Response to Referee #1 (GMD-2023-22)

We Thank Reviewer for his/her constructive comments.

Responses to the comments:

Comments: This resubmitted manuscript (ID: GMD-2023-22) has been well improved from the previous version (ID: GMD-2021-259), especially in terms of the explanation of the QDA method. Now we can clarify the differences and/or relationship between the QDA method and previous methods such as SAA, FS, and IPR. However, I have fundamental questions about the combination of QDA and IPR as described in Section 2.2. In my understanding, the component of "wetdep" in IPR (I believe this term is corresponded to "CLDS" in CMAQ's IPR; https://www.cmascenter.org/cmaq/science_documentation/pdf/ch16.pdf) includes wet deposition process and the aqueous-phase chemistry which is highly important in the sulfate aerosol production. If so, it cannot be separated as Eqs. (26) and (27), because "wetdep" term can be attributed both in M and C in QDA. As replied in the previous review process, the aqueous-phase chemistry is included in the "gaschem" in this case, right? The current manuscript is still not clear regarding this point. If the readers know CMAQ IPR, the current description will lead to confusion. In addition, as listed in Table 5, "gaschem" was also continuously zero through this analyzed period. Did this stand for no production via the aqueous-phase chemistry in all four stages? In this case, what is the pathway of sulfate aerosol? The term "ISORR" seems to be the main component of C in QDA; however, I do not follow why the sulfate production is attributed to "ISORR". At the current quality, it is required for furthermore revisions in Section 2.2 and Table 3 to point out what stands for each IPR component, and a more in-depth discussion of the production process.

Reply: Thanks for this comment. I would reply in the following parts.

Comment 1: For the question about the "wetdep" cannot be separated as Eqs. (26) and (27), because "wetdep" term can be attributed both in M and C in QDA.

Reply: The component of "wetdep" in CMAQ's IPR includes wet deposition process and the aqueous-phase chemistry, while the component of "wetdep" in NAQPMS's IPR only includes wet deposition process, so "wetdep" in our study can only contribute in M.

Comment 2: As replied in the previous review process, the aqueous-phase chemistry is included in the "gaschem" in this case, right? The current manuscript is still not clear regarding this point. If the readers know CMAQ IPR, the current description will lead to confusion.

Reply: We feel sorry that we did not provide enough description on the difference between CAMQ's IPR and NAQPMS's IPR and we have revised the manuscript accordingly. In the revised manuscript, this issue has been supplemented in section 2.2 as: "It should be noted that the aqueous-phase chemistry is calculated in the gas chemistry module ("gaschem" in Table 3) of NAQPMS, while the aqueous-phase chemistry of CMAQ is calculated in the wet deposition module, which may lead to different results of IPR in different models."

Changes in the manuscript: lines 286-288.

Comment 3: In addition, as listed in Table 5, "gaschem" was also continuously zero through

this analyzed period. Did this stand for no production via the aqueous-phase chemistry in all four stages? In this case, what is the pathway of sulfate aerosol? The term "ISORR" seems to be the main component of C in QDA; however, I do not follow why the sulfate production is attributed to "ISORR". At the current quality, it is required for furthermore revisions in Section 2.2 and Table 3 to point out what stands for each IPR component, and a more in-depth discussion of the production process.

Reply: In Table 5, "gaschem" was also continuously zero because what we analyzed in this study is the quantity of $PM_{2.5}$ produced directly by each process. For example, aqueous-phase chemistry in "gaschem" produce liquid sulfuric acid (this step would not increase the amount of $PM_{2.5}$), and these liquid sulfuric acid should then undergo gas-particle partitioning to produce sulfate particles (this step would increase the amount of $PM_{2.5}$). In NAQPMS model, the gas-particle partitioning process is included in "ISORR" while the aqueous-phase chemistry is included in "gaschem", which make the term "ISORR" the main component of C in QDA. We have modified the description of the process in section 2.2 and Table 3.

Changes in the manuscript: lines 425-430, lines 1000-1002.