Reply to interactive comments on "Temporary and net sinks of atmospheric CO₂ due to chemical weathering in subtropical catchment with mixing carbonate and silicate lithology" (bg-2019-310)

Responses to Anonymous Referee #1:

Thank you for your time and sincere evaluation for our manuscript. Thank you very much for your constructive comments, and they are very useful for improving our manuscript. We have revised the manuscript according to the suggestions and comments, and the responses to questions one by one are as follows.

Responses to the questions:

<u>Question 1</u>: During the calculation of chemical weathering rates, the authors ignore the anthropogenic origins of major ions except for SO_4^{2-} , show reasons.

Answer 1: Thank you for your question.

There are two reasons.

(1) Two main characteristics of much polluted rivers are that TDS is greater than 500 mg/L and the Cl-/Na⁺ molar ratio is greater than that of sea salts (about 1.16) (Cao et al., 2016; Gaillardet et al., 1999). The TDS in the study area ranged from 73.79 to 230.16 mg·L⁻¹ and the low TDS implied that the anthropogenic origins of major ions could be ignored in the study. The Beijiang River is characterized as a typical region suffered from serve acid deposition (Larssen et al., 2006) and active mining area (Li et al., 2019). The acid deposition and acid mining discharge contribute to the highest concentration of $SO_4^{2^2}$.

(2) Natural origin of SO_4^{2-} is the dissolution of evaporite, such as gypsum, while no evaporite was

found in the study area. because if SO_4^{2-} comes from the gypsum dissolution, the ratios of Ca^{2+} and SO_4^{2-} should be close to 1:1. The stoichiometric analysis showed that the ratio between Ca^{2+} and SO_4^{2-} deviated from 1:1 and also proved this point (Fig.11 in the manuscript and also showed below).

The two reasons have been added in the lines 141-152.

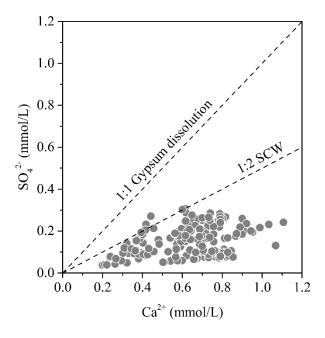


Fig.11 Stoichiometric relationship between Ca²⁺ and SO4²⁻

Question 2: Part 5.1 is too long, may be it is a good idea to separate it into two parts.

Answer 2: Thank you for your suggestion. The part 5.1 has been separated into two parts: 5.1.1 Chemical weathering rates and 5.1.2 Factors affecting chemical weathering.

Question 3: Unify the reference format throughout the paper

Answer 3: Thank you for your suggestion. The format of reference has been modified throughout the paper.

Responses to the specific comments:

<u>Comment 1</u>: Lines 36-37, "regulating the atmosphere-land-ocean fluxes and earth's climate" should be "regulating the atmosphere-land-ocean carbon fluxes and earth's climate"

Answer 1: Thanks a lot. It has been modified in the lines 37-39.

Comment 2: Lines 38-39, delete the "A profound case in point"

Answer 2: Thanks a lot. It has been modified in the lines 41-44.

Comment 3: Lines 54, delete "because"

Answer 3: Thanks a lot. It has been modified in the lines 56-57.

Comment 4: Lines 56, delete "(sulfide oxidation)"

Answer 4: Thanks a lot. It has been modified in the lines 58-59.

Comment 5: Lines 93, change into "it covers an area of 52068 km²"

Answer 5: Thanks a lot. It has been modified in the lines 95-96.

Comment 6: Lines 129 delete "According to the principle of the mass balance"

Answer 6: Thanks a lot. It has been modified in the line 134.

Comment 7: Lines 245 change into "chemical compositions"

Answer 7: Thanks a lot. It has been modified in the lines 265.

Comment 8: Lines 282-284, "Nov", "Jun" and "Feb" should give full names.

Answer 8: Thanks a lot. It has been modified in the lines 305-308.

Comment 9: Line 289, "It is" should be "It was".

Answer 9: Thanks a lot. It has been modified in the lines 313-314.

<u>Comment 10:</u> Lines 450-451, Equations are not labeled a Eq. number. <u>Answer 10:</u> Thanks a lot. It has been modified in the lines 472-473.

<u>Comment 11:</u> Lines 466, "SCW" should give a explain in the Fig. 11 <u>Answer 11:</u> Thanks a lot. It has been modified in the lines 489-490.

<u>**Comment 12:**</u> Line 485, "The result of CCR_{Total} , CCR_{CCW} , CCR_{CSW} and CCR_{NET} were summarized in Table 4" should be "The results of CCR_{Total} , CCR_{CCW} , CCR_{CSW} and CCR_{NET} are summarized in Table 4".

Answer 12: Thanks a lot. It has been modified in the lines 509.

Comment 13: Line 514, "significant influence" should be "significant influences".

Answer 13: Thanks a lot. It has been modified in the lines 544-545.

Comment 14: Line 518, "Runoff manly controlled" should be "Runoff mainly controlled".

Answer 14: Thanks a lot. It has been modified in the lines 549-550.

<u>Comment 15:</u> Lines 530-531, How human activities induced sulfur acid deposition altered the CO2 sinks, increased or decreased?

<u>Answer 15:</u> Thank you for your question. In addition to the chemical weathering induced by H_2CO_3 , sulfuric acid (H_2SO_4) of anthropogenic origins produced by sulfide oxidation such as acid deposition caused by fossil fuel burning and acid mining discharge (AMD) also becomes an important chemical weathering agent in the catchment scale. Many studies have shown the importance of sulfide oxidation and subsequent dissolution of other minerals by the resulting sulfuric acid at catchment scale (Hercod et al., 1998; Spence and Telmer, 2005). Depending on the fate of sulfate in the oceans, sulfide oxidation coupled with carbonate dissolution could facilitate a release of CO_2 to the atmosphere (Spence and Telmer, 2005), the carbonate weathering by H_2SO_4 plays a very important role in quantifying and validating the ultimate CO_2 consumption rate.

Reference

- Cao, Y., Tang, C., Cao, G., Wang, X., 2016. Hydrochemical zoning: natural and anthropogenic origins of the major elements in the surface water of Taizi River Basin, Northeast China. Environmental Earth Sciences, 75(9): 811.
- Gaillardet, J., Dupré, B., Louvat, P., Allegre, C., 1999. Global silicate weathering and CO2 consumption rates deduced from the chemistry of large rivers. Chemical geology, 159(1-4): 3-30.
- Larssen, T., Lydersen, E., Tang, D., He, Y., Gao, J., Liu, H., Duan, L., Seip, H. M., Vogt, R. D., Mulder, J., Shao, M., Wang, Y., Shang, H., Zhang, X., Solberg, S., Aas, W., Okland, T., Eilertsen, O., Angell, V., Li, Q., Zhao, D., Xiang, R., Xiao, J., and Luo, J.: Acid Rain in China, Environmental Science & Technology, 40, 418-425, 10.1021/es0626133, 2006
- Li, R., Tang, C., Li, X., Jiang, T., Shi, Y., and Cao, Y.: Reconstructing the historical pollution levels and ecological risks over the past sixty years in sediments of the Beijiang River, South

China, Science of The Total Environment, 649, 448-460, 2019.

- Hercod, D. J., Brady, P. V., and Gregory, R. T.: Catchment-scale coupling between pyrite oxidation and calcite weathering, Chemical Geology, 151, 259-276, 1998.
- Spence, J., and Telmer, K.: The role of sulfur in chemical weathering and atmospheric CO₂ fluxes: Evidence from major ions, $\delta^{13}C_{DIC}$, and $\delta^{34}S_{SO4}$ in rivers of the Canadian Cordillera, Geochimica et Cosmochimica Acta, 69, 5441-5458, 2005.