Response to Referee comments # 1

Review of paper Sensitivity of Chemical Transport Model Simulations to the Duration of Chemical and Transport Operators: A Case Study with GEOS-Chem v10-01 by S. Philip, R. V. Martin, and C. A. Keller

This study presents sensitivity simulations designed to understand the variability of simulated concentrations to the model spatial resolution and the temporal integration. These simulations are done using the chemistry-transport model GEOS-Chem v10-01, a state of the art model. The main goal is to determine the best compromise between computational expense and simulation accuracy.

The main motivation for this work is clear and certainly one major concern for atmospheric modellers, often searching for more accuracy and less computational time at the same time.

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.

But the methodology exposed in this paper is very simple and the main conclusions are difficult to understand and thus to consider as robust and acceptable.

In fact, the question could be: is there really an optimal couple between computation and accuracy? This couple exists and a sensitivity study is not needed:

1. for the transport, the CFL must be respected

2. For the chemistry (faster), some error criteria exist and have to be apply. see for examples all papers about the stiff solvers, as one you are citing: Mallet et al 2007, among many others.

Also, obviously: http://acmg.seas.harvard.edu/education/jacob_lectures_ctms_chap3.pdf (section 3.2)

If you consider that you need to have 1 ppb as maximal numerical error, you can use solvers criteria to calculate it. If the criteria is not respected, you just have to automatically increase your time step and rerun until the required precision is reached.

We thank the referee for these comments. However, we believe that these concerns arose from the confusing terminologies of "timesteps" used in the initial sections of the paper (as pointed out by the second referee as major comment-1). The prime focus of this paper was to understand the effects of "operator durations" (not "integration timesteps" used in the advection and chemistry solvers). The range of operator durations (10 min to 60 min) tested in this model at typical horizontal scales from 50 km to 250 km already satisfied the limitations/criterion of these integration timesteps, such as CFL criterion. However, the operator durations from 10 min to 60 min substantially increased the CPU time (as shown in Figure 1). We now use the term "operator duration" to avoid confusion with "integration timestep".

The main concerns are:

The conclusion is to invite modellers to prioritize the fine spatial resolution before the temporal resolution. This conclusion is clearly false. As presented in the paper (introduction), the CFL is the most important limiter for the computational time. In a chemistry-transport model, when increasing the spatial resolution, the time-step will automatically increase too. These two considerations are thus not independent and there is no priority to give between the two.

We have revised this statement to clarify the specificity to operator duration, and to avoid the association with CFL, in lines 294-303, "The simulation error for all species at 4^o x 5^o resolution increases by an order of magnitude compared to 2^o x 2.5^o 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^o x 2.5^o resolution, but not at 4^o x 5^o 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."

Note that the CFL has to be expressed in Δt and Δx (and not the absolute value t and x).

We have eliminated this equation to reduce confusion about the objectives of this paper.

The bibliography is not sufficient: a lot of citations are old (25/72 are used to explain how CTMs are working but are before 2000). Numerous sensitivity studies currently exist to examine this problem and it would be necessary to refresh this bibliography and then the arguments used in the paper.

We have eliminated 7 citations prior to the year 2000 (while retaining those with much historical significance), and have added 17 new citations after the year 2000.

A lack of measurements: In this paper, there is one simulation with a specific accuracy and considered as the reference. How can you be sure this is a 'good reference'? Are you sure that the difference between measurement and the reference is lower than between the sensitivity simulations? i.e that your reference is really good? If this is not the case, the study is not very useful, because you are just calculating differences all far from the reality.

To address these questions, we have rephrased the text, 163-167, "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."

The conclusions about the mass variability of aerosols or the concentration variability of gaseous species is very strange. The main goal of a parameterization is to be conservative in mass whatever is the spatial resolution. If the model has results significantly different when the resolution is 1, 2 or 4 degrees, it means there is a real problem in a scheme (chemistry? deposition?).

We have rephrased these results to clarify the effects of spatial structure (e.g., local emission enhancement) and connect to prior literature, 294-299, "The simulation error for all species at 4^o x 5^o resolution increases by an order of magnitude compared to 2^o x 2.5^o 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^o x 2.5^o resolution, but not at 4^o x 5^o resolution. Nonlinear chemistry at different horizontal resolutions (e.g., Wild and Prather, 2006) also plays a role."

All conclusions are probably valid only for this specific CTM, its strengths and weaknesses. The sensitivity study is not done in a way that some results could be used in another context.

We respectfully contend that the conclusion about operator duration could be applicable to other CTMs. We hope this work will inspire related CTM studies to investigate further.

Details:

p.9590, l.8: why tracers? in the paper, the study examines chemically active species and not tracers.

We replaced the term "tracers" with "species" throughout the manuscript as suggested.

p.9591, l.4: CTM stands for chemistry-transport model (and not chemical).

We acknowledge the fact that there is no universally accepted terminology. Nonetheless, we replaced the term "Chemical Transport Model" with "Chemistry-Transport Model" throughout this manuscript as suggested.

p.9591,l8: why "a" mass continuity equation. Is there different equations? Does it mean that some simplifications are done in the case of GeosChem. If yes, what simplifications?

We were describing the general idea/working of CTMs in one sentence. We changed "a" to "the" in lines 46-48: "Typically, Eulerian models divide the atmosphere into numerous (10^4-10^8) grid boxes and solve the mass continuity equation to simulate atmospheric composition."

p.9594, l2: the First level is very high (130m ASL) and probably not adapted to fast chemistry as above urbanized areas.

Perhaps. However, that is beyond the scope of this study.

p.9594: Reference to (Fairlie et al., 2007): More a sensitivity study than a mineral dust production scheme.

We cite Fairlie et al. (2007) to acknowledge the implementation of mineral dust into GEOS-Chem. This implementation includes the dust entrainment and deposition (DEAD) scheme of Zender et al. (2003) combined with a "source function" as in Ginoux et al. (2001). We added two more references to that sentence (line 137), Zender et al. (2003) and Ginoux et al. (2001).

p.9595, l.11: How this influence is reduced? Is it significant compared to the other variabilities studied here?

This influence is reduced through chemical decay. There is no significant influence for this study since the same initial conditions are used throughout.

p.9596, l.19: To make the sum of the SIA may hide a very high variability due to a chemistry badly solved if the time-step is not correct.

Secondary inorganic aerosol was selected in particular to summarize the impact on chemically active aerosol simulation. We discuss the variability of individual SIA components, sulfate, nitrate and ammonium in lines 225-241 while describing Figure 2b.

p.9597, l.20: "a factor of 4" this means there is no impact of the CFL, but only the impact to have four times more cells. This "linearity" is difficult to understand.

Indeed, the grid cells are sufficiently large that the CFL criteria has no influence on the speed of computation. The number of grid boxes is approximately proportional to the CPU time. We added lines 208-211, "This linearity 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."