delta y)^2\alpha^3 - w(\Delta z)^2]. \end{aligned}$$

We compare the factor in braces for the modified scheme with $\Theta = 1$ to the classical scheme ($\Theta = 0$). Considering the signs within this factor, we see that it always is reduced by a certain amount. From typical values of diffusion coefficients [4, 7] we find that near the numerical hole we have, after scaling, $\alpha = -\frac{1}{3}$ approximately. For $\Delta y = \Delta z = 1$ the factor in braces is then reduced to $\frac{2}{5}$ of its classical value. Similarly, with $\Delta y = 1$ and $\Delta z = \frac{1}{3}$, even a reduction to zero occurs.

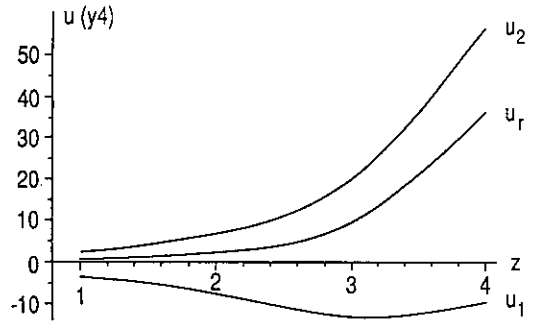


FIG. 1. Part of a vertical cross section of normalized solution of problem (2), for $K_{yy} = 3$, $K_{yz} = -1$, $K_{zz} = 0.5$, $\gamma = 0.4$, $\kappa = 0.16$, without advection (i.e., $v = w = 0$), $\gamma = 0.98$, $\nu = 0.163$, $u_0 = 1$, calculated by three different methods: u_1: $\Delta y = \Delta z = 1$, $\rho_y = \rho_z = 0$; u_r: $\Delta y = \Delta z = 1$, $\rho_y = \rho_z = 0.167$; u_2: $\Delta y = 1$, $\Delta z = \frac{1}{3}$, $\rho_y = \rho_z = 0$.

In Figs. 1 and 2 we show typical examples for numerical solutions in stationary cases, represented by a suitably normalized mixing ratio u along parts of a vertical line $y = y4$ in the middle of the model rectangular domain of width 5 and height 8 (corresponding to 50° meridional and 16 km vertical side lengths; the picture is restricted to the interesting part of the line, thus avoiding values of large magnitude outside). One sees that negative values showing up in the standard u_1 approximation (i.e., $\Delta y = \Delta z = 1$) disappear, if an additional ρ-term is introduced in the u_r approximation. The negative values also disappear for u_2 if a more accurate discretization is chosen (with $\Delta y = 1$, $\Delta z = \frac{1}{3}$), however, at the expense of higher computational costs. The stationary state is achieved by following the solution until a sufficiently large time.

3. APPLICATION TO THE GENERAL CASE

In the case of the complete system (1) with variable coefficients and a large number of tracer species, the diffusion part of the flux density F_φ discretization is also given by the two-term sum in (5) applied to the mixing ratio and multiplied by the air concentration M (the coefficients including ρ_y being taken at the midpoints of the lateral cell-boundaries). The advective part with the difference star

$$M \begin{bmatrix} 0 & 0 \\ \frac{v}{2} & \frac{v}{2} \\ 0 & 0 \end{bmatrix}$$

is added, using the arithmetic mean of mixing ratios in adjacent cells. In a similar way the vertical flux density is treated at the midpoints of upper and lower cell-boundaries. Observe that in combining the four flux approximations at the cell-boundaries into the discrete divergence operator one uses coefficients at different points which, in general, are not equal. Although, the diagonal coefficients of the nine-point star can be made nonnegative by choosing

$$\rho_y \geq \frac{1}{4}\left|\frac{K_{yz}}{\Delta z}\right|, \quad \rho_z \geq \frac{1}{4}\left|\frac{K_{yz}}{\Delta y}\right|$$

at the midpoints of each cell-boundary. On the other hand, for the axis-parallel coefficients diagonal dominance of the transformed diffusion coefficients matrix, even without advection terms, is no longer sufficient for positivity as in the constant coefficients case; this is also stated in [8], although there a different discretization is applied. As in [8], strict diagonal dominance (i.e., $|\overline{K}_{yz}| < \min\{\overline{K}_{yy}, \overline{K}_{zz}\}$) is sufficient for positivity if the step sizes are sufficiently small and the coefficients are continuous.

Since the discrete model from [7] is implemented with fixed step sizes and diagonal dominance is violated in some regions, particularly near the "hole," full positivity cannot be achieved by introducing our additional terms. Despite this we modified the program by our method with

$$\rho_y = \Theta_y \frac{1}{4}\left|\frac{K_{yz}}{\Delta z}\right|, \quad \rho_z = \Theta_z \frac{1}{4}\left|\frac{K_{yz}}{\Delta y}\right|,$$

where Θ_y and Θ_z are chosen suitably from the interval (0, 1].

Several calculations have been carried out with the full system (1) and detailed chemistry as in [7], with and without our modification. One example with $\Theta_y = \Theta_z = 1$ is shown in Fig. 3. In all cases we have obtained positive distributions when choosing $\Theta_y = \Theta_z = 1$. It turns out that our ρ-scheme acts specifically on the hole region of the ClO_x-family and has almost no influence in the other regions and on the other chemical families which present smooth gradients.

It is particularly recommended to apply this modified scheme

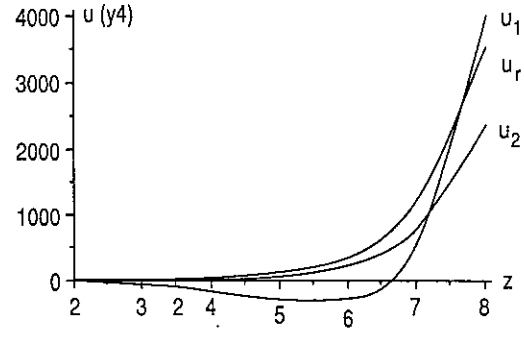


FIG. 2. Part of a vertical cross section of normalized solution of problem (2), for $K_{yy} = 3$, $K_{yz} = -1$, $K_{zz} = 0.5$, $\kappa = 0.08$, with vertical advection $v = 0$, $w = 0.083$, $\gamma = 0.985$, $\nu = 0.081$, $u_0 = 1$, calculated by three different methods: u_1: $\Delta y = \Delta z = 1$, $\rho_y = \rho_z = 0$; u_r: $\Delta y = \Delta z = 1$, $\rho_y = \rho_z = 0.417$; u_2: $\Delta y = 1$, $\Delta z = \frac{1}{3}$, $\rho_y = \rho_z = 0$.

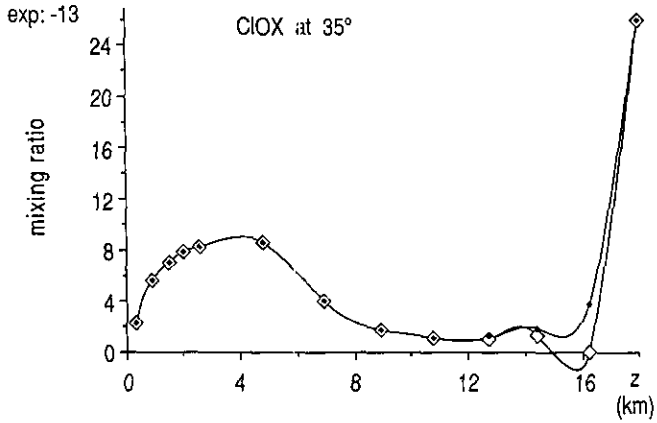


FIG. 3. We show part of the cross section at 35° N latitude for the calculated solution of (1), for the ClO_x-family in north winter for 10° steps in latitude, approximately 2-km steps in altitude (the first five steps are $\frac{1}{2}$ km), after calculation for 1 year in steps of 2 h, whereby: ◇: $\rho_y = \rho_z = 0$; ◆: $\rho_y = |K_{yz}|/(4\Delta z)$, $\rho_z = |K_{yz}|/(4\Delta y)$.

if diffusional transport and rapid chemical destruction of a family of species leads to sharp gradients in the corresponding mixing ratio in an area where diagonal diffusion is present.

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