Sensitivity of Chemistry-Transport Model Simulations to the Duration of Chemical and Transport Operators: A Case Study with GEOS-Chem v10-01

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Abstract

Chemistry-transport models involve considerable computational expense. Fine temporal resolution offers accuracy at the expense of computation time. Assessment is needed of the sensitivity of simulation accuracy to the duration of chemical and transport operators. We conduct a series of simulations with the GEOS-Chem chemistry-transport model at different temporal and spatial resolutions to examine the sensitivity of simulated atmospheric composition to operator duration. Subsequently, we compare the species simulated with operator durations from 10 min to
60 min as typically used by global chemistry-transport models, and identify the operator durations that optimize both computational expense and simulation accuracy. We find that longer continuous transport operator duration increases concentrations of emitted species such as nitrogen oxides and carbon monoxide since a more homogeneous distribution reduces loss through chemical reactions and dry deposition. The increased concentrations of ozone precursors increase ozone production with longer transport operator duration. Longer chemical operator duration decreases sulfate and ammonium but increases nitrate due to feedbacks with in-cloud sulfur dioxide oxidation and aerosol thermodynamics. The simulation duration decreases by up to a factor of 5 from fine (5 min) to coarse (60 min) operator duration. We assess the change in simulation accuracy with resolution by comparing the root mean square difference in ground-level concentrations of nitrogen oxides, secondary inorganic aerosols, ozone and carbon monoxide with a finer temporal or spatial resolution taken as “truth”. Relative simulation error for these species increases by more than a factor of 5 from the shortest (5 min) to longest (60 min) operator duration. Chemical operator duration twice that of the transport operator duration offers more simulation accuracy per unit computation. However, relative simulation error from coarser spatial resolution generally exceeds that from longer operator duration; e.g. degrading from 2° x 2.5° to 4° x 5° increases error by an order of magnitude. We recommend prioritizing fine spatial resolution before considering different operator durations in offline chemistry-transport models. We encourage chemistry-transport model users to specify in publications the durations of operators due to their effects on simulation accuracy.

1 Introduction

Global and regional chemistry-transport models (CTMs) have a wide range of applications in studies of climate, air quality, and biogeochemical cycling. The last few decades have witnessed
rapid development of modeling sophistication to tackle these issues, but that development is
associated with increasing computational expense. Typically, Eulerian models divide the
atmosphere into numerous \((10^4\text{-}10^5)\) grid boxes and solve the mass continuity equation to simulate
atmospheric composition. Numerical solution of the mass continuity equation involves separating
the different chemical and transport processes (or operators) through operator splitting. The
concentrations of simulated species are sensitive to the duration of operators used in the CTM.
Attention is needed to understand how operator duration affects model performance.

Numerous studies have examined the sensitivity of simulations to grid resolution for ozone (Jang
et al., 1995; Esler et al., 2004; Ito et al., 2009; Yu et al., 2016), ozone production efficiency (Liang
and Jacobson 2000), and ozone sensitivity to precursor emissions (Cohan et al., 2006; Henderson
et al., 2010). Simulation error increases proportional to the size of the horizontal grid (Wild and
Prather, 2006; Prather et al., 2008). Biases can be reduced by simulating sub grid scale processes
such as emission plumes from point sources (Sillman et al., 1990; Valin et al., 2011), aircraft
exhaust (Kraabøl et al., 2002), ship exhaust (Vinken et al., 2011), mineral dust emissions (Ridley
et al., 2013), and lightning (Cooper et al., 2014). The spatial and temporal resolution of the
meteorological fields used in CTMs can also influence model processes (Bian et al., 2009). The
spatiotemporal variation of carbon monoxide is better represented with finer grid resolution (Wang
et al., 2004; Chen et al., 2009; Yan et al., 2014). Moreover, fine horizontal resolution is important
for air quality exposure assessment and health impact studies (Punger and West, 2013; Fountoukis
et al., 2013; Thompson et al., 2014; Li et al., 2015). Fine vertical resolution can better represent
the effects of convection (Rind et al., 2007; Arteta et al., 2009). Simulations are also sensitive to
operator durations (Mallet et al., 2007; Santillana et al., 2016), however, few studies have
examined this sensitivity.
CTMs solve the continuity equation for tens to hundreds of chemical species, each with number density \( n \), for individual grid boxes defined in the Eulerian model.

\[
\frac{\partial n}{\partial t} = -\nabla \cdot nU + P - L
\]  

(1)

\( \frac{\partial n}{\partial t} \) represents the local temporal evolution of \( n \). \(-\nabla \cdot nU\) represents the transport flux divergence term, where \( U \) is the wind velocity vector. \( P \) and \( L \) are the local production and loss terms respectively. Typically, the above equation is discretized in space, and the continuity equation is simulated as a system of coupled non-linear partial differential equations with chemical and transport operators. These chemical and transport operators are usually simulated sequentially through operator splitting to increase computational efficiency (Hundsdorfer and Verwer, 2003).

The transport operator involves solving the 3-D advection equation using efficient numerical schemes (Prather, 1986; Lin and Rood, 1996). Boundary layer mixing, convection, emission and deposition are often simulated as individual operators. The chemical operator representing the temporal evolution of local sources and sinks involves numerically solving a system of coupled ordinary differential equations using efficient solvers (Jacobson and Turco, 1994; Damian et al., 2002). The integration timestep in a differential equation solver is important for efficient and accurate solution (Jacobson and Turco, 1994). Moreover, the model accuracy is affected by the duration of chemical and transport operators (Mallet and Sportisse, 2006; Mallet et al., 2007), and the order in which these operators are applied (Sportisse, 2000; Santillana et al., 2016). The operator splitting method requires the coupling between individual operators to be negligible over the operator duration. However, reducing operator durations increases computational expense. Attention is needed to this tradeoff.

We examine the sensitivity of a CTM to operator duration by conducting a series of simulations at different horizontal resolutions and operator durations. We then identify the optimal operator...
duration from the range of operator durations from 10 min to 60 min usually used by global CTMs (e.g., Horowitz et al., 2003; Huijnen et al. 2010). Section 2 describes the sensitivity simulations, the method to quantify the simulation error, as well as the method to identify the simulation operator durations that best account for both computational expense and simulation accuracy. Comparison of the sensitivity simulations, description of resolution-dependent errors, and the identification of appropriate chemical and transport operator durations are examined in section 3.

2 Materials and Methods

2.1 GEOS-Chem simulations

We conduct a series of sensitivity simulations with the GEOS-Chem CTM (version 10-01; www.geos-chem.org) at different horizontal resolutions and operator durations to examine the individual sensitivities to chemical and transport operator durations. The GEOS-Chem model (Bey et al., 2001) is used by about 100 research groups worldwide to simulate the oxidant-aerosol system. GEOS-Chem has the capability to be driven with several generations of assimilated meteorological data from the Goddard Earth Observing System (GEOS) at the NASA Global Modeling Assimilation Office (GMAO). For computational expedience, GEOS-Chem global simulations are often conducted using horizontal resolutions of either 4° x 5° or 2° x 2.5° degraded from the native resolution of GEOS meteorology. GEOS-Chem also has the capability for nested regional simulations where the global model provides dynamic boundary condition to the finer regional grids (Wang et al., 2004; Chen et al., 2009; Zhang et al., 2011; van Donkelaar et al., 2012).

We use the GEOS-5.2.0 meteorology available at a native horizontal resolution of 0.5° x 0.667° (Rienecker et al., 2008). It includes three-hour averaged 2-D fields such as mixed layer depth, and six-hour averaged 3-D fields such as zonal and meridional wind, and convective mass flux. The
height of the lowest level of the model is approximately 130 meters above the sea level, with 47 vertical levels.

GEOS-Chem performs species advection (A), vertical mixing (V), cloud convection (Z) and wet deposition (W) for every transport operator duration (T), as well as dry deposition (D), emissions (E), and chemistry (G) for every chemical operator duration (C) in the following order,

\[ A(T) \cdot D(C) \cdot E(C) \cdot V(T) \cdot Z(T) \cdot G(C) \cdot W(T) \] (2)

The traditional transport operator durations are 30 minutes at 4° x 5° resolution, 15 minutes at 2° x 2.5° resolution, and 10 min at 0.5° x 0.667° resolution. The traditional chemical operator duration is set to either 60 min or twice the transport operator duration based on the Strang operator splitting scheme (Strang, 1968) which follows \( T \cdot C \cdot T \cdot C \cdot T \) order repetitively with \( C = 2 \times T \). Transport operations are repeated twice before a chemical operation when \( C = 2 \times T \). We also consider an alternate splitting scheme which follows \( T \cdot C \cdot T \cdot C \) order repetitively with \( C = T \). Changes in operator duration from \( C = 2 \times T \) to \( C = T \) include effects of both time truncation \( (T \cdot T \) to \( T \) and operator splitting.

Advection is based on the multi-dimensional flux-form semi-Lagrangian advection scheme (Lin and Rood, 1996; Lin et al., 1994), with an additional pressure-fixer algorithm implemented for the conservation of species mass (Rotman et al., 2004). The cloud convection operator couples transport by convection (Balkanski et al., 1993; Wu et al., 2007) with gas-aerosol wet deposition (Liu et al., 2001; Wang et al., 2011; Amos et al., 2012). GEOS-Chem uses an internal integration timestep of 5 min for convective mixing within the cloud convection operator. The wet deposition operator includes scavenging by large-scale precipitation through first order operators, rainout and washout (Balkanski et al., 1993). We use a non-local boundary layer mixing scheme for vertical
transport (Holtslag and Boville, 1993, Lin and McElroy, 2010). Emissions are processed through the HEMCO module (Keller et al., 2014). A resistance-in-series method is used for dry deposition of species (Wesely, 1989; Wang et al., 1998; Zhang et al., 2001; Fisher et al., 2011).

GEOS-Chem uses a Sparse Matrix Vectorized GEAR II chemistry solver (Jacobson and Turco, 1994; Jacobson, 1995; 1998). The oxidant-aerosol chemistry simulation includes organic and black carbon (Park et al., 2003), mineral dust (Fairlie et al., 2007; Zender et al., 2003; Ginoux et al., 2001), sea salt (Alexander et al., 2005; Jaegle et al., 2011), and the sulfate-nitrate-ammonium system (Park et al., 2004). The photolysis frequency is calculated (Mao et al., 2010; Eastham et al., 2014) at the middle of the chemical operator duration using the Fast-JX algorithm (Bian and Prather, 2002). Simulation of gas-aerosol interactions are performed within the chemistry operator by aerosol extinction effects on photolysis rates (Martin et al., 2003), and heterogeneous chemistry (Jacob, 2000) including aerosol uptake of N_2O_5 (Evans and Jacob, 2005) and HO_2 (Mao et al., 2013). The ISORROPIA II thermodynamic module (Fontoukis and Nenes, 2007) performs aerosol-gas partitioning (Pye et al., 2009).

We conduct simulations for 2010 July at two horizontal resolutions of 4° x 5° and 2° x 2.5° globally, and 0.5° x 0.667° over the North America (140°W–40°W, 10°N–70°N) and East Asia (70°W–150°W, 11°S–55°N) nested regions. We use the 4° x 5° global simulation to archive dynamic boundary conditions every three hours for the nested simulations. We use one month spin up with each GEOS-Chem simulation to reduce the influence of initial conditions.

2.2 Computing platform

We conduct all simulations on the same computing platform to compare their computational performance. We use the Glooscap cluster of the Atlantic Computational Excellence Network
(ACENET) Consortium of Canadian Universities (http://www.ace-net.ca/wiki/Glooscap). The operating system is Linux 4.8. We use Intel Fortran compiler version 12. Each GEOS-Chem simulation is submitted as a 16-thread parallelized job on a single node.

We calculate the CPU time for the month of July for each operator separately using the Fortran-intrinsic routine, CPU_TIME. We found this value identical to the one calculated using the Linux command ‘qacct –j’. To reduce the effects of other jobs on the shared cluster, we repeat simulations five times, while excluding data output operations to minimize sensitivity to system input/output, and use the median to represent CPU time. We also report the standard error over the five simulations.

2.3 Assessing the relative simulation error

We treat the simulation with the shortest operator duration as the most accurate. This approach exploits the reduction in error associated with coupling across operators as operator duration diminishes. Assessing simulation error versus operator duration through comparison with observations is impaired by imperfect model processes, by the sparseness of measurements, and by model-observation representativeness biases. We take as “truth” the concentrations simulated with a chemical operator duration (C) of 10 minutes and a transport operator duration (T) of 5 minutes (represented as C10T05). Finer resolutions are computationally prohibitive. We define the relative simulation error $E_{\text{sim}}$ for species $s$ as the root mean square error (RMSE) of the species concentrations simulated with the finest resolution (“truth”) and the simulation under consideration ($\text{Sim}$), normalized by the concentrations in simulation “truth”,

$$E_{\text{sim}} = \sqrt{N_s \sum_{i=1}^{N_s} (\text{Truth}^i_s - \text{Sim}^i_s)^2} / \sum_{i=1}^{N_s} \text{Truth}^i_s$$

(3)
where, $i$ represents a particular grid box, with a total number of $N$ grid boxes of interest. RMSE in the numerator is chosen instead of absolute difference to more heavily penalize extrema. Normalization with the mass of the “true” simulation is intended to cross-compare $E_{sim}^s$ of different species. $E_{sim}^s$ captures the variation of a species $s$ from the “true” simulation.

We focus on four key species relevant to atmospheric chemistry, namely nitrogen oxides ($NO_x = NO + NO_2$), secondary inorganic aerosols (SIA: sum of sulfate, nitrate and ammonium), ozone ($O_3$), and carbon monoxide (CO). These species represent a range of lifetimes from a day ($NO_x$) to weeks (CO). The focus on SIA is designed to devote more attention to chemically active species than to mineral dust and sea salt. We sample the instantaneous values of simulated ground-level concentrations of these atmospheric species every 60 min to span the diurnal variation of chemical environments. We focus on concentrations in July near the Earth’s surface when and where chemical and transport timescales tend to be short.

### 2.4 Identifying the optimal operator duration

A practical way to select optimal chemical and transport operator durations is to identify the simulation with the lowest error ($E_{sim}^s$) per unit of computation time. To quantify the simulation accuracy per unit CPU time, we propose a simple metric, the CPU-time adjusted Composite Normalized Error (CNE) which represents a tradeoff between the simulation accuracy, and the associated computation expense. This is performed by normalizing the relative simulation error $E_{sim}^s$ for species $s$ by the CPU time $t$ for the simulation under consideration $t_{sim}$ and for a reference simulation $t_{ref}$, and taking the mean of the four species.
We normalize $E_{\text{sim}}^s$ by the reference $E_{\text{ref}}^s$ so that the CPU-time adjusted Composite Normalized Error for each species is of similar magnitude. The variation of CNE across operator durations is unaffected by the choice of reference simulation; C10T10 used here. The relative value of CPU time versus simulation accuracy is subjective and depends on scientific objective. This definition of CNE gives equal weighting to the respective cost of CPU time and simulation accuracy. The simulation with the lowest CNE is used to identify an optimal chemical and transport operator duration.

3 Results and discussion

Figure 1 shows the computational performance for the series of GEOS-Chem simulations conducted here. The CPU time decreases by factors of 3-5 from fine to coarse operator duration. The CPU time increases by about a factor of 4 from 4° x 5° to 2° x 2.5° and another factor of 2 to a single nested simulation at 0.5° x 0.667°. The linearity from 4° x 5° to 2° x 2.5° implies that grid boxes are sufficiently large that CPU time is proportional to the number of grid boxes, and that transport integration timesteps constrained by the Courant-Freidrich-Lewy criterion (Courant et al., 1967) are largely unaffected by changes to grid box size at these resolutions. Comparison of individual CPU times for chemical and transport operators shows that performing a single cycle of all chemical operations takes ~4 times that of a single cycle of transport operations at the global scale. This factor is reduced for nested simulations due in part to the additional CPU time for simulating boundary conditions.

Figure 2 illustrates the sensitivity of the simulations to chemical and transport operators at 2° x 2.5° horizontal resolution. The left column shows the species concentrations for the “true”
simulation (C10T05). The middle column shows the difference in species concentrations from doubling the transport operator duration. This doubling is in practice a change in time truncation of the transport operator from $T \cdot C \cdot T \cdot C \cdot T$ to $T \cdot C \cdot T \cdot C$ since the transport operator must keep pace with the chemistry operator. Increasing the transport operator duration tends to increase concentrations of emitted species like CO and NO$_x$ over source regions since species are more uniformly mixed by long continuous operator durations before loss processes such as dry deposition and chemistry occur. More homogeneous fields have lower dry deposition rates as a larger fraction is mixed aloft, and lower chemical loss rates depending on the chemical regime. The increase in CO over source regions is partly associated with decreases in OH. Increasing concentrations of ozone precursors increases ozone production (P[O$_3$]). Wild and Prather (2006) similarly found that ozone production increases at coarser horizontal resolution. Increasing the transport operator duration increases SIA components, especially over the source regions of East Asia, North India, and North America.

The right column in Fig. 2 shows the change in species concentrations from increasing the chemical operator duration. Hydroxyl radical concentrations increase, NO$_x$ concentrations decrease, and P[O$_3$] decreases with increasing chemical operator durations over source regions. Berntsen and Isaken (1997) found that the error introduced by coarser chemical operator durations is higher in polluted regions than the clean background due to the increased time lag, and invariant production and loss across rapid chemical cycles. A longer chemical operator duration decreases sulfate and ammonium but increases nitrate over source regions. Inspection of SO$_2$ and H$_2$O$_2$ fields indicates that sulfate formation through H$_2$O$_2$ in clouds decreases at longer chemical operator durations. In turn, SO$_2$ and NH$_3$ concentrations increase at longer chemical operator durations due to the corresponding decreases in ammonium sulfate or ammonium bisulfate. The additional free
ammonia at longer chemical operator durations tends to promote regional ammonium nitrate
formation depending on local thermodynamics. An increase of total SIA mass with increasing
chemical operator duration is driven by nitrate and ammonium, and partially compensated by a
reduction in sulfate, especially downwind of source regions. We find similar spatial patterns for
other operator duration combinations, and other horizontal resolutions.

Figure 3 shows the sensitivity of simulated species to changes in operator duration (C20T10 to
C10T05) at two other horizontal resolutions (global 4° x 5°, and nested North America 0.5° x 0.67°
simulations) considered here. Spatial patterns of monthly mean ground-level concentrations, and
absolute differences are similar, albeit with finer spatial heterogeneity resolved in the nested
simulation. However, some resolution dependent differences do arise reflecting nonlinear
feedbacks.

Figures 4 shows the relative simulation error for nitrogen oxides, secondary inorganic aerosols,
ozone and carbon monoxide with varying operator durations at 2° x 2.5° horizontal resolution.
Relative simulation errors for all these major species increase by more than a factor of 5 from the
shortest to longest operator duration. Errors increase fairly smoothly with increasing chemical and
transport operator duration until the transport operator duration exceeds 30 min. Then errors
increase by an order of magnitude for long lived species of O₃ and CO. The saw-tooth pattern for
CO versus O₃ reflects a greater sensitivity of CO to transport operator duration and a greater
sensitivity of O₃ to chemical operator duration. Relative simulation errors for other horizontal
resolutions follow similar pattern. These relative errors of 5% - 35% for NOₓ and SIA are
comparable to typical model-observation errors of ~30% for NOₓ (Boersma et al., 2008; Hudman
et al., 2006) and 20 - 40% for SIA (Philip et al., 2014; Heald et al., 2012). Operator duration errors
of <2% for O$_3$ and CO are smaller than typical model-observation errors of ~20% for ozone (Zhang et al., 2011; Wang et al., 2009) and 10 - 20% for CO (Duncan et al., 2007; Shindell et al., 2006).

Figure 5 shows the difference in simulated species at 2° x 2.5° horizontal resolution for the GEOS-Chem traditional (C30T15) minus the finest operator durations considered (C10T05). The spatial variation for the monthly mean ground-level concentrations is generally within 5-15% for short lived species like NO$_x$ and SIA, and within 1% for longer lived species like O$_3$ and CO. Santillana et al. (2016) similarly found an upper limit of 10% for operator splitting errors. However, the maximum hourly spatial variation can exceed 50% for short lived species and 5% for longer lived species. The spatial pattern of extrema resembles that of the monthly mean, albeit with more heterogeneity from synoptic variation.

We also examined the diurnal variation and vertical profile of extrema. Extrema arise from all times of day with a slight tendency for larger values for NO$_x$ at night, for O$_3$ near sunrise and sunset, and for SIA and CO near noon. Zonal mean vertical profiles exhibit largest differences in the lower troposphere for NO$_x$ and SIA, with more homogeneous differences throughout the troposphere for O$_3$ and CO. Near the subtropical jets of the upper troposphere O$_3$ and CO have maximum extrema of up to 3%.

Figure 6 shows the CPU-time adjusted Composite Normalized Error for the GEOS-Chem simulations at various horizontal resolutions and operator durations. The CNE is significantly higher with C = T than C = 2 x T. We confirmed this tendency with different choices of “truth” (such as C05T05, C10T10) or reference (such as C10T05) simulations. This finding motivates the traditional approach of using C = 2 x T in GEOS-Chem simulations. Applying the chemical operator as frequently as the transport operator (with C = T) appears to increase computation cost.
with little benefit in accuracy. The CNE for all three horizontal resolutions have noisy minima with a chemical operator duration of 20 min and a transport operator duration of 10 min (C20T10). A unit of computation time has a similar efficiency for a small range of operator durations from 10 min to 20 min. We found similar patterns in the variation of CNE with operator durations with CNE calculated for selected domains, such as over Northern Hemisphere, nested model regions, land grid boxes, and over the entire troposphere. We conducted additional simulations at $4^\circ \times 5^\circ$ horizontal resolution for January 2011 with a spin up of 7 months, and found similar patterns in CNE.

The relative simulation error decreases by 40-50% (Fig. 4) by changing the operator duration from the traditional (C30T15) to the optimal (C20T10) at $2^\circ \times 2.5^\circ$ horizontal resolution. The relative spatial variations are $<20\%$ for NO$_x$ and SIA, and $<1\%$ for O$_3$ and CO. However, the CPU time increases by 20% by the decrease in operator duration.

Table 1 shows the relative simulation error at $4^\circ \times 5^\circ$ horizontal resolution with “truth” at $2^\circ \times 2.5^\circ$ resolution (C10T05) to investigate the tradeoff between horizontal resolution and operator duration. The simulation error for all species at $4^\circ \times 5^\circ$ resolution increases by an order of magnitude compared to $2^\circ \times 2.5^\circ$ resolution for any choice of operator duration tested here. The error in this configuration is insensitive to operator duration, and dominated by representativeness differences due to spatial structure resolved at $2^\circ \times 2.5^\circ$ resolution, but not at $4^\circ \times 5^\circ$ resolution.

Nonlinear chemistry at different horizontal resolutions (e.g., Wild and Prather, 2006) also plays a role. Numerical errors due to advection processes generally exceed those from operator splitting (e.g., Prather et al., 2008; Santillana et al., 2016). We therefore recommend prioritizing horizontal resolution over operator duration for offline CTMs using time-averaged meteorological fields as tested here. As meteorological fields used in CTMs become available at finer temporal and spatial
resolution, the value of shorter operator duration should further increase. We encourage CTM users to specify in publication the duration of operators due to its effect on simulation accuracy.

4 Conclusions

The computational expense of chemistry-transport models warrants investigation into their efficiency and accuracy. Solving the continuity equation in CTMs through operator splitting method offers numerical efficiency, however, few studies have examined the implications of operator duration on simulation accuracy. We conducted simulations with the GEOS-Chem model for multiple choices of operator duration from 10 min to 60 min as typically used by global CTMs. We found that longer continuous transport operator durations increase ozone precursors and ozone production over source regions since a more homogeneous distribution reduces loss through chemical reactions and dry deposition. Longer chemical operator durations decrease NOx and ozone production over source regions. Longer chemical operator durations reduce sulfate and ammonium concentrations, however increase nitrate due to feedbacks with in-cloud SO2 oxidation and local aerosol thermodynamics.

We investigated the computational efficiency with the GEOS-Chem model, and found that the simulation computation time decreases by up to a factor of 5 from short (C10T05) to long (C60T60) operator duration. The chemical operator consumes about four times the CPU time of the transport operator. We subsequently compared the root mean square differences in the ground-level concentrations of nitrogen oxides, secondary inorganic aerosols (SIA), ozone and carbon monoxide with a finer temporal or spatial resolution taken as “truth”, and estimated the relative simulation error. The relative simulation error for these species increases by more than a factor of 5 from the shortest to longest operator duration. Monthly mean simulation errors of about 30% for
NO\textsubscript{x} and SIA from long operator duration are comparable to typical model-observation errors, while simulation errors for CO and O\textsubscript{3} tend to be less than 2\% for operator duration < 30 min.

In order to account for simulation accuracy with computational cost, we proposed a metric, CPU-time adjusted Composite Normalized Error that identifies the operator duration with respect to CPU cost. We find greater efficiency of using $C = 2 \times T$ than $C = T$ for all horizontal resolutions. The Composite Normalized Error exhibits a noisy minimum for a chemical operator duration of 20 min and transport operator duration of 10 min for the range of operator durations and horizontal resolutions considered here. Nonetheless, the relative simulation error from changing horizontal resolution exceeds that from changing operator durations within a horizontal resolution. We recommend prioritizing fine spatial resolution before considering different operator durations in offline CTMs with time-averaged archived meteorological fields as tested here. The importance of shorter operator durations should increase with the availability of time-averaged meteorological fields at higher temporal resolution. Short operator durations could offer even greater benefits to simulation accuracy in online CTMs that offer meteorological fields at temporal resolutions closer to operator duration. We encourage CTM users to specify in publications the durations of operators due to their effects on simulation accuracy.
Code Availability

The GEOS-Chem code is freely accessible to the public, by following the guidelines in http://wiki.geos-chem.org/). This work used GEOS-Chem version 10-01.

Acknowledgements

We thank Colette Heald, Daniel Jacob and Patrick Kim for useful comments at the early stages of this research. We are grateful to three anonymous reviewers for helpful comments. This work was supported by National Science and Engineering Research Council, Canada, and Atlantic Computational Excellence Network (http://www.ace-net.ca/).

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Table 1: Comparison of mean* relative simulation error versus horizontal resolution, with “truth” defined at 2° x 2.5° horizontal resolution

| Species                      | Mean relative simulation error (unitless) |
|------------------------------|-------------------------------------------|
|                              | 4° x 5° resolution | 2° x 2.5° resolution |
| Nitrogen oxides              | 2.1                         | 0.092                |
| Secondary inorganic aerosols | 1.0                         | 0.14                 |
| Ozone                        | 0.17                        | 0.004                |
| Carbon monoxide              | 0.36                        | 0.005                |

* Mean taken for operator durations ≤ 30 min.
Figure 1: CPU time for GEOS-Chem simulations with various operator durations at three horizontal resolutions. Global simulations are at 4° x 5° (top) and 2° x 2.5° (middle) resolutions. The bottom panel contains results for the average of two nested regions North America and East Asia at 0.5° x 0.667° resolution. Colored lines represent the CPU time for simulating transport (red) and chemical (blue) operators, and the sum of the two (green). Error bars represent standard error over five simulations. Simulations are represented in the abscissa as CccTtt with chemical operator duration, C = cc minutes, and transport operator duration, T = tt minutes.
Figure 2a: Sensitivity of simulated species to the duration of chemical and transport operators. The left column contains monthly mean ground-level concentrations simulated with the shortest operator duration considered (C10T05) at 2° x 2.5° horizontal resolution. Other columns contain the absolute differences from doubling the transport operator duration to C10T10 (middle), and doubling the chemical operator duration to C20T05 (right). Each row from top to bottom represents carbon monoxide (CO), nitrogen oxides (NO\textsubscript{x}), hydroxyl radical (OH), and the production of ozone (P[O3]). Simulations are represented as CccTtt with chemical operator duration, C = cc minutes, and transport operator duration, T = tt minutes.
Figure 2b: As described in Fig. 2a, but each row from top to bottom represents ozone (O$_3$), sulfur dioxide (SO$_2$), sulfate (SO$_4^{2-}$), and nitrate (NO$_3^-$).
Figure 3: Sensitivity of simulated species to changes in operator duration (C20T10 to C10T05) at two different horizontal resolutions over North America (global 4° x 5°, and nested 0.5° x 0.67° simulations). The upper two rows contain monthly mean ground-level concentrations simulated with the C20T10 operator duration for 4° x 5° (top row) and 0.5° x 0.67° (second row) resolutions. The two lower rows contain the monthly mean differences (C20T10 minus C10T05) for 4° x 5° (third row) and 0.5° x 0.67° (bottom row) resolutions. Each column from left to right represents nitrogen oxides (NOx), secondary inorganic aerosols (SIA), ozone (O3), and carbon monoxide (CO).
Figure 4: Relative simulation error of different species ($E_{sim}$, eq. 3) with various operator durations at 2° x 2.5° horizontal resolution. Colored lines and dots represent the relative simulation error for nitrogen oxides (NO$_x$; red), secondary inorganic aerosols (SIA; blue), ozone (O$_3$; green), and carbon monoxide (CO; magenta). Simulations are represented in the abscissa as CccTtt with chemical operator duration, C = cc minutes, and transport operator duration, T = tt minutes.
Figure 5: Effect on simulated species of changing from the GEOS-Chem traditional operator durations (C30T15) to the shortest operator durations considered (C10T05). The top row contains monthly mean ground-level concentrations simulated with the C30T15 operator duration at 2° x 2.5° horizontal resolution. The next two rows contain the monthly mean differences (C30T15 minus C10T05) for absolute (second row) and relative (third row) differences. The two lowest rows contain the maximum differences (C30T15 minus C10T05) for absolute (fourth row) and relative (bottom row) differences. Each column from left to right represents nitrogen oxides (NO$_x$), secondary inorganic aerosols (SIA), ozone (O$_3$), and carbon monoxide (CO).
Figure 6: CPU-time adjusted Composite Normalized Error (CNE, eq. 4) for GEOS-Chem simulations with various horizontal resolutions and operator durations. Colored lines and dots represent the CNE for the global simulations at $4^\circ \times 5^\circ$ (red) and $2^\circ \times 2.5^\circ$ (blue), and the nested simulations at $0.5^\circ \times 0.667^\circ$ (green) horizontal resolutions. Error bars represent standard error in CPU time. Simulations are represented in the abscissa as CccTtt with chemical operator duration, $C = cc$ minutes, and transport operator duration, $T = tt$ minutes.