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The terminator “toy”-chemistry test: a simple tool to assess errors in transport schemes

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Abstract

This test extends the evaluation of transport schemes from prescribed advection of inert scalars to reactive species. The test consists of transporting two interacting chemical species in the Nair and Lauritzen 2-D idealized flow field. The sources and sinks for these two species are given by a simple, but non-linear, “toy” chemistry which represents combination ($X+X \rightarrow X_2$) and dissociation ($X_2 \rightarrow X+X$). This chemistry mimics photolysis-driven conditions near the solar terminator, where strong gradients in the spatial distribution of the species develop near its edge. Despite the large spatial variations in each species the weighted sum $X_T = X + 2X_2$ should always be preserved (up to molecular diffusion). The terminator test demonstrates how well the advection/transport scheme preserves linear correlations. Chemistry-transport (physics-dynamics) coupling can also be studied with this test. Examples of the consequences of this test are shown for illustration.

1 Introduction

Tracer transport is a basic component of any atmospheric dynamical core. Typically, transport accuracy is evaluated in ideal tests before being developed further or implemented in full models. Several tests for 2-D passive and inert transport exist in the literature (Williamson et al., 1992; Nair and Machenhauer, 2002; Nair and Jablonowski, 2008; Nair and Lauritzen, 2010). To facilitate the intercomparison of transport operators under challenging flow conditions, Lauritzen et al. (2012) proposed a standard suite of tests that was exercised by a number of state-of-the-art schemes in Lauritzen et al. (2014). These tests evaluate each advection scheme’s ability to transport an inert tracer with respect to a wide range of diagnostics as well as the ability of each transport scheme to maintain non-linear tracer correlations between pairs of tracers (Lauritzen and Thuburn, 2012). While such evaluations provide useful information about the ability of each transport operator to advect inert scalars, these idealized tests do not shed light on how transport methods perform under forced conditions, e.g., how the method interacts with sub-grid scale processes.

Idealized chemical processes have readily available analytic expressions for the forcing terms. The implementation of these processes as sub-grid scale forcing involves “only” solving forced continuity equations rather than the full Navier Stokes, primitive or shallow water equations that add extra levels of complexity. Indeed several simplified systems, where two species interact non-linearly, have been developed and studied quite extensively in the literature. For example, the Lotka and Volterra equations (also known as *predator-prey equations*) that are a pair of first-order, differential equations describing the dynamics of biological systems in which two species interact, one as a predator and the other as prey. For a dynamical systems analysis of the Lotka and Volterra equations, e.g., see Chapter 4 in Prigogine (1981). The equations are the same for simple chemistry systems where each chemical species is transformed to the others. A more complicated system, but also consisting of just two independent variables (and two variables held constant), is the *Brusselator* system (Prigogine and Lefever, 1968) that allows for a rich set of solutions (Prigogine, 1981). The real Belousov-Zhabotinsky reaction has similar transient complex oscillations as found in the Brusselator system. Pudykiewicz (2006) coupled the Brusselator reactions to the advection–diffusion equations in a shallow water flow. The linearized system has analytic solutions (Turing, 1952) that can be used to assess the accuracy of the numerical solution to the differential equations. Pudykiewicz (2006, 2011) solved the full non-linear system, which is basically a forced advection–diffusion equation with flow prescribed from the shallow water solution, and examined the solutions qualitatively since the analytic solution is not known. Similar idealized systems for reactive species have been developed in the context of convective boundary layers (e.g., Kristensen et al., 2010).

The test we develop in this paper extends the Nair and Lauritzen (2010) test to two reactive species, adding one extra level of complexity while retaining the simplicity of analytic prescribed flow and known analytic solution. The inspiration for the idealized chemical reactions is photolysis-driven chemistry in which sunlight strongly influences the production and loss processes, creating very steep gradients in the individual tracer distributions near the terminator boundary (as observed for chlorine species and bromine in the stratosphere; see, e.g., Anderson et al., 1991; Salawitch et al., 2009; Brasseur and Solomon, 2005).

Hence these reaction coefficients lead to strong gradients coinciding with a “terminator-like” line (Lander and Hoskins, 1997). Another inspiration for this test is that the atomic concentration is conserved for each air parcel, while the molecular species react non-linearly with each other, e.g., total organic and inorganic chlorine in the stratosphere up to the scales where molecular diffusion matter (Edouard et al., 1996; Strahan et al., 2011; Prather and Jaffe, 1990). So by choosing the initial condition for two tracers so that the total amount (i.e. the total weighted mass of a chemical constituent X) is a constant throughout the domain then the atomic concentration should remain constant in space and time (as long as the chemistry exactly conserved the total of the constituents). This concept is used in this test case so that an analytic solution for the atomic concentration is readily available irrespective of the complexity of the flow and non-linearity of the chemical reactions.

The paper is organized as follows. In Sect. 2 the idealized chemistry, referred to as “toy chemistry”, is defined. An analysis in terms of steady-state solutions is presented. The transport operator is discussed in the context of linear tracer correlations in Sect. 3. The combination of the “toy” chemistry forcing with advection prescribed by the Nair and Lauritzen (2010) wind field (see Appendix A) defines the *terminator* test. The discrete terminator test is defined in Sect. 4. Section 5 shows example solutions from the Community Atmosphere Model (CAM) Finite-Volume dynamical core (CAM-FV; Lin, 2004) and the CAM Spectral Elements dynamical core (CAM-SE; Dennis et al., 2012). In particular, we show that the terminator test exacerbates errors associated with the preservation of linear relations and limiters as well as highlights differences in chemistry-transport (physics-dynamics) coupling approaches. The summary and conclusions are in Sect. 6.

2 Toy chemistry

In this section, we use the nomenclature X to describe the number density (number of molecules of compound X divided by the number of molecules of air, see Adreus et al., 1987). A molecule composed of two atoms X is written as X_2 . The number density associated with the total number of atoms X (in X and X_2) is denoted X_T .

The non-linear “toy” chemistry equations for X_2 and X are



where k_1 and k_2 are the reaction rates of the production pathways for X and X_2 , respectively. The reactions are designed to conserve the total number of X atoms

$$X_T = X + 2X_2. \quad (3)$$

The kinetic equations corresponding to the above system (Eqs. 2 and 1) are given by

$$\frac{dX}{dt} = 2k_1X_2 - 2k_2XX, \quad (4)$$

$$\frac{dX_2}{dt} = -k_1X_2 + k_2XX, \quad (5)$$

where d/dt is the material (or total) derivative $d/dt = \partial/\partial t + \mathbf{v} \cdot \nabla$ and \mathbf{v} is the wind vector. It is easily verified that the weighted sum of X and X_2 is conserved along characteristics of the flow

$$\frac{dX_T}{dt} = \frac{d}{dt}[X + 2X_2] = 0. \quad (6)$$

If the initial condition for X_T is constant (as we assume here), X_T is not a function of time and is therefore equal to its initial value.

$$\begin{aligned} X_T &= X(t) + 2X_2(t), \\ &= X(0) + 2X_2(0), \end{aligned} \quad (7)$$

and hence,

$$X_2(t) = \frac{1}{2}(X_T - X(t)). \quad (8)$$

The reaction coefficient, k_1 , represents the photolytic breaking of molecule X_2 and can be represented as the cosine of the solar zenith angle for when the sun's zenith is at (λ_c, θ_c) (see Fig. 1). The reaction coefficient, k_2 , represents the recombination and is assumed constant over the globe.

$$k_1(\lambda, \theta) = \max[0, \sin \theta \sin \theta_c + \cos \theta \cos \theta_c \cos(\lambda - \lambda_c)], \quad (9)$$

$$k_2(\lambda, \theta) = 1, \quad (10)$$

where λ and θ are longitude and latitude, respectively, and (λ_c, θ_c) are chosen as (20° N, 300° E) to align with the flow field. The terminator is the continuous boundary between day and night regions. These reaction rates produce very steep gradients in the X species near the terminator. This setup is of direct application to the real atmosphere as the total chlorine in the stratosphere is conserved (except for molecular diffusion), while photolysis and chemical reactions partition the various components and lead to narrow gradients across the terminator.

An analytic steady-state solution of the chemical concentrations for the condition of no flow is derived in Appendix B. The initial condition is specified as being the steady-state solution under no flow. Reaction rates are quite rapid compared to the model time-steps as shown in Appendix C.

3 Transport operator and correlations

Let \mathcal{T} be the discrete transport operator that advances, in time, the numerical solution to the passive and inert continuity equation for species X and X_2

$$\frac{D\phi}{Dt} = 0, \quad \phi = \tilde{X}, \tilde{X}_2, \quad (11)$$

at grid point or grid cell i . In this equation, \tilde{X} refers to the volume mixing ratio of X (Adrews et al., 1987), which is related to the number density of X through scaling by the fixed ratio

of the atomic weight of X to the molecular weight of dry air.

$$\phi_i^{n+1} = \phi_i^n + \Delta t_{\text{tracer}} \mathcal{T}(\phi_j^n), \quad j \in \mathcal{H}, \quad \phi = \tilde{X}, \tilde{X}_2 \quad (12)$$

where n is the time-level index, Δt_{tracer} time-step for the transport operator, and \mathcal{H} is the set of indices defining the stencil required by \mathcal{T} to update ϕ_i^n . Note that the transport operator may not solve the prognostic equation for ϕ in advective form as used in Eqs. (4) and (5). For example, it is common practice for finite-volume schemes to base the discretization on a flux-form formulation of the continuity equation (here written without forcing terms)

$$\frac{\partial(\rho\phi)}{\partial t} = -\nabla \cdot (\mathbf{v}\rho\phi), \quad (13)$$

where ρ is air density. To deduce the mixing ratio from Eq. (13) one needs to solve the continuity equation for air. For a non-divergent wind field and an initial condition of ρ that is constant, the exact solution for ρ is that it remains constant in time and space. For the terminator test, in which we use a non-divergent flow field and constant initial condition for ρ (if applicable), it has been found crucial to solve for ρ rather than prescribing the analytic solution for ρ . Usually a transport scheme using $\rho\phi$ as a prognostic variable will not preserve a $\rho\phi = \text{constant}$ initial condition whereas it will preserve a constant mixing ratio. So if ρ is analytically prescribed ϕ will not be preserved in areas where it would otherwise be constant. Such errors can be exacerbated by the terminator chemistry. For a fuller discussion of tracer-air coupling see, e.g., Lauritzen et al. (2011) and Nair and Lauritzen (2010).

For the theoretical discussion it is convenient to define the property *semi-linear*: A transport operator \mathcal{T} is semi-linear if it satisfies

$$\mathcal{T}(a\phi_i + b) = a\mathcal{T}(\phi_i) + b\mathcal{T}(1) = a\mathcal{T}(\phi_i) + b, \quad (14)$$

for any constants a and b (Lin and Rood, 1996; Thuburn and McIntyre, 1997). A semi-linear transport operator preserves linear correlation between two trace species. Note that the semi-linear property subsumes that the transport operator preserves a constant mixing

ratio

$$\mathcal{T}(b) = b. \quad (15)$$

Since X_T is simply the weighted sum of just two species, X_T will be conserved in the numerical model if \mathcal{T} is semi-linear. The semi-linear property, however, does not imply that a weighted (linear) sum of more than 2 species is conserved (Lauritzen and Thuburn, 2012). The chemical reactions (Equations 1 and 2), even in discrete form, will preserve the sum of species. Consequently, a semi-linear transport operator combined with the terminator chemistry will produce no error in X_T .

Several transport operators \mathcal{T} in the literature are semi-linear when limiter/filters are not applied. For example, Lin and Rood (1996) show that their scheme, based on the widely used Piecewise-Parabolic Method (PPM; Colella and Woodward, 1984) for reconstructing sub-grid scale tracer fields, preserves linear correlations. The CSLAM scheme (Lauritzen et al., 2010), also based on polynomials, preserves linear correlations (see proof in Appendix A of Harris et al., 2010). An example of a scheme that is not semi-linear is the transport operator based on rational functions described in Xiao et al. (2002) due to the non-linearity of the reconstruction function.

Typically, transport operators are not applied in their unlimited versions in full models. Shape-preserving filters are applied to ensure physically realizable solutions such as the prevention of negative mixing ratios or unphysical oscillations in the numerical solutions (e.g. Durran, 2010). The filter method/algorithm depends on the advection scheme formulation and discretization. Finite-volume discretizations that are based on cell-average prognostic variables ($\rho\phi$) usually make use of either sub-grid-cell reconstruction function filters, e.g. van Leer type 1-D limiters (Lin et al., 1994), or flux-limiting methods such as flux-corrected transport (Zalesak, 1979). The reconstruction filter can be applied for schemes that are based on Lagrangian or Eulerian finite-volume discretizations, where the integration is based on swept areas (in either dimensionally split 1D operators such as Lin and Rood, 1996, or fully 2D) that span the domain without gaps or overlaps. Examples of reconstruction function

filters are Colella and Woodward (1984) and Lin and Rood (1996). Limiting through flux-correction, where low-order shape-preserving fluxes are optimally blended with higher-order fluxes, can only be applied in flux-form schemes. For discretizations based on a non-conservative form (advective form), where the prognostic variables are mixing ratio ϕ rather than $\rho\phi$, tracer mass conservation is not inherent and is usually restored *a posteriori* with *ad hoc* methods (e.g., Priestley, 1993; Gravel and Staniforth, 1994). Since the mass-restoration algorithm may alter ϕ , the mass-fixer and shape-preservation algorithms are intrinsically related. Usually this problem is solved using optimization/variational methods (e.g., White and Dongarra, 2011). For methods where the prognostic variables are represented with series-expansions (e.g., Galerkin methods), shape-preservation can also be enforced with optimization methods (e.g., Guba et al., 2014).

Shape-preserving filters may render an otherwise semi-linear transport operator non-semi-linear. Some limiters, however, are semi-linear. For example, van Leer type 1-D limiters (Lin et al., 1994) preserve linear correlations (Lin and Rood, 1996). Flux-corrected transport limiters with and without selective limiting preserves linear correlations (Blossey and Durran, 2008; Harris et al., 2010). The limiter by Barth and Jespersen (1989), that scales the reconstruction functions so that it is within the range of the surrounding cell average values, preserves linear correlations (Harris et al., 2010). Positive definite limiters that insure positivity-preservation and “clipping” algorithms that simply remove negative values (see, e.g., Skamarock and Weisman, 2009, for applications in a weather forecast model), are certain to violate linear correlations as the filter only affects the species that is about to become negative and not the other species. Note that “clipping” may occur in the physical parameterization package. *A posteriori* filters (e.g., optimization-based shape-preserving filters) may or may not be semi-linear and the details of the implementation can effect the semi-linearity (e.g., iteration thresholds, logic in the code).

We note that instead of advecting each species separately by solving the advection equations in (11) or (13) with $\phi = X, X_2$, one may also choose to advect the sum X_T and one of the species, e.g. X , and then diagnose the remaining species, X_2 , from (8). If the transport operator conserves a constant, which many transport operators do, then the constant sum

X_T is trivially conserved if chosen as a prognostic variable. This approach is commonly used in chemistry transport models for some families of species. E.g., Douglass *et al.* (e.g., 2004) advected the sum of total inorganic chlorine and bromine to avoid spurious maxima and minima in their distributions in the stratosphere. Idealized tests to evaluate how well families of species that add up to a constant are preserved see, e.g., Lauritzen and Thuburn (2012).

4 Discrete terminator test

Coupling the chemistry parameterization with advection can be done in multiple ways. A common approach in weather/climate modeling is to update the species evolution in time incrementally by first updating the mixing ratios with respect to sub-grid-scale forcings (chemistry) and then apply the transport operator based on the chemistry-updated state (or in reverse order). Since the computation of the sub-grid-scale tendencies in full models is computationally costly, the dynamical core (in this case the transport scheme) is usually subcycled with respect to chemistry. For fast chemistry this may be reversed.

A model will operate with a chemistry (physics) time-step Δt_{chem} , a tracer time-step Δt_{tracer} and a chemistry-transport (physics-dynamics) coupling time-step Δt_{cpl} . For example, in the default CAM-SE setup $\Delta t_{tracer} = 300s$, $\Delta t_{chem} = 1800s$ and $\Delta t_{cpl} = 600s$. Hence the chemistry tendencies, F_X and F_{X_2} , are computed every 30 minutes, and the species are updated every 600s with the chemistry tendencies. A detailed description of the CAM-SE implementation of physics-dynamics coupling in terms of its namelist variables is given in Appendix E.

It is, of course, up to the model developer to choose which coupling method and time-step to use. To facilitate comparison the model developer is encouraged to use the analytically computed forcing terms F_X and F_{X_2} given in Appendix F, and to use a chemistry (physics) time-step of $\Delta t_{chem} = 1800s$. The initial conditions are given by the steady state asymptotic solutions Eqs. (B14) and (B15) with a mixing ratio of $X_T = 4 \times 10^{-6}$ (Fortran code for the initial conditions is given in Appendix G and in the Supplement).

For simplicity the velocity field for the transport operator \mathcal{T} is prescribed. We use the deformational flow of Nair and Lauritzen (Case-2; 2010) that was also used in the standard test case suite of Lauritzen et al. (2012, 2014). For completeness the components of the non-divergent velocity vector $\mathbf{V}(\lambda, \theta, t)$ and the stream function are repeated in Appendix A. The test is run for 12 days (or 5 non-dimensional time-units) exactly as prescribed in Nair and Lauritzen (2010). Note that the test case methodology can be applied in any velocity field including a full 3-D dynamical core.

5 Results

It is the purpose of this section to show exploratory terminator test results. An in-depth analysis of why the limiters do not preserve linear relations (and the derivation of possible remedies) is up to the scheme developers.

5.1 Model setup

Terminator test results are shown for two dynamical cores (transport schemes) available in the CAM: CAM-FV (Lin, 2004) and CAM-SE (Dennis et al., 2012) that are documented within the framework of CAM in Neale et al. (2010). The transport scheme in CAM-FV is the widely used finite-volume scheme of Lin and Rood (1996). CAM-SE performs tracer transport using the spectral element method based on degree three polynomials. Further details on CAM-SE are given in Appendix H.

As discussed in detail in Nair and Lauritzen (2010) and briefly in Sect. 3, care must be taken in the handling of tracer mixing ratio and tracer mass coupling for schemes that prognose tracer mass. In general the transport scheme will not preserve a constant tracer density field ($\rho\phi = \text{constant}$) since the discrete divergence operator is non-zero despite the analytical wind field being non-divergent (zero divergence). However, the scheme will preserve a constant mixing ratio if q is recovered from $\rho\phi$ by dividing the tracer density with the prognosed air density ρ . If one does not prognose ρ and simply specifies the analytic solution ($\rho = \text{constant}$), a constant mixing ratio will not be preserved.

For all simulations the chemistry (physics) time-step is $\Delta t_{chem} = 1800$ s. The horizontal resolution is approximately 1° : For CAM-FV that is the 0.9×1.25 configuration (192 latitudes and 288 longitudes) and for CAM-SE it is the NE30NP4 configuration in which there are 30×30 elements on each cubed-sphere panel and 4×4 Gauss–Lobatto–Legendre (GLL) quadrature points in each element. The tracer time step Δt_{tracer} is 900 s for CAM-FV and 300 s for CAM-SE. During the tracer transport scheme time-step the analytic winds of Nair and Lauritzen (2010) are held constant following the CAM-Chem setup (Lamarque et al., 2012). Unless explicitly stated otherwise, the coupling time-step for CAM-SE is $\Delta t_{cpl} = 600$ and $\Delta t_{cpl} = 1800$ for CAM-FV. For details on CAM chemistry-transport coupling see Appendix E.

The sample results shown next are divided into four sections. First of all, baseline results for CAM-FV and CAM-SE using their default configurations. Next, results from experiments varying the limiter in CAM-SE are presented. Then the consequences of using different chemistry-transport (physics-dynamics) coupling methods (in CAM-SE) are discussed. Lastly, the results are quantified.

5.2 Default CAM-FV and default CAM-SE results

Figure 3 shows the distributions X_T after 1 and 6 simulated days for CAM-FV and CAM-SE. Ideally X_T should be conserved. Both CAM-FV and CAM-SE show deviations from constancy in X_T (note that the color-scale on the Figures is not linear). The errors in X_T are produced at the terminator when the limiter is most challenged. After the errors are introduced they propagate away from the terminator following Lagrangian trajectories of the prescribed flow. This is most visible for CAM-SE at day 6 (see Fig. 3 and/or animations in Supplement).

CAM-FV transport is based on the dimensionally split Lin and Rood (1996) scheme. The scheme produces errors in X_T since the limiter used in CAM-FV (described in Appendix B of Lin, 2004) does not strictly conserve linear relations. The errors appear to be largest when the flow is aligned with the terminator at a 45° angle (see animation in Supplement). In that situation the dimensionally split approach is most challenged; the shape-preserving limiter

is not strictly shape-preserving in the cross direction since the one-dimensional limiters are only applied in the coordinate directions (Lauritzen, 2007).

CAM-SE does not preserve linear relations either and the errors in X_T are about an order of magnitude larger than CAM-FV. The CAM-SE limiter is optimization-based (using Least-squares) and guarantees no under- or over-shoots at the element level while maintaining mass-conservation at the element level (Guba et al., 2014). While the optimization-based limiter preserves linear relations with exact arithmetic, its present implementation in CAM-SE does not lead to such preservation (most likely due to iteration thresholds, if-statements, etc. that can be non-linear).

To further understand this behavior we have performed some tests (not shown) turning the chemistry off and advecting linearly correlated Cosine hills and linearly correlated step-functions. The Cosine hills are C^0 continuous (the function is continuous but its derivatives are not) and the limiter exactly preserves the linear relationship. For the step-functions, which are discontinuous distributions, the correlation preservation is only maintained up to $O(10^{-8})$ due to an $O(10^{-8})$ overshoot in one of the tracers. So the advection operator introduces an $O(10^{-8})$ error. The terminator chemistry constantly enforces a discontinuity in the distributions and in combination with the CAM-SE limiter, strong error growth is produced. It is beyond the scope of this paper to trace down exactly where in the implementation this error is introduced and to find a remedy. The terminator test is designed to enable scheme developers to test their scheme in setups that are directly relevant to some of the issues seen in chemistry application. In this particular case, the terminator test clearly exacerbates small errors in correlation preservation that may be easily overlooked in inert transport testing (such as the tests in Lauritzen et al., 2012).

5.3 CAM-SE: limiter experiments

In addition to the quasi-monotone mass-conservative limiter used by default in CAM-SE, the model has options for performing tracer advection without any limiter and with a positive definite limiter. Results for terminator test runs using those configurations are shown on Fig. 4. As expected the unlimited version of the CAM-SE transport exactly preserves linear

relations i.e. X_T is conserved to machine precision. By looking at cross sections of the individual distributions of X and X_2 on Fig. 5, it is immediately apparent (and expected) that Gibbs phenomena manifests itself near the terminator when no limiter is used. The stability analysis discussed in Appendix D and illustrated in Figs. B1 and B2 indicates that the terminator chemistry will drive a negative mixing ratio even more negative. From the experiments, however, the amplitude of the spurious oscillations near the terminator remain nearly constant in time. In other words, the instability associated with negative mixing ratios in the terminator chemistry is weak in our present setup.

When using a positive definite limiter, Gibbs phenomena is eliminated near the base of the terminator but not near the maximum. This obviously violates linear relations and produces large errors in X_T . Similar results are expected from mass-filling algorithms in which negative values are simply set to zero. This emphasizes the importance of using carefully designed limiters in transport schemes for applications in which preservation of linear pre-existing relations is important, e.g., chemistry applications (for a fuller discussion see, e.g., Lauritzen and Thuburn, 2012).

5.4 CAM-SE: Chemistry-tracer (physics-dynamics) coupling experiments

As explained in Sect. 4 the tracer transport (dynamics) and chemistry (physics) can be coupled in various ways. Here we discuss results based on different coupling time-steps, Δt_{cpl} , in CAM-SE.

On Fig. 6 X_T is shown using $\Delta t_{cpl} = 1800s$, $\Delta t_{cpl} = 600s$, and $\Delta t_{cpl} = 300s$, respectively¹. In all experiments the tracer time-step and chemistry time-step are held fixed: $\Delta t_{tracer} = 300s$ and $\Delta t_{chem} = 1800s$, respectively. The experiments evaluate the sensitivity to coupling time-step size, in other words, how often the species are adjusted with chemistry tendencies.

Near the western edge of the terminator (located at approximately 130° W on Fig. 5) where the gradients are steepest, the errors in X_T are largest for $\Delta t_{cpl} = 1800s$. The chem-

¹ in terms of CAM-SE namelist these configurations correspond to (a) `fctype=1, nsplit=1, rsplit=6`, (b) `fctype=0, nsplit=2, rsplit=3`, and (c) `fctype=0, nsplit=6, rsplit=1`

istry adjustments that steepen the gradients are largest at the western edge and consequently produces states that challenges the limiters more. When the chemistry tendency is added gradually throughout the tracer transport the errors are reduced as Δt_{cpl} is decreased.

At the eastern edge of the terminator (located at approximately 30° E on Fig. 5) the gradients are less steep compared to the western edge. In fact, the location of the gradient near the eastern edge propagates (see animation in Supplement) whereas the gradients at the western edge of the terminator are static in space. The chemistry tendencies in this area are not stationary in space and are weaker so the transport signal is larger. This means that for any given point in the eastern area, the state used for computing the chemistry tendencies changes during the tracer subcycling. As a result the gradients will have propagated during the transport step but the chemistry tendencies will steepen gradients in the “old” location. This “inconsistency” is present with $\Delta t_{cpl} \neq \Delta t_{chem}$. For $\Delta t_{cpl} = 1800$ s the chemistry update is based on the “correct” in time state. The temporal inconsistency in the state used for computing chemistry tendencies for $\Delta t_{cpl} = 600$ s and $\Delta t_{cpl} = 300$ s produce an increase in errors near the eastern edge of the terminator compared to $\Delta t_{cpl} = 1800$ s.

Physical parameterization packages may contain code that sets negative mixing ratios to zero. Or similarly there may be code that prevent tendencies to be added to the state if it is zero or negative. The terminator test may be a useful tool to diagnose such alternations in large complicated codes.

5.5 Quantification of X_T errors

To quantify the errors introduced in the terminator test, we suggest to compute standard error norms for X_T . The global normalized error norms used are $\ell_2(t)$ and $\ell_\infty(t)$ (e.g.,

Williamson et al., 1992):

$$\ell_2(t) = \sqrt{\frac{I[(X_T(t) - X_T(0))^2]}{I[(X_T(0))^2]}}, \quad (16)$$

$$\ell_\infty = \frac{\max_{\forall \lambda, \theta} |X_T(t) - X_T(0)|}{\max_{\forall \lambda, \theta} |X_T(0)|}, \quad (17)$$

where $X_T(0) = 4 \times 10^{-6}$ is the globally-uniform initial condition and the global integral I is defined as follows,

$$I(\phi) = \frac{1}{4\pi} \int_0^{2\pi} \int_{-\pi/2}^{\pi/2} \phi(\lambda, \theta, t) \cos\theta d\lambda d\theta. \quad (18)$$

As a reference we show the time-evolution of $\ell_2(t)$ and $\ell_\infty(t)$ for CAM-FV and CAM-SE on Fig. 7.

6 Conclusions

A simple idealized “toy” chemistry test case is defined. It consists of advecting two reactive species (X and X_2) in the Nair and Lauritzen (2010) flow field. The simplified non-linear chemistry creates strong gradients in the species similar to what is observed for photolysis-driven species in the stratosphere. The forcing terms for the continuity equations for X and X_2 are computed analytically over one time-step (assuming no advection) and Fortran codes for computing the forcing terms are provided as Supplement. Hence, model developers who have already setup the standard test case suite of Lauritzen et al. (2012) can, with modest effort, setup the terminator test by adding the forcing terms to their codes. As in the test case of Nair and Lauritzen (2010) this forced advection problem has an analytic solution.

The “toy” chemistry, by design, does not disrupt pre-existing linear relations between the species. So the only source of error is from the transport scheme and/or the chemistry-transport (physics-dynamics) coupling. The terminator test is setup so that X_T is a constant so any deviation from constancy is an error in the preservation of linear correlations. Many transport schemes preserve linear relations when no shape-preserving limiter/filter is applied and are therefore not challenged with respect to conserving X_T . However, many shape-preserving limiters/filters render the transport scheme non-conserving with respect to X_T . While preservation of linear correlations can indeed be verified in inert advection setup, the terminator chemistry exacerbates the non-conservation problem through the constant forcing that creates very steep gradients. It is demonstrated in this paper that the terminator test is useful for challenging the limiters with strong grid-scale forcing. In particular, it is shown that positive definite limiters severely disrupt linear correlations near the terminator.

In addition, the terminator test assesses the accuracy of chemistry-tracer (physics-dynamics) coupling methods in an idealized setup. Different coupling methods (such as those available in CAM-SE) lead to different distributions of X_T . Also, chemistry-transport (physics-dynamics) coupling layer or the physical parameterization package may contain code that sets negative mixing ratios to zero and/or contain if-statements that prevent tendencies to be added to the state if it is zero or negative. The terminator test may be a useful tool to diagnose such alternations in large complicated codes.

The terminator test is easily accessible to advection scheme developers from an implementation perspective since the software engineering associated with extensive parameterization packages is avoided. The test forces the model developer to consider how their scheme is coupled to sub-grid scale parameterizations and, if solving the continuity equation in flux-form, forces the developer to consider tracer-air mass coupling. Also, the idealized forcing proposed here has an analytic formulation and the continuous set of forced transport equations have, contrary to the Brusselator forcing, an analytic solution for the weighted sum of the correlated species irrespective of the flow field.

We encourage dynamical core developers to implement the toy chemistry in their test suite as it has the potential to identify tracer transport issues that standard tests (with unreactive/inert tracers) would not generate.

Appendix A: Idealized flow field

In the terminator test we use the deformational flow of Nair and Lauritzen (Case-2; 2010). The components of the non-divergent velocity vector $\mathbf{V}(\lambda, \theta, t)$ and the stream function

$$u = -\frac{\partial\psi}{\partial\theta}, \quad (\text{A1})$$

$$v = \frac{1}{\cos\theta} \frac{\partial\psi}{\partial\lambda}, \quad (\text{A2})$$

are given by

$$u(\lambda, \theta, t) = \frac{10R}{T} \sin^2(\lambda') \sin(2\theta) \cos\left(\frac{\pi t}{T}\right) + \frac{2\pi R}{T} \cos(\theta) \quad (\text{A3})$$

$$v(\lambda, \theta, t) = \frac{10R}{T} \sin(2\lambda') \cos(\theta) \cos\left(\frac{\pi t}{T}\right), \quad (\text{A4})$$

$$\psi(\lambda, \theta, t) = \frac{10R}{T} \sin^2(\lambda') \cos^2(\theta) \cos\left(\frac{\pi t}{T}\right) - \frac{2\pi R}{T} \sin(\theta), \quad (\text{A5})$$

respectively, where λ is longitude, θ is latitude, t is time and the underlying solid-body rotation is added through the translation $\lambda' = \lambda - 2\pi t/T$. The period of the flow is $T = 12$ days and $R = 6.3172 \times 10^6$ m (in non-dimensional units $T = 5$ and $R = 1$). Schemes based on characteristics, e.g. Lagrangian and semi-Lagrangian schemes, may use the semi-analytic

trajectory formulas given in (Nair and Lauritzen, 2010). Note that it is not necessary to use an analytic flow field for this test case setup. In fact, one may use winds from a weather or climate model simulation.

Appendix B: Analytic solution for no flow

To gain more insight into the toy chemistry (and to formulate “spun-up” initial conditions), it is useful to consider the special case of no flow. For $v = 0$ the prognostic equations for X and X_2 (Eqs. 4 and 5, respectively) can be solved analytically. Assume the reaction rates are positive (and non-zero for k_2),

$$k_1 \geq 0, \tag{B1}$$

$$k_2 > 0 \tag{B2}$$

and the mixing ratios are non-negative,

$$X(0) \geq 0, \tag{B3}$$

$$X_2(0) \geq 0. \tag{B4}$$

From the kinetic Eqs. (4) and (5) above, as well as the conservation Eq. (3) we can write

$$\frac{dX}{dt} = k_1(X_T - X) - 2k_2XX \tag{B5}$$

For (algebraic) convenience, define the quantities,

$$r = \frac{k_1}{4k_2}, \tag{B6}$$

$$D = \sqrt{r^2 + 2rX_T}, \tag{B7}$$

$$E(t) = e^{-4k_2Dt}. \tag{B8}$$

Completing the square on the right-hand side leads to the expression

$$\frac{dX}{dt} = -2k_2[(X+r)^2 - D^2]. \quad (\text{B9})$$

The right-hand side can be factored, and the following partial fraction expansion can be constructed:

$$\frac{dX}{(X+r)-D} - \frac{dX}{(X+r)+D} = -4Dk_2dt \quad (\text{B10})$$

Integration of each of these terms from time $t = 0$ to t , yields the expression

$$\ln \left(\frac{(X(t)+r-D)(X(0)+r+D)}{(X(t)+r+D)(X(0)+r-D)} \right) = -4Dk_2t, \quad (\text{B11})$$

leading to the solutions Eq. (B12). The analytic solution for $X(t)$ is

$$X(t) = \begin{cases} D \left(\frac{(X(0)+r)(1+E(t))+D(1-E(t))}{(X(0)+r)(1-E(t))+D(1+E(t))} \right) - r & \text{if } r > 0, \\ \frac{X(0)}{1+2k_2tX(0)} & \text{if } r = 0. \end{cases} \quad (\text{B12})$$

$$X_2(t) = \frac{1}{2}(X_T - X(t)) \quad (\text{B13})$$

For long times, $X(t)$ and $X_2(t)$ converge to the steady state solutions,

$$\lim_{t \rightarrow \infty} X(t) = D - r, \quad (\text{B14})$$

$$\lim_{t \rightarrow \infty} X_2(t) = \frac{1}{2}(X_T - D + r), \quad (\text{B15})$$

and are shown on Fig. 2. The steady state solutions are specified as initial conditions for the terminator test case. For a stability analysis of the terminator ‘‘toy’’ chemistry see Appendix D.

Appendix C: Local Linear Convergence

If the flow is slow compared to the rate at which the chemistry returns to equilibrium, then the concentrations will stay near the steady-state solution derived in Appendix B. Under these conditions, the rate of convergence to the steady-state solution can be computed. For convenience, define X_s as the right-hand side of Eq (B14). From Eq. (B5), a perturbation from steady state, $\epsilon(t) = X(t) - X_s$, can be seen to solve the equation,

$$\frac{d\epsilon(t)}{dt} = -k_1\epsilon(t) - 4k_2X_s\epsilon(t) - 2k_2\epsilon^2(t) \quad (C1)$$

which implies a locally-linearized convergence rate for $\epsilon(t)$ of

$$-k_1 - 4k_2X_s \quad (C2)$$

Since the maximum of X is 4×10^{-6} and $k_2 = 1$, this convergence rate is dominated by $k_1 \in [0, 1]$. It is clear that the reaction convergence is very rapid in regions of sunlight and much slower in dark regions. Peak rates in this computation are 1/second.

Appendix D: Stability of chemical kinetics

Relation Eq. (B9) is plotted in Figs. B1 and B2. As can be seen in Fig. B1, for $k_1 > 0$, X converges to $D - r$. For $k_1 = 0$ Fig. B2 shows that X converges to zero for values greater than zero, but diverges for $X < 0$. While X should never be negative, numerical errors can lead to negative values. This divergence is slow, in the sense that the divergence is algebraic as can be seen in Eq. (B12). The divergence is also slow in the sense that the time required to double a negative X concentration is

$$t_2 = -\frac{1}{4k_2X}. \quad (D1)$$

Thus, for very small (negative) X , the time will have to be particularly large. However, for time of $2 \cdot t_2$, the solution is singular, reaching a value of $-\infty$.

Appendix E: CAM-SE chemistry-tracer (physics-dynamics) coupling

Algorithm 1 Pseudo-code explaining the different levels of subcycling and chemistry-transport (physics-dynamics) coupling used in CAM-SE.

Outer loop advances solution Δt_{chem} in time:

for $t = 1, 2, \dots$ **do**

 Compute chemistry tendencies $F_i, i = X, X_2$

for $ns = 1, 2, \dots, nsplit$ **do**

 Update state with chemistry/physics tendencies:

$$C_i = C_i + \frac{\Delta t_{chem}}{nsplit} F_i, i = X, X_2$$

for $rs = 1, 2, \dots, rsplit$ **do**

 subcycling of tracer advection:

$$C_i = C_i + \frac{\Delta t_{chem}}{nsplit \times rsplit} \mathcal{T}(C_i), i = X, X_2$$

end for

end for

end for

The different levels of subcycling used in CAM-SE are explained via pseudo-code in algorithm 1 using CAM-SE namelist conventions: `nsplit` and `rsplit`. The outer time-stepping loop starts with a call to chemistry that computes the chemistry tendencies over the entire chemistry time-step Δt_{chem} . The full chemistry tendencies are divided into `nsplit` adjustments of equal size and in each iteration of the `nsplit` loop the adjustments are added to the state. The tracer transport scheme may not be stable on the chemistry time-step (Δt_{chem}) or the coupling/adjustment time-step ($\Delta t_{cpl} = \Delta t_{chem}/nsplit$) so it must be subcycled with respect to the chemistry adjustments. The number of iterations of the tracer transport subcycling loop is `rsplit`. Note that since the `nsplit` and `rsplit` loops are nested the tracer time-step is $\Delta t_{tracer} = \Delta t_{chem}/(nsplit \times rsplit)$.

We distinguish between the `nsplit = 1` and `nsplit > 1` configurations and refer to them as `ftype = 1` and `ftype = 0`, respectively, based on CAM-SE namelist terminology (`ftype`

refers to *forcing type*)². In the CAM code, if $f_{\text{type}} = 1$ the state is updated with chemistry (physics) tendencies in the physics code whereas for $f_{\text{type}} = 0$ the adjustments take place in the dynamical core. CAM-FV uses a $f_{\text{type}} = 1$ configuration (with the caveat the chemistry tendencies are added after the transport is complete) and CAM-SE supports both $f_{\text{type}} = 0$ and $f_{\text{type}} = 1$. The current default CAM-SE uses $f_{\text{type}} = 0$ where the tendencies are split into n_{split} equal-sized adjustments.

In full model runs, if the physics time-step is large the $f_{\text{type}} = 1$ coupling method may produce large physics tendencies that drive the state much out of balance. When the dynamical core is given the physics updated state that is strongly (and locally) out of balance, the dynamical core may produce excessive gravity waves. To alleviate this one may choose to update the state with respect to physics tendencies throughout the tracer subcycling. This approach of adding the physics tendencies as several equal-sized adjustments is the $f_{\text{type}} = 0$ configuration that was explained above.

For the 1° setup (NE30NP4) we use the standard/recommended configuration with $n_{\text{split}} = 3$ and $n_{\text{split}} = 2$ (see pseudo-code in algorithm 1) so that for every third tracer time-step half of the chemistry tendencies are added to the state. In terms of tracer, chemistry and coupling time-steps this configuration corresponds to $\Delta t_{\text{tracer}} = 300\text{s}$, $\Delta t_{\text{chem}} = 1800\text{s}$ and $\Delta t_{\text{cpl}} = 600\text{s}$. CAM-FV uses $f_{\text{type}} = 1$ configuration where the chemistry tendencies are added once³. For CAM-FV $\Delta t_{\text{tracer}} = 900\text{s}$, $\Delta t_{\text{chem}} = 1800\text{s}$ and $\Delta t_{\text{cpl}} = 1800\text{s}$.

²when running the 3-D CAM-SE dynamical core n_{split} defines the vertical remapping time-step; if $f_{\text{type}} = 0$ then n_{split} also defines the adjustment time-step whereas if $f_{\text{type}} = 1$ then n_{split} only defines the remapping time-step as the full adjustments are added at the beginning of dynamics only.

³Note, however, that in CAM-FV the tendencies are added after tracer transport and not before

Appendix F: Analytic chemical forcing term

The analytic solution of the equations leads to an explicit solution for the change in concentrations during a time step with no flow.

$$F_X^n = -L_{\Delta t_{chem}} \frac{(X^n - D + r)(X^n + D + r)}{1 + E(\Delta t_{chem}) + \Delta t_{chem} L_{\Delta t_{chem}} (X^n + r)}. \quad (\text{F1})$$

where X^n is the value of X at the beginning of the n 'th time step,

$$L_{\Delta t_{chem}} = \begin{cases} \frac{1 - e^{-4k_2 D \Delta t_{chem}}}{D \Delta t_{chem}} & \text{if } D > 0 \\ 4k_2 & \text{if } D = 0. \end{cases} \quad (\text{F2})$$

and by conservation,

$$F_{X_2}^n = -\frac{1}{2} F_X^n. \quad (\text{F3})$$

In implementation, $L_{\Delta t_{chem}}$ needs some care. As $4k_2 D \Delta t$ approaches machine precision, it is useful to simply use the formula for $D = 0$ rather than the expression for $D > 0$.

Appendix G: Fortran code

In terms of Fortran code the analytical forcing is given by:

```
! dt is size of chemistry/physics time step
XT = X + 2.0*X2

r = k1/(4.0*k2)
d = sqrt(r*r + 2.0*r*XT)
e = exp(-4.0*k2*d*dt)
```



```

if(abs(d*k2*dt).gt. 1e-16)
  e1 = (1.0-e)/(d*dt)
else
  e1 = 4.0*k2
endif

f_X = -e1 * (X-d+r) * (X+d+r)/(1.0 + e + dt*e1*(X+r))
f_X2 = -f_X/2.0

```

The reaction rates are defined by

```

! k1 and k2 are reaction rates
k1_lat_center = 20.0 ! degrees
k1_lon_center = 300.0 ! degrees
k1 = max(0.d0,
         sin(lat)*sin(k1_lat_center)
         + cos(lat)*cos(k1_lat_center)
         *cos(lon-k1_lon_center))
k2 = 1.0

```

The initial condition is defined by

```

XT = 4.0e-6

r = k1/(4.0*k2)
d = sqrt(r*r + 2.0*XT*r)

X = d-r
X2 = XT/2.0 - (d-r)/2.0

```

These specifications are implemented in Fortran code in the Supplement.

Appendix H: CAM-SE time-stepping

The tracer algorithm and dynamical core use the same time step which is controlled by the maximum anticipated wind speed, but the dynamics uses more stages of a second-order accurate N-stage Runge–Kutta (RK) method in order to maintain stability. CAM-SE's tracer advection algorithm is based on a 3-stage RK strong-stability-preserving (SSP) time-stepping method (Spiteri and Ruuth, 2002). The SSP method ensures that the time step will preserve any monotonicity properties preserved by the underlying spatial discretization. CAM-SE uses monotone limiter in its advection scheme coupled with a monotone hyper-viscosity operator (Guba et al., 2014). This option renders the advection scheme second-order. The time-stepping scheme in the dynamical core uses a third-order accurate 5-stage RK method (modified version of Kinnmark and Gray, 1984a, b; P. A. Ullrich, personal communication, 2013) The extra stages are chosen to maximize the stable time step size. We also note that the hyper-diffusion in the dynamical core requires three subcycled iterations for each dynamics time step (in the NE30NP4 configuration).

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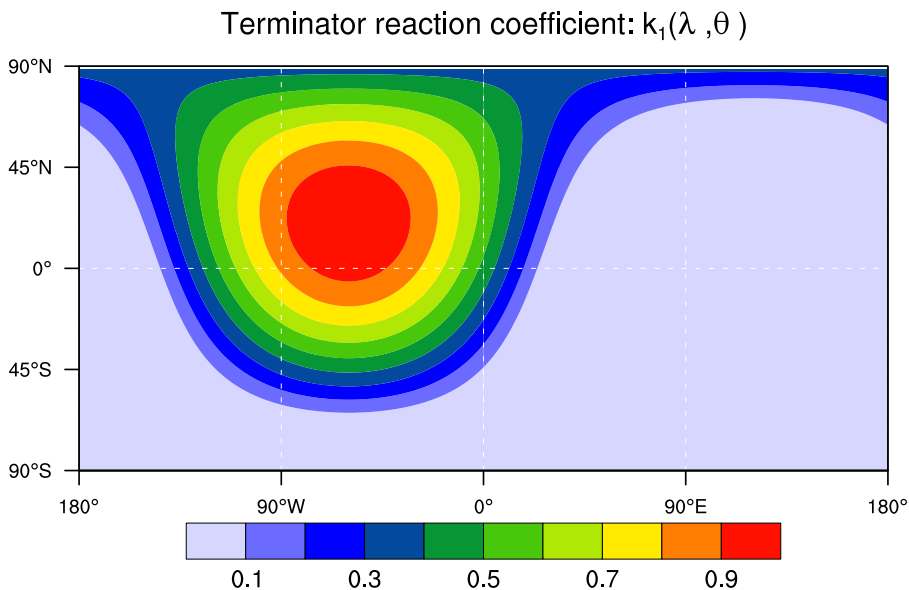


Figure 1. Contour plot of the terminator-“like” reaction coefficient $k_1(\lambda, \theta)$ where λ and θ are longitude and latitude, respectively.

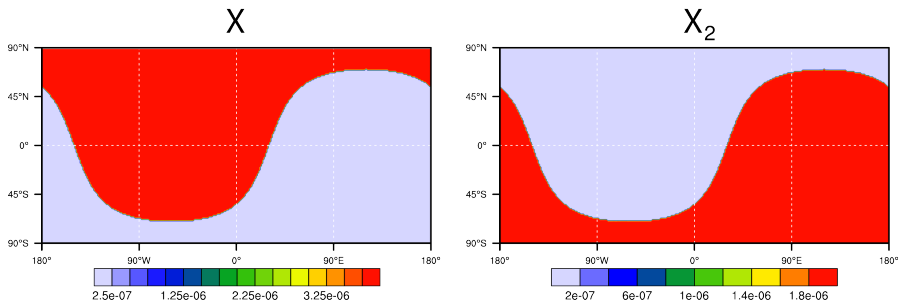


Figure 2. Contour plots of the steady-state solutions, assuming no flow, for X (left) and X₂ (right), respectively, computed from initial conditions $X = 4.0 \times 10^{-6}$ and $X_2 = 0$.

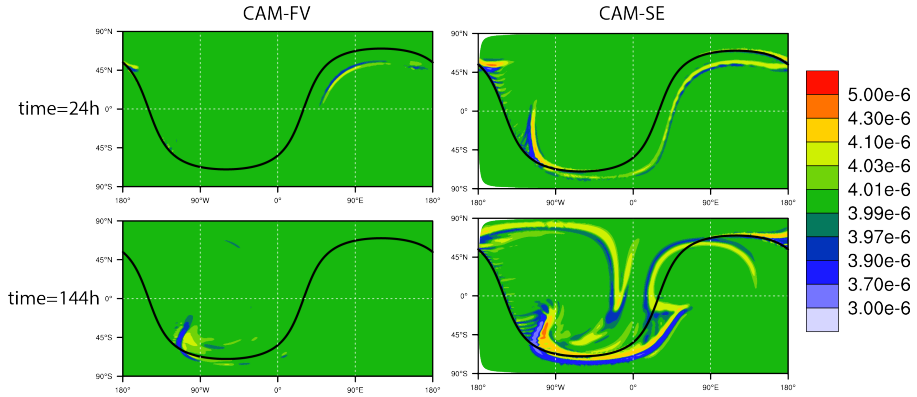


Figure 3. Contour plots of X_T for (left column) CAM-FV and for (right column) CAM-SE in $\text{ftype} = 0$ configuration at day 1 (upper row) and day 6 (lower row), respectively. Solid black line is the location of the terminator line. Note that the contour levels are not linear.

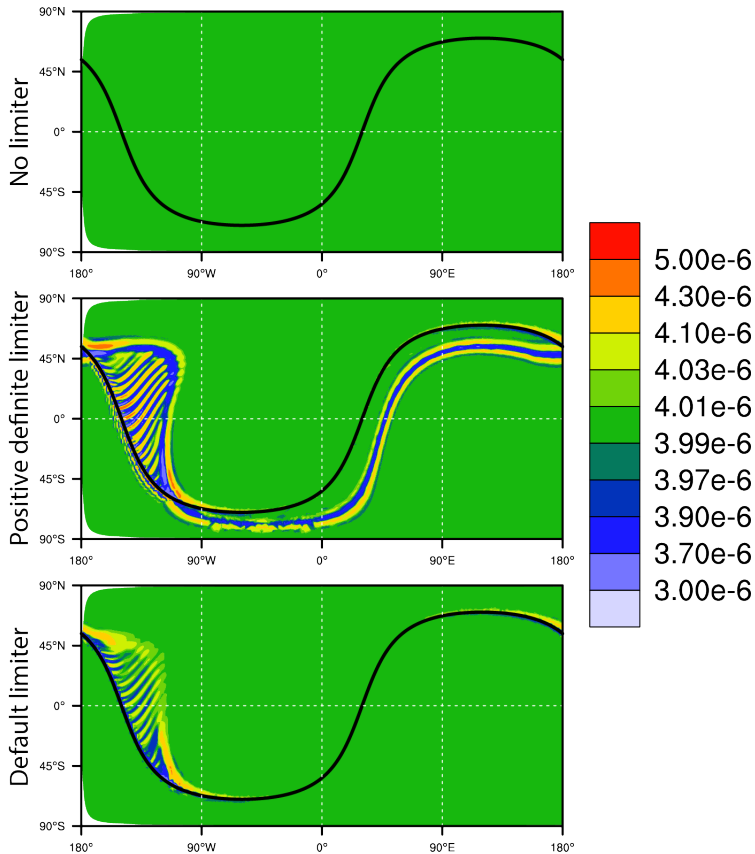


Figure 4. Contour plot of X_T at day 1 using CAM-SE in `ftype = 1` configuration where (upper) no limiter, (middle) positive definite limiter, and the default CAM-SE limiter is applied, respectively. The solid black line depicts the location of the terminator line. Note that the contour levels are not linear.

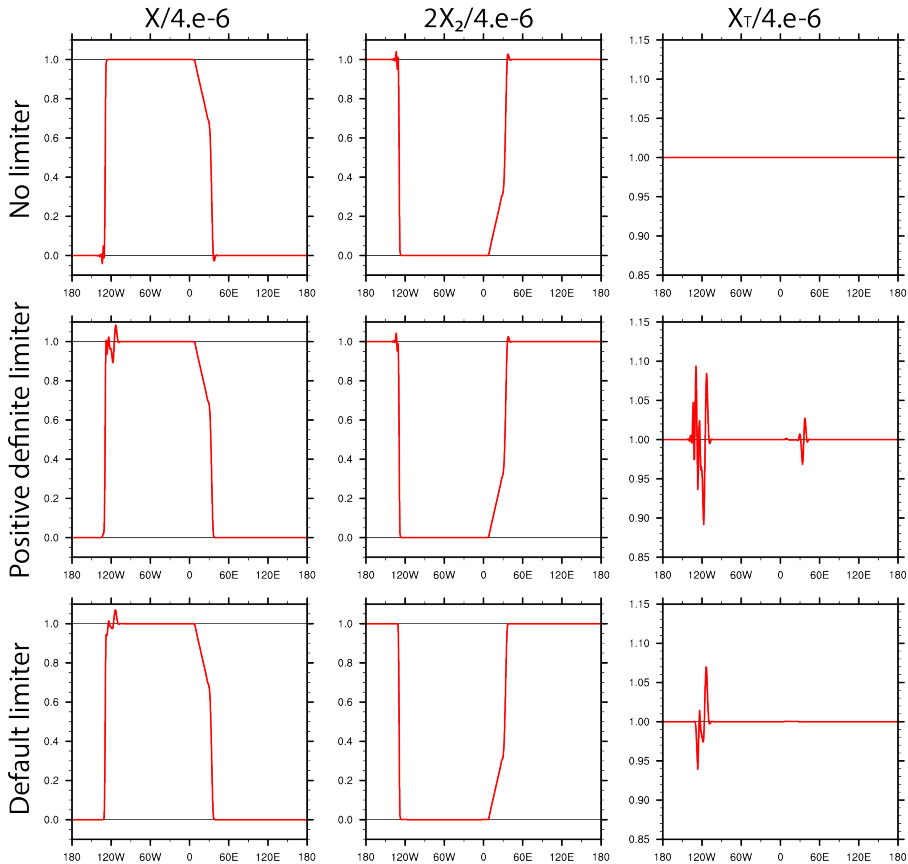


Figure 5. Cross sections of day 1 (left column) X , (middle column) $2 \times X_2$, and (right column) X_T at 45° S based on CAM-SE with (top row) no limiter, (middle row) positive definite limiter, (lower row) and default limiter, respectively. Results are normalized by 4×10^{-6} (the initial value of X_T).

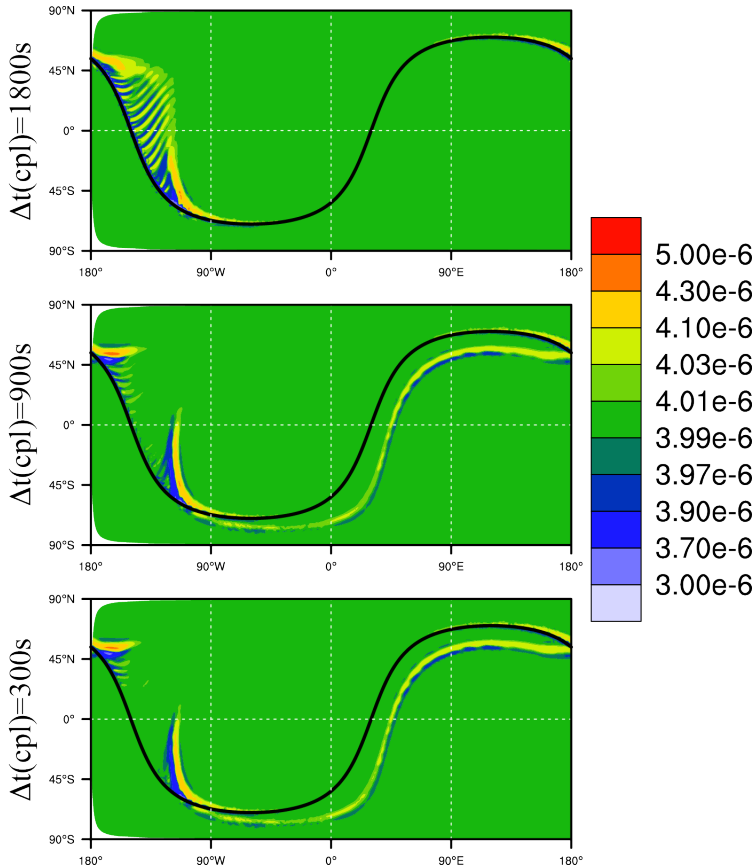


Figure 6. Contour plots of X_T at day 1 using CAM-SE based on (upper) $\Delta t_{cpl} = 1800s$, (middle) $\Delta t_{cpl} = 900s$, and (lower) $\Delta t_{cpl} = 300s$, respectively. In all simulations the tracer and chemistry time-step is constant $\Delta t_{tracer} = 300s$ and $\Delta t_{chem} = 1800s$, respectively.

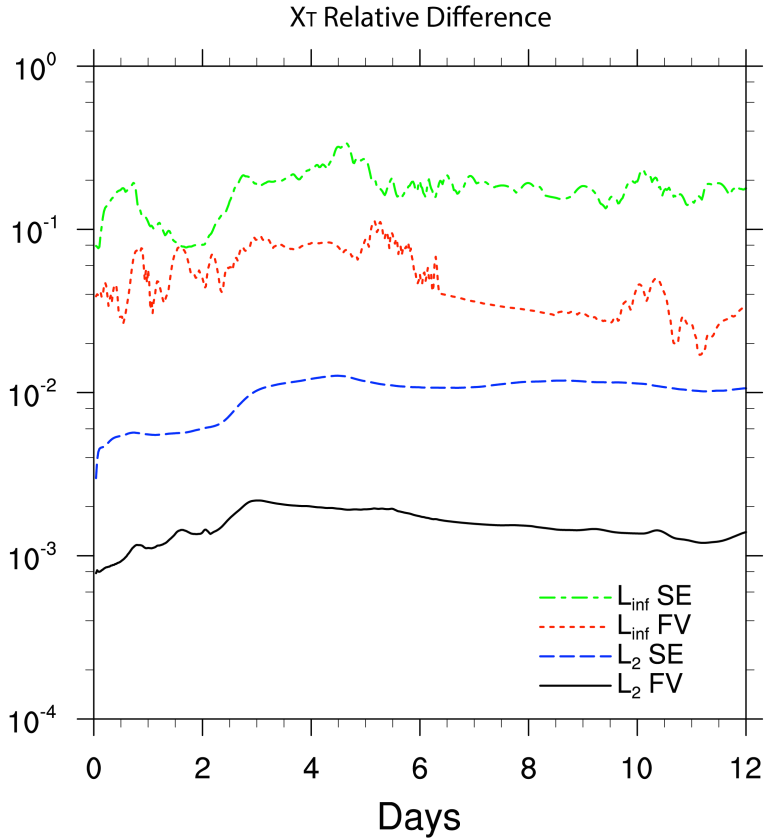


Figure 7. Time-evolution of standard error norms l_2 and l_{∞} for X_T using CAM-FV and CAM-SE dynamical cores. Note that the y axis is logarithmic.

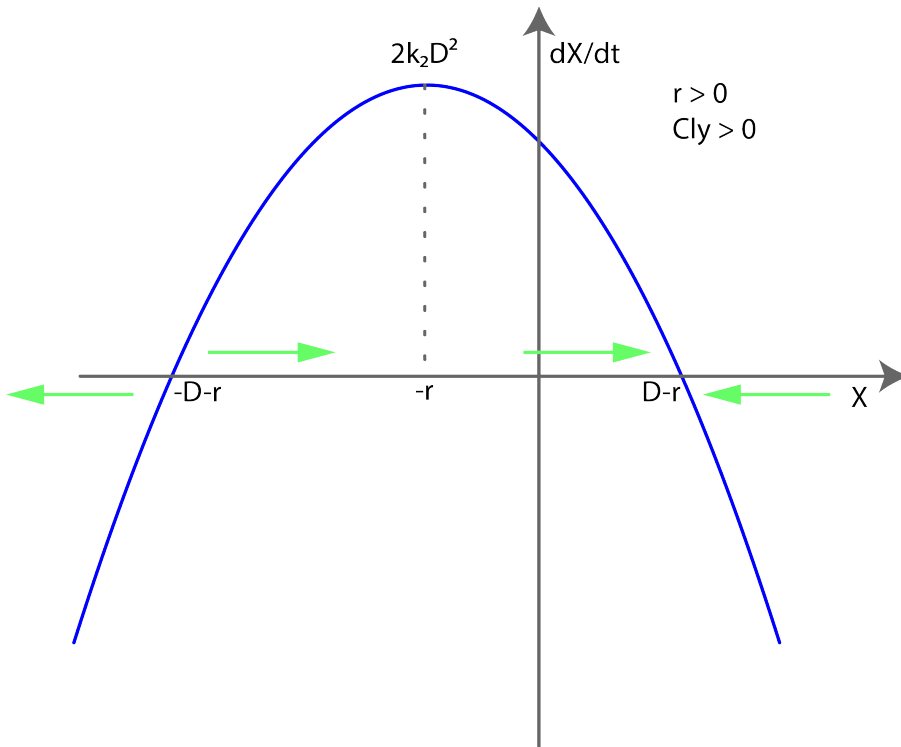


Figure B1. When $k_1 > 0$, or equivalently $r > 0$, there is a single stable limit point. X will converge to $D - r$ as long as $X > -D - r$.

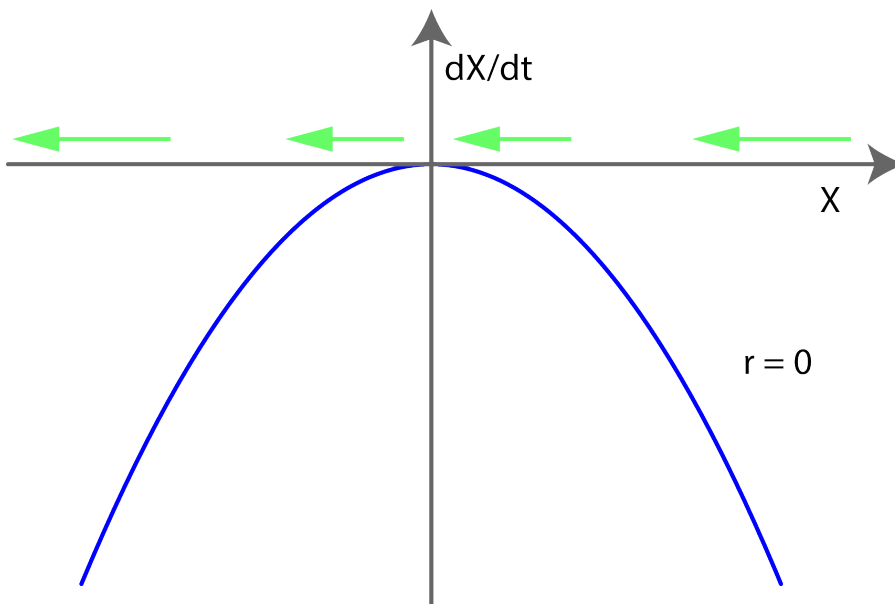


Figure B2. When $k_1 = 0$, or equivalently $r = 0$, X converges to zero, but if, for some numerical reason, X is driven negative, the kinetic equations will drive the concentrations even more negative.