

Response to Referee comments # 3

Review of Philip et al., Sensitivity of Chemistry-Transport Model simulations to the duration of chemical and transport operators: A case study with GEOS-Chem v10-01

The manuscript provides an analysis of the growth of errors from using longer model timesteps to achieve faster model execution with the GEOS-Chem chemical transport model. In particular, the paper focuses on the growth of error as the timestep used for the time split solution of individual processes in the model is increased. The paper presents an analysis of a fundamental choice that chemical transport modellers must make, that has not received much analysis to the best of my knowledge. The paper is well organized and the use of the ‘CPU-time adjusted Composite Normalized Error’ provides a nice summary of the trade-offs of runtime versus accuracy, the paper has a few omissions that hinder a full understanding of the results. I would find the paper acceptable for publication after these concerns are addressed.

I should mention at the outset that this is my first review of the manuscript – I was not one of the two original reviewers – and that I have read and taken into consideration the comments and replies made during the first round of reviews.

We thank the referee for valuable comments. Our responses to referee comments are below in blue. We have modified the article based on these suggestions. Line numbers refer to the revised manuscript.

My most significant concern of the manuscript in its present form is the interpretation of the differences between the C10T05 and C10T10 simulations that are shown in Figure 2 and discussed on lines 215-227. In particular, in interpreting the differences between the C10T05 and C10T10 simulations the authors state ‘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 operator durations before loss processes such as deposition and chemistry occur.’ (lines 218-221). The differences between C10T05 and C10T10 are due to a change in going from an operator order of C-T-T-C-T-T... to C-T-C-T-C-T..., if I have correctly understood how the model timestepping works. In both cases, the transport operator

must still calculate a full 10 minutes of model time to keep pace with the chemistry operator for each model timestep. The differences you have shown in the middle column of Figure 2, I would argue, are more due to the different time truncation errors associated with each transport operator calculation than a change in the errors due to time splitting. For a single timestep of 10 minutes, advection will produce different solutions depending on the timestep used (2x5 minutes or 10 minutes). Even a simple calculation such as dry deposition can produce different results whether called twice or once to cover 10 minutes, particularly depending on the details of the numerical treatment (implicit, explicit, analytic). The discussion seems to suggest the differences are due to time splitting errors ('long operator durations') but each individual operator in both the C10T05 and C10T10 case has the same operator duration. The discussion presented in this section would be much more applicable to the differences between C20T05 and C10T05, as the operator duration has actually changed in this case. If you keep in mind the idea that there are two sources of error (time truncation and operator splitting) it may help in the interpretation of the differences between the C=T and C=2xT simulations, in particular the saw-tooth pattern of error growth in O₃ and CO shown in Figure 4.

Helpful suggestion. Indeed both time truncation and operator splitting errors contribute to the operator duration differences in Figure 2 in ways that are not fully separable. We thus refer to both as operator duration effects. We define transport operator duration as the duration of a single call (e.g., T rather than T · T) to avoid ambiguity in the duration of a single call. We added lines 25-26, " We find that longer continuous transport operator duration increases ..."; lines 127-128, "Changes in operator duration from C = 2 × T to C = T include effects of both time truncation (T · T to T) and operator splitting."; lines 225-227, "This doubling is in practice a change in time truncation of the transport operator from T · C · T · T · C · T to T · C · T · C since the transport operator must keep pace with the chemistry operator."; lines 263-265, " 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."; and line 322, " We found that longer continuous transport operator durations increase ..."

A second concern is the presentation of the operator structure of GEOS-Chem, found around lines 113-124. In equation (2), there is a photolysis operator that is separate from the chemistry operator. Is the photolysis operator really a separate operation that modifies the model solution

(the chemical fields) and is calculated separately from the chemistry? I am guessing this is the calculation of photolysis rates, but if so this calculation would not strictly be counted as a separate model operator as the photolysis rates only affect the model solution through the calculation of chemistry. In that case, photolysis should be removed from equation (2), perhaps with a note that photolysis rates are calculated as part of the chemistry operator.

We agree. Removed photolysis from equation 2. We already have a description regarding this in lines 144-146, “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).”

On lines 129-130 there is a short mention that ‘GEOS-Chem uses an internal integration timestep of 5 min for convective mixing.’ I assume that no matter what the chemical and transport operator durations are set to, the convection operator shown in equation (2) is calculated with an internal 5 minute timestep. This would seem to be an important caveat that should be included in the preceding paragraph discussing the time splitting.

We have added to line 134 the emphasis that this internal timestep occurs within the convection operator. However, we respectfully retain the position of this sentence since it fits well within the discussion of other details on individual operators.

On lines 127-129, there is the statement that ‘Transport by convection is coupled with gas-aerosol wet deposition.’ yet equation (2) shows separate operators for cloud convection (Z) and wet deposition (W). I would assume the wet deposition operator is only from large-scale (stratiform) precipitation. If so, it should be made more transparent to the reader that wet deposition from convective precipitation is calculated as part of the convective transport operator and stratiform wet deposition is in the separate W operator.

We agree. We added lines 134-136, “The wet deposition operator includes scavenging by large-scale precipitation through first order operators, rainout and washout (Balkanski et al., 1993).”

Lastly, regarding the form of the operators in GEOS-Chem, there is no mention of how aerosol (and cloud?) chemistry and gas-phase chemistry interact. Are they two separate operators? And if so they should probably be presented as separate operators in equation (2). This could be an

issue as mention is made on lines 234-236 of differences in in-cloud sulphate oxidation due to H₂O₂ differences with longer chemical operator durations.

They are a single operator. We added to lines 146-147, “ Simulation of gas-aerosol interactions are performed within the chemistry operator by ...”

Other minor comments are given below.

Lines 48-50: very generally the form of CTMs is introduced in the first paragraph and discussion of operator duration is broached, but there is no mention of why operator splitting in CTMs is required.

We added a sentence in lines 50-51, “Numerical solution of the mass continuity equation involves separating the different chemical and transport processes (or operators) through operator splitting.”

Lines 64-65: a minor quibble but here it is stated that ‘Fine vertical resolution can better represent convection’. To be more exact, this statement should probably say ‘can better represent the effects of convection...’.

Modified lines 66-67, “Fine vertical resolution can better represent the effects of convection (Rind et al., 2007; Arteta et al., 2009).”

Lines 67-79: here the continuity equation is discussed and advection by resolved winds along with chemical production and destruction are explicitly discussed but there is no mention of sub-grid scale transport – turbulent mixing, convection – or other terms such as emissions, dry deposition.

We added a sentence to lines 80-81, “Boundary layer mixing, convection, emission and deposition are often simulated as individual operators.”

Line 119-120: The sentence ‘The traditional chemical operator durations have varied from either 60 min or twice the transport operator duration...’ is overly complex. On re-reading the reference

to 60 minutes is twice the maximum of 30 minutes given for the transport operator, but the construction is a bit convoluted and it took a bit of rereading to figure out what was being expressed. How about ‘The traditional chemical operator duration is set to twice the transport operator duration...’

We modified lines 122-124, “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 $T \cdot C \cdot T \cdot T \cdot C \cdot T$ order repetitively with $C = 2 \times T$.”

Lines 159-160: Would you have the range of execution times for the five repetitions of each simulation handy? A passing mention of the range would help the reader understand the robustness of the timing.

We added a sentence to lines 166-167, “We also report the standard error over the five simulations.” These values are in Figure 1.

Lines 290-291: In comparing the errors and execution time of the C30T15 and C20T10 simulations you say that ‘the CPU time increases by 20% by the decrease in operator duration.’ If it was completely linear, going from C30T15 to C20T10 should increase the model execution time by 50% because you now need 50% more chemical and transport operations to complete a given amount of model time. Do you have an idea of why this particular case does not seem to scale linearly?

This case is not unusual. Inspection of Figure 1 reveals that there is additional computational overhead that behaves as an offset term. For example progressing from C60T60 to C10T05 increases CPU time by a factor of 4 rather than 6.

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

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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 $2^\circ \times 2.5^\circ$ to $4^\circ \times 5^\circ$ 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.

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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 - 10^8) 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.

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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.

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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.

$$\frac{\partial n}{\partial t} = -\nabla \cdot nU + P - L \quad (1)$$

$\partial n/\partial t$ 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 (Hundsdoerfer 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

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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 $4^\circ \times 5^\circ$ or $2^\circ \times 2.5^\circ$ 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 $0.5^\circ \times 0.667^\circ$ (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

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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,

$$A(T) \cdot D(C) \cdot E(C) \cdot V(T) \cdot Z(T) \cdot G(C) \cdot W(T) \quad (2)$$

The traditional transport operator durations are 30 minutes at $4^\circ \times 5^\circ$ resolution, 15 minutes at $2^\circ \times 2.5^\circ$ resolution, and 10 min at $0.5^\circ \times 0.667^\circ$ 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 $T \cdot C \cdot T \cdot T \cdot C \cdot T$ 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

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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).

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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 $4^\circ \times 5^\circ$ and $2^\circ \times 2.5^\circ$ globally, and $0.5^\circ \times 0.667^\circ$ 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 $4^\circ \times 5^\circ$ 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_{sim}^s 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 (Sim), normalized by the concentrations in simulation “truth”,

$$E_{sim}^s = \frac{\sqrt{\sum_{i=1}^{i=N} (Truth_i^s - Sim_i^s)^2}}{\sum_{i=1}^{i=N} Truth_i^s} \quad (3)$$

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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 ($\text{NO}_x = \text{NO} + \text{NO}_2$), 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.

$$CNE = \left(\frac{1}{4} \times \sum_s \frac{E_{sim}^s}{E_{ref}^s} \right) \times \left(\frac{t_{sim}}{t_{ref}} \right)$$

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$$(4) \quad CNE = \left(\frac{1}{4} \times \sum_s \frac{E_{sim}^s}{E_{ref}^s} \right) \times \left(\frac{t_{sim}}{t_{ref}} \right)$$

We normalize E_{sim}^s by the reference E_{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 $4^\circ \times 5^\circ$ to $2^\circ \times 2.5^\circ$ and another factor of 2 to a single nested simulation at $0.5^\circ \times 0.667^\circ$. The linearity from $4^\circ \times 5^\circ$ to $2^\circ \times 2.5^\circ$ 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 $2^\circ \times 2.5^\circ$ horizontal resolution. The left column shows the species concentrations for the “true”

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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 $T \cdot C \cdot T \cdot T \cdot C \cdot T$ 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[O₃]). 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[O₃] 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 SO₂ and H₂O₂ fields indicates that sulfate formation through H₂O₂ in clouds decreases at longer chemical operator durations. In turn, SO₂ and NH₃ concentrations increase at longer chemical operator durations due to the corresponding decreases in ammonium sulfate or ammonium bisulfate. The additional free

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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.

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Figure 3 shows the sensitivity of simulated species to changes in operator duration (C20T10 to C10T05) at two other horizontal resolutions (global $4^\circ \times 5^\circ$, and nested North America $0.5^\circ \times 0.67^\circ$ 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 $2^\circ \times 2.5^\circ$ 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 O_3 and CO. The saw-tooth pattern for CO versus O_3 reflects a greater sensitivity of CO to transport operator duration and a greater sensitivity of O_3 to chemical operator duration. Relative simulation errors for other horizontal resolutions follow similar pattern. These relative errors of 5% - 35% for NO_x and SIA are comparable to typical model-observation errors of ~30% for NO_x (Boersma et al., 2008; Hudman et al., 2006) and 20 - 40% for SIA (Philip et al., 2014; Heald et al., 2012). Operator duration errors

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of <2% for O₃ 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 O₃ 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 O₃ 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 O₃ and CO. Near the subtropical jets of the upper troposphere O₃ 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

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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.

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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 NO_x 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

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NO_x and SIA from long operator duration are comparable to typical model-observation errors, while simulation errors for CO and O₃ 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.

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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.

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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.

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Acknowledgements

~~T~~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.

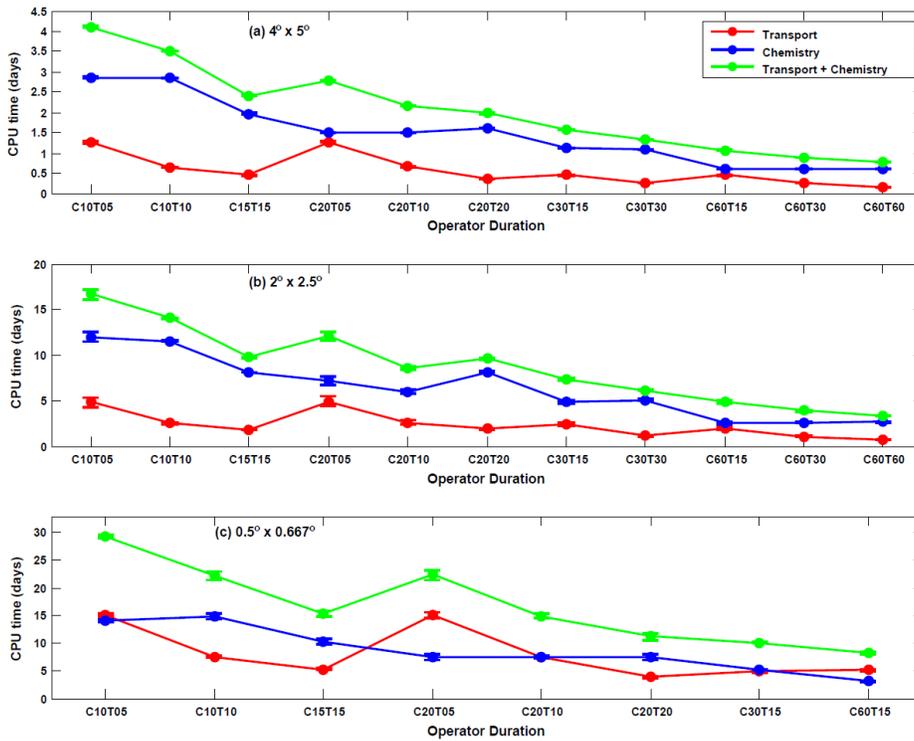


Figure 1: CPU time for GEOS-Chem simulations with various operator durations at three horizontal resolutions. Global simulations are at $4^\circ \times 5^\circ$ (top) and $2^\circ \times 2.5^\circ$ (middle) resolutions. The bottom panel contains results for the average of two nested regions North America and East Asia at $0.5^\circ \times 0.667^\circ$ 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.

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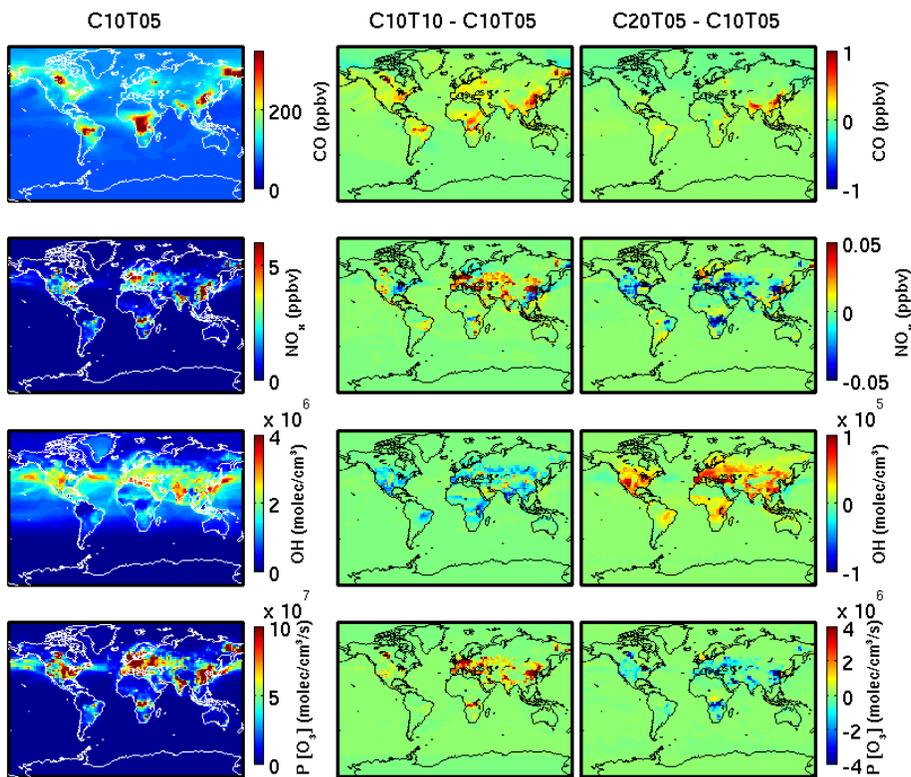


Figure 2a: Sensitivity of simulated species to the duration of chemical and transport operators. The left column contains monthly mean ground-level concentrations simulated with the shortest operator duration considered (C10T05) at $2^\circ \times 2.5^\circ$ horizontal resolution. Other columns contain 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_x), hydroxyl radical (OH), and the production of ozone ($\text{P}[\text{O}_3]$). Simulations are represented as CccTtt with chemical operator duration, C = cc minutes, and transport operator duration, T = tt minutes.

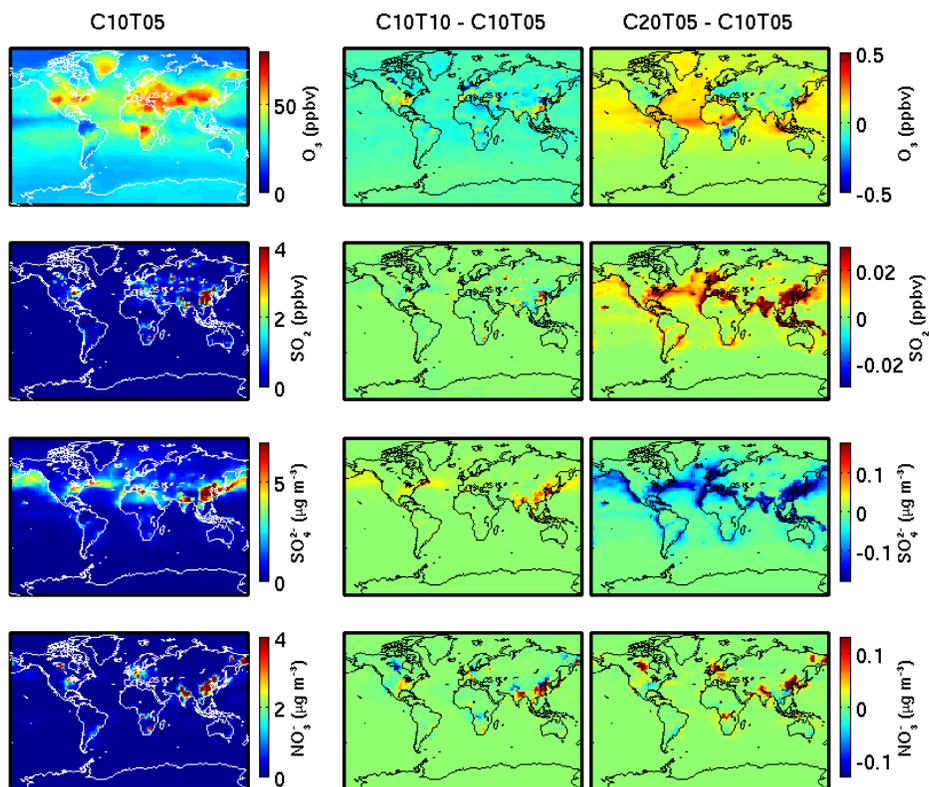


Figure 2b: As described in Fig. 2a, but each row from top to bottom represents ozone (O₃), sulfur dioxide (SO₂), sulfate (SO₄²⁻), and nitrate (NO₃⁻).

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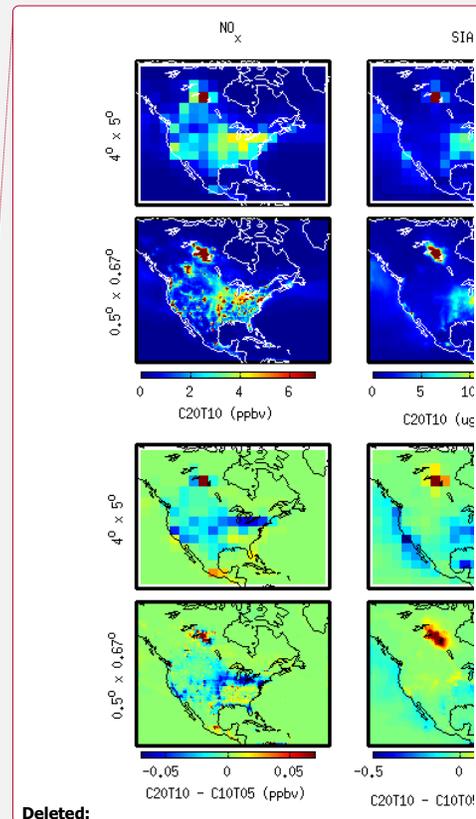
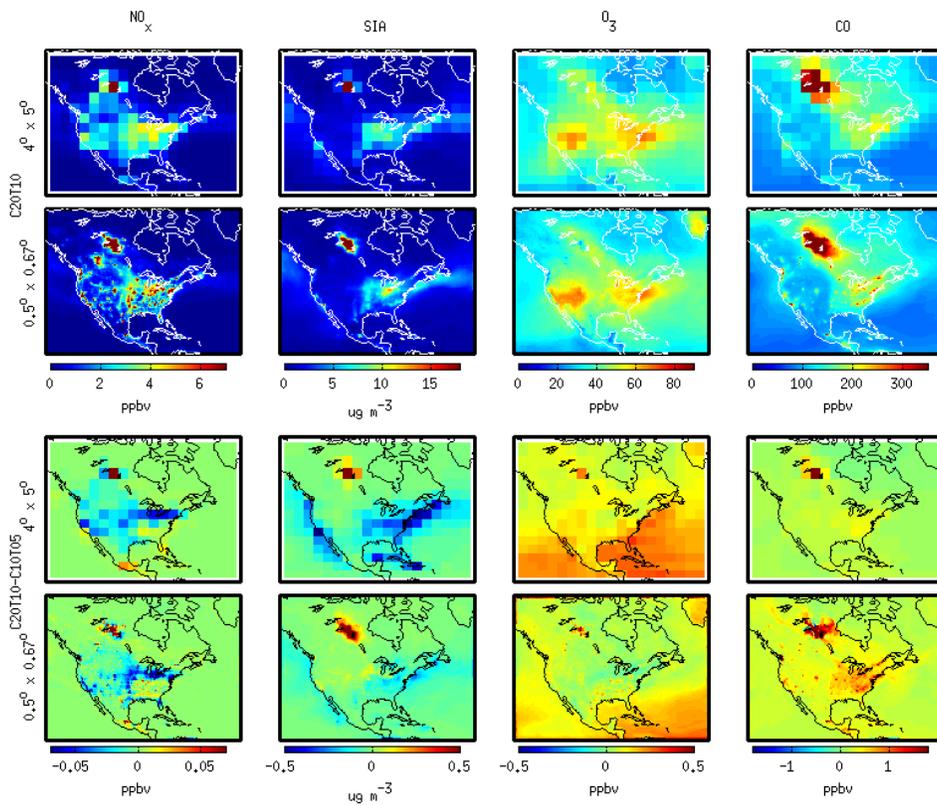


Figure 3: Sensitivity of simulated species to changes in operator duration (C20T10 to C10T05) at two different horizontal resolutions over North America (global $4^\circ \times 5^\circ$, and nested $0.5^\circ \times 0.67^\circ$ simulations). The upper two rows contain monthly mean ground-level concentrations simulated with the C20T10 operator duration for $4^\circ \times 5^\circ$ (top row) and $0.5^\circ \times 0.67^\circ$ (second row) resolutions. The two lower rows contain the monthly mean differences (C20T10 minus C10T05) for $4^\circ \times 5^\circ$ (third row) and $0.5^\circ \times 0.67^\circ$ (bottom row) resolutions. Each column from left to right represents nitrogen oxides (NO_x), secondary inorganic aerosols (SIA), ozone (O_3), and carbon monoxide (CO).

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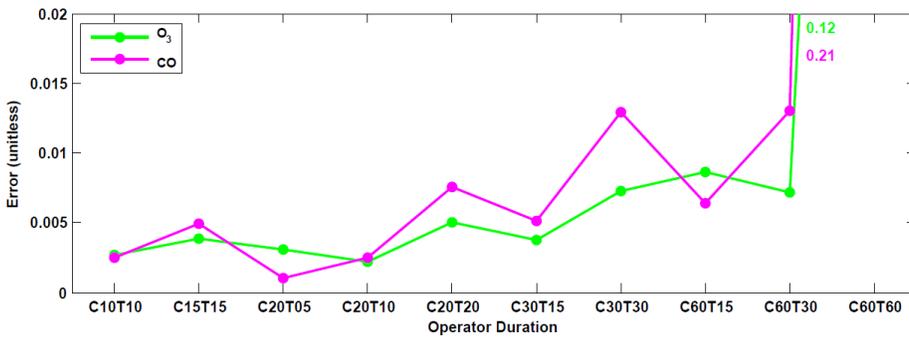
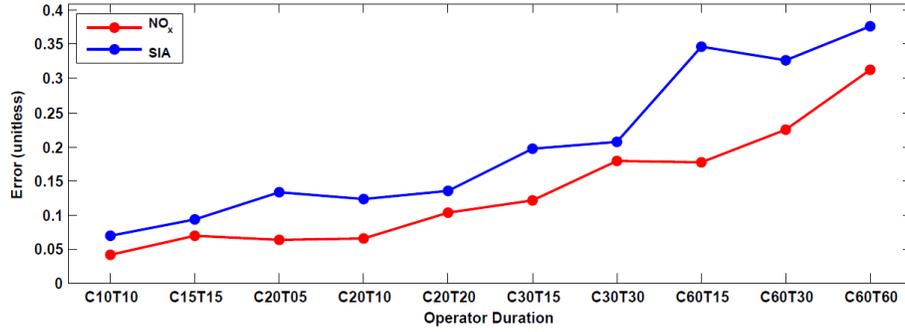


Figure 4: Relative simulation error of different species (E_{sim}^s , eq. 3) with various operator durations at $2^\circ \times 2.5^\circ$ horizontal resolution. Colored lines and dots represent the relative simulation error for nitrogen oxides (NO_x; red), secondary inorganic aerosols (SIA; blue), ozone (O₃; 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.

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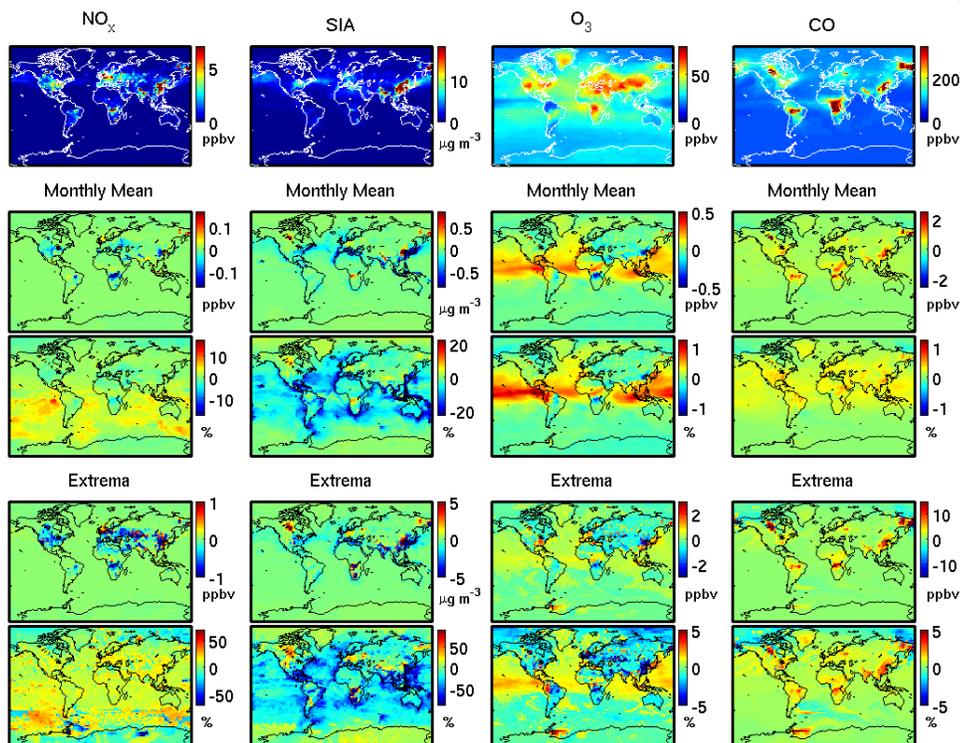


Figure 5: Effect on simulated species of changing from the GEOS-Chem traditional operator durations (C30T15) to the shortest operator durations considered (C10T05). The top row contains monthly mean ground-level concentrations simulated with the C30T15 operator duration at $2^\circ \times 2.5^\circ$ 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).

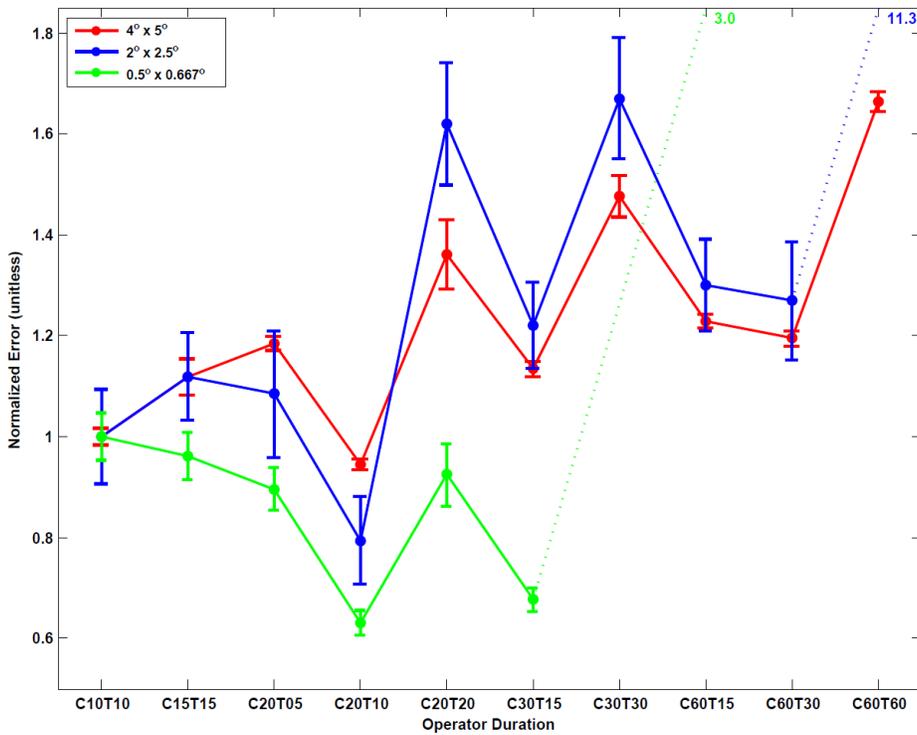


Figure 6: CPU-time adjusted Composite Normalized Error (CNE, eq. 4) for GEOS-Chem simulations with various horizontal resolutions and operator durations. Colored lines and dots represent the CNE for the global simulations at $4^\circ \times 5^\circ$ (red) and $2^\circ \times 2.5^\circ$ (blue), and the 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.