Manuscript number: EGUSPHERE-2025-44

Title: Improving the computation efficiency of a source-oriented chemical mechanism for the simultaneous source apportionment of ozone and secondary particulate pollutants

Reviewer comment #2:

Overall, this is a strong paper which demonstrates a new method that has the potential to be useful in the field of study. In this paper you have two key achievements. The development of a method to reduce the number of reactions needed for source apportionment, and the implementation of an EBI solver for source apportionment. One difficulty with reading through the paper was identifying which method was used. I suggest giving a name to your method of reaction reduction, such as ‘relative rate reaction reduction’ (or something shorter), to be able to specify when you have used this method. Otherwise, it is often unclear that both methods have been used.

In addition, it is my understanding that the reaction reduction method does not actually add any error, yet you only show error when including the EBI. Given that this reaction reduction is a new method, I would suggest including some result showing that there is no error from reducing the number of reactions as you have done when the original SMVGEAR solver is used with the new reactions.

Unless my understanding is incorrect, these two methods do not need to be used in tandem, and it seems that the reaction reduction could be implemented relatively easily without major modification to a run, so I suggest highlighting that these methods can be used separately but are complementary of each other.

In your discussion of method error, you show that EBI has a tolerable level of error on average across the area measured. However, I think that more detail would be helpful. The biggest concern when introducing a new solver is the propagation of error over time. Here, your runs only go for 24 hours. It would be helpful to justify why you chose that run length. Furthermore, it would be helpful to include some discussion of how the error changes over time. Does the error increase as the run goes on, or is it relatively stable?

Response: We would like to express our sincere gratitude for the detailed and constructive feedback on our manuscript from the eviewer's positive comments and valuable suggestions. We confirm that all your concerns and suggestions have been fully addressed in the revised manuscript. This includes modifications to the main technical issues, enhancements to the figures and tables, as well as substantial improvements to the clarity and precision of the language throughout the text. Specifically, revisions include clarifications regarding the main purpose of this study, EBI scheme's application and limitations, enhanced explanations of the proposed methods (dual-tagged reaction reduction and successive under-relaxation), corrections to equations and terminology, more detailed analysis of the dynamic under-relaxation factor, and additional discussion of error propagation over time. Below are the point-to-point responses to all of thecomments (The comments are marked in black color and the responses are markedin dark blue color). Note that the following line numbers are shown in the corrected version.

General comments:

Comment 1: I suggest giving a name to your method of reaction reduction, such as 'relative rate reaction reduction' (or something shorter), to be able to specify when you have used this method. Otherwise, it is often unclear that both methods have been used.

Response: Thank you for your suggestion. The name should be 'dual-tagged reaction reduction', and the name has been added in proper places in the text.

Comment 2: In addition, it is my understanding that the reaction reduction method does not actually add any error, yet you only show error when including the EBI. Given that this reaction reduction is a new method, I would suggest including some result showing that there is no error from reducing the number of reactions as you have done when the original SMVGEAR solver is used with the new reactions.

Response: Thanks for the comment. The result comparison of the dual-tagged reaction reduction method is being added as the appendix Figure 1 and highlighted in the manuscript at the end of paragraph 2 of 2.3. (Line 323-326)

Comment 3: so I suggest highlighting that these methods can be used separately but are complementary of each other. (Reaction reduction and implement of source apportionment on EBI, highlighting that these two methods can be used separately but are complementary of each other.)

Response: Thanks for the comment. Yes, these two methods are not necessary to be used simultaneously, however they both contribute to computational cost reduction. The mechanism file generated by the CHEMMECH module from tagged-mechanism reduction method could be directly used by SMVGEAR. The Highlight added to second paragraph of section 2.3. (Line 316-318)

Comment 4:

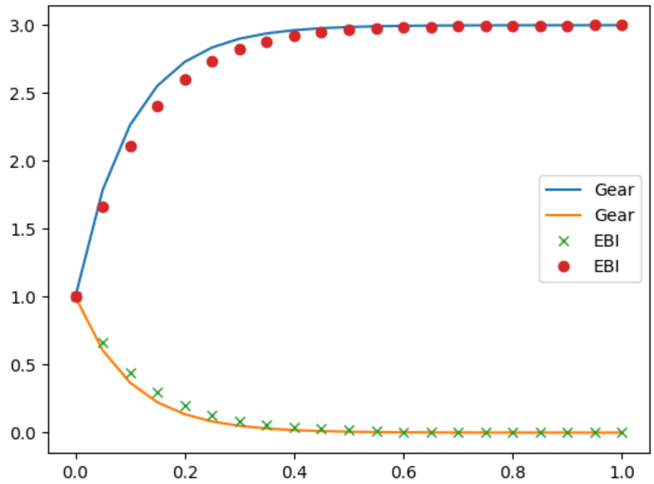
In discussion of method error, you show that EBI has a tolerable level of error on average across the area measured. More detail would be helpful. The biggest concern when introducing a new solver is the propagation of error over time. Here, your runs only go for 24 hours. It would be helpful to justify why you chose that run length. Furthermore, it would be helpful to include some discussion of how the error changes over time. Does the error increase as the run goes on, or is it relatively stable?

Response: Thanks for the comment. In the performance assessment run, totally 3 days simulation has been done while only the first day results were selected to present in the manuscript. The reason for this is we want to inspect the maximum extent of discrepancy of EBI, in comparison to SMVGEAR results, and whether it is acceptable. It was found the error of EBI scheme reduced as flow time increased, and gradually diminished to the range within less than 1% by the end of day 3. The error of EBI is mainly caused by the intrinsic nature of Eulerian backward (fully implicit) method, which generally produces a slower dynamic response in comparison to the real solution.

Here is a simple example of the solution predicted by EBI and the real solution (SMVGEAR), of the two first order problems:

(1) $dC/dt = -kC$, $k=10.0$, $C_0=1.0$; (2) $dC/dt = P - kC$, $k=10.0$, $P=30.0$, and $C_0=1.0$.

It can be seen in the dynamic stage, the solution by EBI algorithm is slower, in both natural decay and external production term cases. Prediction from EBI approach to the real solution as the t increases but steady state values of these two are equal. As EBI results are always slower, in the decaying case the EBI predictions are higher and vice versa for the climbing case.



The initial condition used for the test run was a real concentration field derived from a normal CMAQ run with seven days spin up period rather than a clean default IC, the positively tagged species (NO_X1-NO_X6, NO_2_X1-NO_2_X6…) are effectively zero, but with anthropogenic emission while the null tagged species (NO_X0, NO_2_X0…) are orders of magnitude higher but have no emission source. The evolution of the former is an accumulation process, and the later is a natural decay process. From the Fig 4 it can be seen that the concentration of null-tagged (IC/BC column) of EBI are higher than the SMVGEAR results (underneath the line of symmetry, especially for NO and NO_2), and the positively tagged species of EBI are lower. This is consistent with the above simple case illustration, and the deviation in total concentrations of are inconspicuous as the higher null-tagged concentration of EBI prediction compensates for the lower positive-tagged results. As the flow time increases, the concentrations of positive-tagged and null-tagged species will reach a new stable state, that is oscillating within a certain bound corresponding to the emission intensities, the errors will eventually diminish to a very small level (most of them are less than 1%). Revisions made by appending additional sentences to the end of the third paragraph section 3.2, also additional data regarding the error on the second and third day has been inserted into Table 3 to show the decaying trend of error in EBI.

Comment 5: typo, should be fails to converge

Response: Thanks for the comment. The words"fail to converge" changed to "fails to converge". (Line 130)

Comment 6: built-in

Response: Thanks for the comment. The words "build-in" has been changed to "built-in". (Line 139)

Comment 7: Line 142-145: Need to rewrite this sentence, it is missing some words and there are grammatical issues.

Response: Thanks for the suggestion. The sentence has been modified to: “The method for improving the efficiency of the source-oriented mechanism through simplification of reaction representation and modification of the EBI ODE solver for source-oriented nitrogen species is described in Section 2. Section 3 details the testing of the improved mechanism and the source-oriented EBI solver.” (Line 157–161)

Comment 8: Line 193: Period instead of semicolon

Response: Thanks for the comment. The punctuation "semicolon" has been changed to "period". (Line 212)

Comment 9: Line 279-282: Rephrase, confusing sentence.

Response: Thanks for the comment. The wording should be a little clearer and the sentence has been rewritten to “As the primary goal of this paper is to evaluate the efficiency of the gas phase algorithm, aerosol results despite being enabled in the simulations along with cloud processes, are not included in the analyses described below.”. (Line 302-304)

Comment 10: Table 1:# of species and # of reactions are rows without values

Response: Thanks for the comment. The number of species and reactions have been added into Table 1. (Line 330)

Comment 11: Section 2.2.2 Successive under-relaxation. Has this phenomenon been observed in prior EBI papers, or is this a new phenomenon. Additionally, did you come up with the relaxation coefficient or was that defined previously as well. How does it effect the time dependent concentration of species that are not at equilibrium?

Response: Thanks for the comment. The under-relaxation method is commonly used in the computational fluid dynamics (CFD) calculations to eliminate the inherent numerical instability, and the for fixed α value 0.8 is commonly used in commercial CFD codes, for scalars such as concentration or energy, and this study adopts such a concept. The successive under-relaxation is introduced for the first time in this study, to achieve quicker convergence for source-tagged mechanisms by variant α value to suit the different iteration stages, and it has not been reported in prior EBI papers. From the printout of solution versus iterations, the predicted solutions without under-relaxation showed a two-phase behavior: a rapid shift towards the true solution's proximity in the initial iterations, followed by oscillations around true solution before convergence, and fixed-point iteration frequently occurs due to numerical instability in source-tagged case. Therefore, in the very first steps larger α is favorable as it shoves the predictive concentration rapidly to true solution and subsequent smaller α is used to damp out the overshoots and avoid numerical instability. Performance with the optimal fixed α remains inferior to the dynamic α scheme due to the absence of an initial thrust. The time-dependent concentration of species is independent of the chosen α, which solely affects the number of iterations required for convergence, not the final converged solutions, whether the simulation is time-dependent or at equilibrium. Also, the statement in 2.2.2 and Eq.11 are apparently wrong, solution for each iteration is updated with a weighted average of predicted value and previous iteration, not by values of current and previous time-step. Thus, a declaration has been inserted to the second paragraph of section 2.2.2. Corrections were made to errors in the first paragraph of section 2.2.2 and Eq. 11. (Line 279-285) Furthermore, a more detailed explanation of the successive under-relaxation method was included in an additional paragraph at the end of section 3.1. (Line 398-409)

Comment 12: Table 3: Is there a time-dependent element to the errors? Do they increase over the course of the run. One concern that is not yet addressed is whether or not this method is stable for longer runtimes, and it is not clear from the results whether the error increases over time.

Response: Thanks for the comment. As the concentration field is stable in correspondent to the external inputs (emission intensity etc.) the error will eventually be within a range. Revisions made at the end of paragraph 3, section 3.2. Also, error results for day 2 & 3 are added to Table3. "For all these species, the maximum normalized error among all grid cells is less than 15% and the mean normalized error does not exceed 4% on the first day. Subsequently, the error gradually decays over the following two days, reaching an order of magnitude of 0.1% to 1% by the third day. This indicates that the accuracy of the source-oriented EBI scheme is acceptable, as the errors are anticipated to diminish further with increasing flow time." (Line 426-431) and Table3. (Line 432)

Comment 13: Figure 3: Include a dotted line for the 1:1 matching as you have in Figure 4.

Response: Thanks for the comment. The 1:1 baseline has been added to each sub-plot of Figure 4.

Comment 14: Suggestion for Figures 3 and 4: stating in the caption which method is baseline and which is new, switching the axis.

Response: Thanks for the suggestion. Adding a description to the legend whether old or new would aid reading. Plots with the new method as the vertical axis are more in line with common sense."New" and "baseline" have been added to the caption of Figures 3 and 4, and axis have been changed to EBI as the Y axis and SVMVGEAR as the X axis.

Comment 15: All CMAQ results figures: please include a brief description of the run in the caption. For example: Results from a one-day CMAQ simulation with XX resolution and meteorological inputs from WRF. I know that these details are given in section 2.3, but from looking at the figures alone, key details are missed.

Response: Thanks for the comment. In all CMAQ result plots, brief description of the run has been added in the caption.

Comment 16: Figures 6 and 7: Consider showing the difference between gear and ebi in a third column, as it is difficult to quantify the differences visually.

Response: Thanks for the comment. A third column has been added to Figures 6 and 7 to show the difference between GEAR and EBI.

Comment 17: SMVEAR typo.

Response: Thanks for the comment, there is a spelling error here that needs to be corrected. The word "SMVEAR" has been changed to "SMVGEAR". (Line 484)

Comment 18: Line 449-451: sentence should be rewritten for clarity.

Response: Thanks for the comment and sorry for the mistake. It is confusing that 'Test cases based on the Texas Air Quality Study 2006' appears in the conclusions for the first time. This phrase, discussing reported study results, was included in error. And it has been moved into introduction part with reference. (Line 104-108)

Comment 19: Line 456: what are "adverse meteorological conditions"? Consider rephrasing.

Response: Thanks for the comment, adverse meteorological conditions means weather patterns that exacerbate pollution, like the stable inversions that trap winter fine PMs or the hot, sunny conditions that enhance summer ozone production. The sentence has been modified to: "under meteorological conditions that exacerbate pollution". (Line 520-521)