Response to reviewers' comments

Response to Reviewer #1:

[Comment] First, I want to apologize for a very late review. I found the authors responses to my comments convincing for the most part and I am happy with the current version with the exception of two small points to look into. Note that line numbers refer to the version with tracked changes.

Which silicate weathering flux is relevant?

Not having played too much with models myself, I did not realize until reading through reviewer 2s comments that the silicate weathering flux that was chosen as a benchmark are based on the Ca + Mg fluxes from silicate weathering only. I think that has to come at the very beginning of the paper in the introduction (e.g. around L 55).

My main confusion is: I believe the estimate by Gaillardet et al. (1999) (2.5×10^{12} mol/yr) does not include volcanic islands with basalts that may be 30% (or more) of the total flux (Dessert et al., 2003; Gaillardet et al., 1999). Isn't that part included in Park et al. (2020)?

Role of lithology:

With regards to my comment on the importantce of lithology. In the responses, you said that taking out ROCK reduces the R2 from 0.81 - 0.78. That seems to be quite a small change. Is that the adjusted R2? If not, I'd be interested to see the change in the adjusted R2 values which may be even smaller.

I am not too familiar with the significance of the MAE change. For all of the variables that you excluded before. Did this statistic show no MAE change? Could it be that this statistic always shows some change, and the absolute value matters (like by including more variables one usually always gets some improvement of R2 - hence the use of adjusted R2)

Response: We thank the reviewer for identifying two places where the manuscript and our previous response needed to be clearer and more quantitative.

1. **Which silicate weathering flux is relevant?**

We have revised the Abstract, Introduction, Methods, and Discussion to state explicitly that the 2.5×10^{12}mol yr^{-1} comparison value is the Ca+Mg silicate flux from Gaillardet et al. (1999), not the total terrestrial silicate-derived alkalinity flux.

We also agree that basaltic weathering had to be treated more carefully. The Gaillardet et al. (1999) large-river benchmark underrepresents small volcanic islands and other basalt-dominated coastal catchments, although basaltic material may occur within some large mixed basins. Our previous wording could be read as saying that basaltic weathering is absent from MErSiM. That is not the case.

First, we would like to clarify that, although we have shown the range of Gaillardet et al. (1999)'s estimates for global Ca+Mg weathering fluxes in gray shading in Figure 9, this global flux of 2.5×10^{12} mol/yr was not the benchmark we used when optimizing the model parameters. Our optimization objective was to minimize the discrepancy between simulated weathering and observed fluxes over 81 river basins (from Gaillardet et al. (1999) and HYBAM, see Figure 10 in the manuscript for details).

Second, the GLiM basic volcanic rock (VB) is included in the mafic lithology class, and the mafic class has the largest Ca+Mg concentration proportionality constant (k_d) of 10317 mol m^{-3} compared to other 5 lithologies (for example the k_d is 1521 mol m^{-3} for felsic lithology and 4759 mol m^{-3} for intermediate lithology). k_d scales the chemical dissolution rate K, which depends on both runoff and temperature:

$$K = k_d(1 - e^{-k_w q})\, e^{\left[\left(\frac{E_a}{R}\right)\left(\frac{1}{T_0} - \frac{1}{T}\right)\right]}$$

This parameterization permits a higher contribution of basaltic rock to the global silicate weathering Ca+Mg flux. Because the dependence of dissolution rate K on runoff q and temperature T is scaled by k_d , the model retains the climatic controls emphasized by Dessert et al. (2003). Therefore, MErSiM includes basaltic/mafic lithologies in the global Ca+Mg calculation. Park et al. (2020) have also included basaltic weathering in this way.

To make this quantitative, we added a new diagnostic calculation. Basic volcanic rock (VB) occupies 4.82×10^6 km^2, corresponding to 3.77% of the lithology-classified land area, and contributes 0.78×10^{12} mol yr^{-1}, or 25.0% of the total MErSiM Ca+Mg flux. The broader mafic group used by the model (basic volcanic and basic plutonic rocks, VB+PB) occupies 5.68×10^6 km^2, or 4.44% of the lithology-classified land area, and contributes 0.94×10^{12} mol yr^{-1}, or 30.3% of the global MErSiM flux.

For global weathering flux, MErSiM predicts $\sim3.1 \times 10^{12}$ mol yr^{-1}, 24% higher than the 2.5×10^{12} mol yr^{-1} Gaillardet et al. estimate. Because the model was evaluated against the 81-basin Ca+Mg dataset rather than tuned only to the global Gaillardet value, this 24% difference is plausibly related in part to the contribution of basalt weathering.

2. Role of lithology

For the ROCK predictor, we apologize for the ambiguity in the earlier lithology comparison. The value 0.81 refers to the six-predictor baseline experiment without PWET, whereas the selected seven-predictor MErSiM erosion model has validation $R^2 = 0.85$. When ROCK is removed from the selected seven-predictor configuration, the validation R^2 decreases to 0.78. Using the validation-set sample size of approximately 745 and the standard adjusted-R^2 formula, these values correspond to adjusted R^2 values of 0.849 for the seven-predictor model and 0.778 for the

ROCK-removed model. Even under the more conservative comparison to the six-predictor baseline, the adjusted R^2 decreases from about 0.808 to 0.778. Thus the penalty for removing ROCK is not due to reducing the number of predictors.

We have also clarified the MAE-based permutation test. This test does not reward a model merely for having more variables. Instead, it evaluates the already-trained model on held-out data after one predictor is randomly given. A positive MAE change therefore means that destroying the information in that predictor increases prediction error. All seven predictors produced positive MAE changes, and ROCK ranked above RUNOFF and LAI in this test.

Response to Reviewer #2:

[Comment] The authors have done a nice job of answering my questions and critiques of the manuscript. I have only one outstanding concern. This involves the target global silicate weathering flux.

My understanding is that the paper targets 2.5×10^{12} mol C/yr as the global silicate weathering flux. This is based upon the 2.5×10^{12} mol Ca + Mg/yr that Gaillardet et al. 1999 report as the Ca + Mg flux to the ocean. As the authors note in their response, this number is substantially lower than the total global silicate weathering flux reported by both Gaillardet et al. (1999) and Moon et al. (2014), which is approximately an order of magnitude higher.

The authors justify this lower rate in two ways: (1) they note that it is only CO2 consumption due to Ca and Mg-silicates that result in removal of C from the ocean-atmosphere on geological timescales (and that CO2 consumption should be 1:1 with the Ca and Mg flux on geological timescales) and (2) Park et al. (2020) uses a similar approach.

I disagree with this approach fairly strongly. It seems bizarre to assume--in a land-based weathering model--how the alkalinity that is supplied by the terrestrial surface will be removed from the ocean. Built into this 2.5×10^{12} mol C/yr term is the assumption that all of this alkalinity will be removed by carbonate burial. That may be a decent assumption for Ca-derived weathering in the modern world, and it may be an ok (?) assumption for Mg-derived weathering (the output of Mg is poorly constrained, and Mg may go into phyllosilicates, resulting in no net burial of C), but it is probably a poor assumption for many parts of Earth history, particularly in those periods when reverse weathering may have played a much greater role in setting steady-state climate and ocean chemistry. Since one goal of the model is to study deep-time Earth, this assumption that is baked into the model will severely limit its usefulness, since to couple this model to any sort of long-term carbon cycle model will require the user to kind of back-out the alkalinity flux that is not partitioned to carbonate burial (but this alkalinity flux can only be guessed at in the current model framework). Similar points could be made about the Na and K-associated alkalinity flux, since the ultimate sink for these cations will help to determine the long-term carbon balance. I should note that, one reason why Park et al. (2020) can get away with this treatment of the weathering system is that their model is really only focused on the late Neogene, when reverse weathering was likely minimal. But extending their model further back in time would present the same problems.

However (and I hadn't appreciated this until this round of reviews), the model also does not apparently attempt to estimate the basaltic weathering flux, which is ~30% of the total flux and does release substantial amounts of Ca and Mg. I presume this is because this are few erosion rate data for basaltic terranes; nevertheless, this is a critical flux that may be quite climate sensitive (does this explain the weak climate sensitivity in the paper?) and may have changed proportions rather dramatically over Earth history, particularly in relation to different climate zones.

With all of that said, these are all clearly decisions that were made to keep this model simple, and, having built plenty of models myself, I understand the need to draw boundaries in model development. However, the authors really need to be clear about the limitations that these decisions impose on this model and these limitations need to be stated upfront, both in the methods (why chose this target global silicate weathering rate; why ignore basalts) and in the discussion. I would certainly not expect to pick up a paper advertising a new global model of terrestrial silicate weathering only to learn that the model does not attempt to parameterize 30% of the flux and bakes in an assumption about the sinks of alkalinity in the ocean in their calculation of the global terrestrial silicate weathering flux. Fine if the authors want to continue this way, but these model decisions need to be clearly explained.

Response: We appreciate this careful and very insightful criticism. We agree that our earlier response was too brief and that the manuscript needed to separate three issues that had been conflated: (i) what flux is used as the model-data benchmark, (ii) whether basaltic lithologies are present in the model, and (iii) whether the terrestrial Ca+Mg flux can be treated as a complete long-term carbon sink without specifying ocean alkalinity sinks.

First, we clarified the benchmark. Although we have shown the range of Gaillardet et al. (1999)'s estimates for global Ca+Mg weathering fluxes in gray shading in Figure 9, this global flux of 2.5×10^{12} mol/yr was not the benchmark against which our model parameters were optimized. Our optimization objective was to minimize the discrepancy between simulated Ca+Mg weathering fluxes and the 81 large river basin observations (from Gaillardet et al. (1999) and HYBAM, see Figure 10 in the manuscript for details).

We now describe the estimates of Gaillardet et al. (1999) as a large-river Ca+Mg reference rather than as the total global silicate-weathering flux. We have therefore revised the manuscript to state that the output of MErSiM should be treated as a terrestrial Ca+Mg source term. The oceanic partitioning of that alkalinity among carbonate burial, reverse weathering, Mg incorporation into secondary minerals, and other sinks must be prescribed or modeled externally, in order to know how much atmospheric CO_2 is eventually consumed, especially for deep-time applications.

Second, we corrected the statement about basaltic weathering. MErSiM does include basaltic lithology: GLiM basic volcanic rock (VB) is included in the mafic lithology class, and the mafic class has a highest Ca+Mg concentration proportionality constant (k_d) of 10317 mol m^{-3} compared to other 5 lithologies (for example the k_d is 1521 mol m^{-3} for felsic lithology and 4759 mol m^-3 for intermediate lithology). k_d scales the chemical dissolution rate K that is dependent on both runoff and temperature:

$$K = k_d(1 - e^{-k_w q})\, e^{\left[\left(\frac{E_a}{R}\right)\left(\frac{1}{T_0}-\frac{1}{T}\right)\right]}$$

This parameter sets permit a higher contribution of basaltic rock in global silicate weathering flux, and also emphasize the role of climate (runoff q and temperature T) on basaltic weathering with a larger scaling factor, as has mentioned in Dessert et al. (2003).

We now added a diagnostic calculation showing that VB covers 4.82 × 10^6 km^2 (3.77% of lithology-classified land area) and contributes 0.78 × 10^{12} mol yr^{-1} (25.0% of the global MErSiM Ca+Mg flux). The broader mafic group (basic volcanic rock + basic plutonic rock, VB+PB) contributes 0.94 × 10^{12} mol yr^{-1} (30.3%). The basaltic rock contribution in MErSiM Ca+Mg flux is surprisingly similar to the estimate of the basaltic rock contribution in global CO_2 consumption flux (30% - 35%) of Dessert et al. (2003), even though they have used a different modeling scheme detailed below. Therefore, we would like to state that basaltic/mafic lithologies are not absent from our flux calculation, and their inclusion plausibly helps explain why our simulated total (~3.1 × 10^{12} mol yr^{-1}) is higher than the Gaillardet et al. (1999) benchmark (~2.5 × 10^{12} mol yr^{-1}) by 24%.

We also checked the erosion model. In the training dataset, the propotion of basic volcanic rock (ROCK=2) is quite limited as the reviewer has noticed: 62 of 3706 basin samples (1.67%), but it has not been completely neglected. The erosion-rate distribution of these samples is highly skewed, with a median of 11.4 mm kyr^{-1} and a mean of 123 mm kyr^{-1} because a small number of high-erosion settings raise the average. This supports a more precise statement: the erosion model is not blind to basic volcanic terrains, but the basaltic subset is small and should not be overinterpreted as a complete representation of island basalt weathering with fast erosion. We are expecting more basalt erosion data to constrain the current analysis.

Third, we now state more clearly what MErSiM does not do, and conduct more experiments to identify the model's limitations. Although basaltic rocks are included through the lithology map and the mafic parameter, MErSiM v1.0 does not include a separate basalt-weathering law. Specifically, Dessert et al. (2003) compiled several small basaltic catchments and volcanic provinces, found that basaltic rivers have high Ca/Na, Mg/Na, and HCO3/Na ratios, and proposed a strong runoff-temperature control on basalt weathering. They have used regression method and found that the specific atmospheric CO_2 consumption rate f_{CO_2} (mol km^{-2} yr^{-1}), is

$$f_{CO_2} = R_f \times 323.44 e^{0.0642T}$$

Where R_f is runoff (mm/yr) and T is temperature (℃), with a regression $R^2 = 0.7$.

The weathering scheme we have adopted in MErSiM v1.0 is from Gabet and Mudd (2009), which has used the compilation of weathering flux from West et al. (2005). We examined the raw dataset and found that it indeed did not include basalt-bearing watersheds. However, we couldn't help but wonder whether this transport/kinetics-limited model framework could, to some extent, describe the characteristics of basalt weathering observed by Dessert et al. (2003).

If this hypothesis holds true, the following experimental results would be expected: (i) The simulated weathering fluxes for basalt weathering zones would align with the relationship identified by Dessert et al. (2003), namely that the ratio of weathering flux to runoff is an exponential function of temperature (as shown in the equation above); (ii) A large portion of the basalt weathering zones would fall within the kinetic-limited range, as climate (temperature and runoff) has a significant impact on weathering flux; (iii) The characteristics of granite (acidic plutonic rock, PA) would differ significantly from those of basalt.

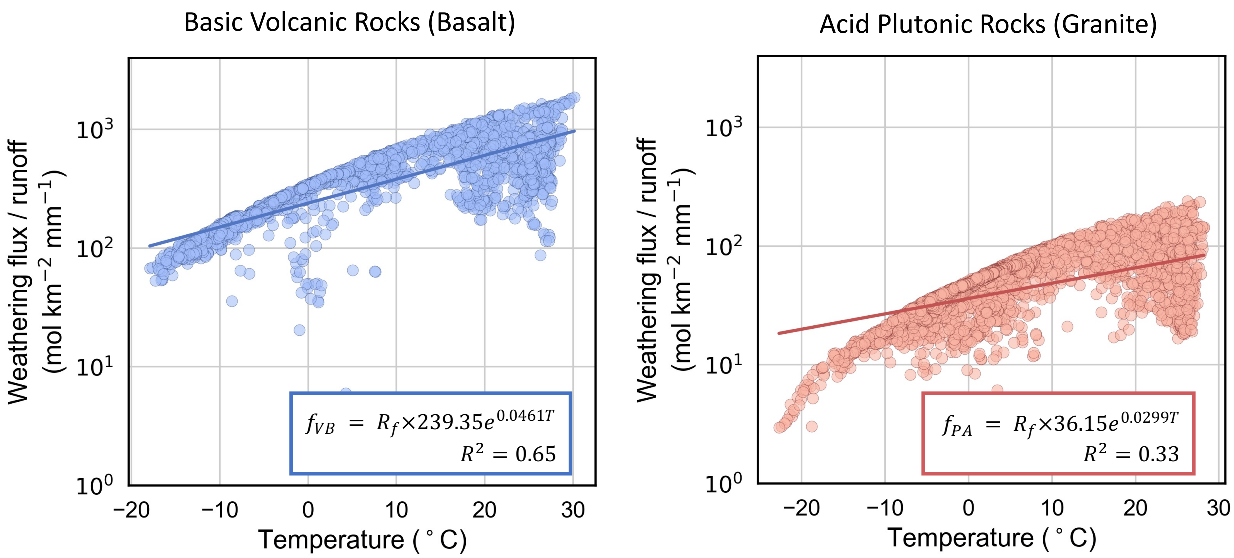

Figure R1 "Dessert-type" relationship between weathering flux and climate. The y-axis represents the ratio of the weathering flux per unit area to runoff. The box shows the mathematical relationship indicated by the straight line and the coefficient of determination R^2. Blue indicates cells with basaltics (Basic Volcanic rocks, VB), and red indicates cells with granites (Acid Plutonic rocks, PA). This is Fig. S6 in the revised Supplementary Material.

We analyzed the weathering fluxes simulated by the MErSiM model and found that in areas with basaltic rock, the simulated weathering fluxes could be described by the product of the runoff and temperature exponential functions, with an R^2 of 0.65 (Figure R1).

$$f_{VB} = R_f \times 239.35e^{0.0461T}$$

This is consistent with the relationship established by Dessert et al. (2003) based on data from a dozen river basins, including the Columbia Plateau and the Deccan Traps, though it should not be interpreted as a direct reproduction of their basalt CO_2-consumption law. Specifically, the fitted coefficients for VB (0.0461 and 239.35) are lower than those of Dessert et al. (2003). This may be because they have used the number of moles of bicarbonate, whereas our model only accounts for the number of moles of Ca and Mg cations. However, we still note that the order of magnitude of these constants is consistent with the results of Dessert et al. (2003). In contrast, weathering fluxes in granite cannot be well described by this relationship with an R^2 of 0.33 (Figure R1), and the sensitivity constants are significantly smaller than basalts (0.0299 and 36.15), highlighting a stronger dependence of basalt weathering on climate.

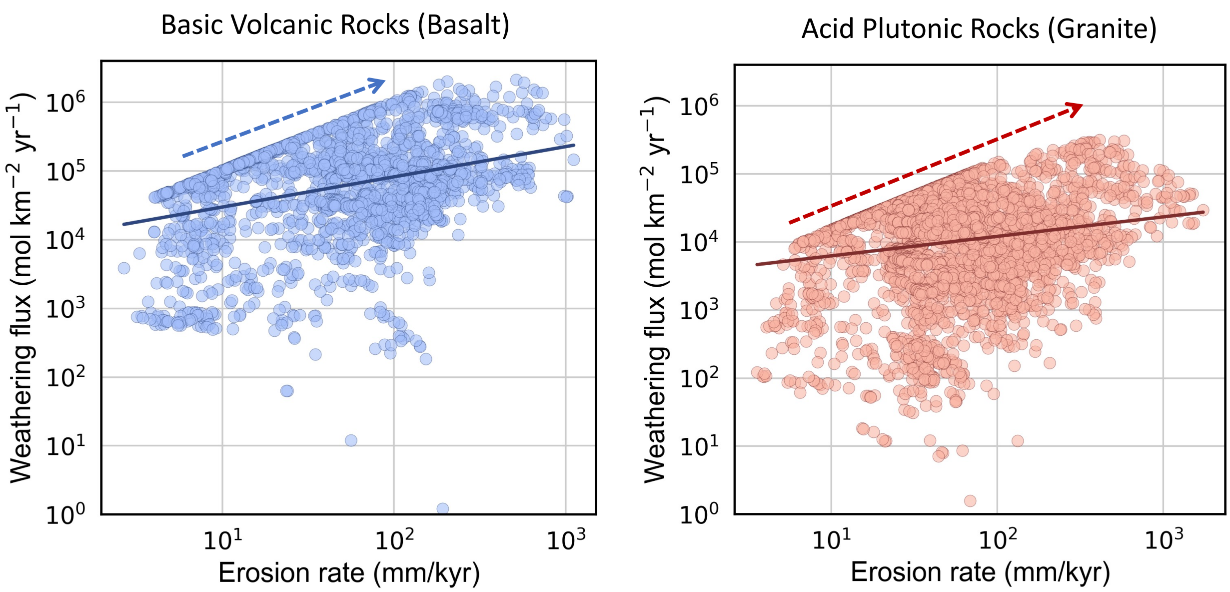

Figure R2 Relationship between erosion and weathering. The fitted straight lines illustrate the power-law relationship. The dashed line with arrow illustrate the limiting effect of erosion on weathering flux. This is Fig. S7 in the revised Supplementary Material.

We also found that in general, weathering flux increases with erosion rate for both basalt and granite (Figure R2). When the erosion rate does not exceed 100 mm/kyr, a significant number of weathering flux are clearly limited by the erosion rate.

Our frequency analysis of basalt erosion rates revealed that, while a considerable number of basalt regions indeed have erosion rates around 100 mm/kyr (in contrast, granite erosion rates are widely distributed around 30 mm/kyr), low-erosion zones with rates around 10 mm/kyr are equally prevalent (Figure R 3), as areas with gentle slopes and low tectonic activity account for a large proportion of the continent's land area. In other words, although many oceanic islands are located in tectonically active zones with mountainous topography and high physical erosion rates—where the supply of fresh rock is not a bottleneck—not all basalt weathering occurs within the kinetics-limited regime. For example, the erosion rates of the Karoo basalt in South Africa's semi-arid, flat areas are extremely low, generally < 4 mm/kyr (Decker et al., 2013). Therefore, island basalt and continental basalt may not be treated as equivalent, as Li and

Elderfield (2013) has suggested when they were trying to estimate the CO_2 consuming rate of continental weathering.

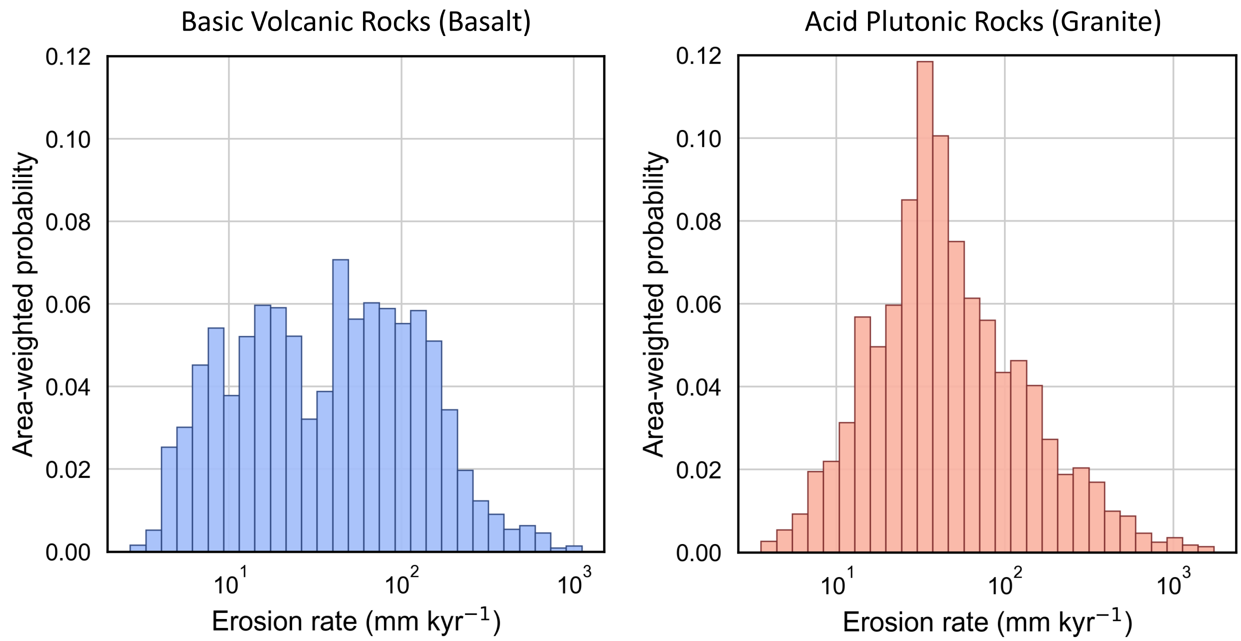

Figure R 3 Frequency Distribution of Erosion Rates. It can be seen that in a large portion of the basalt weathering areas, the erosion rate exceeds 100 mm/kyr, which means that the weathering flux is less affected by erosion. This is Fig. S8 in the revised Supplementary Material.

Moreover, even some of the island basalt weathering is limited by erosion. The erosion module in MErSiM yields lower erosion rates for tropical islands compared to Park et al.(2020) (Figure 5d in the manuscript). The role of basalt in carbon consumption in Southeast Asia emphasized by Park et al. (2020) is partly because basalt is assigned a high dissolution rate in weathering calculations, and partly because their SPIM model highlights the effects of large runoff on erosion rate in these tropical islands. Park et al. (2020) proposed that enhancement of Southeast Asian mafic island weathering was a driver of Neogene cooling, but this increasing flux of unradiogenic mafic is not reconciled with the trend toward more radiogenic Sr and Os isotope values (Caves Rugenstein et al., 2021). We have therefore expanded our discussion of mafic weathering in the revised manuscript to address these issues.

In summary, although the MErSiM v1.0 weathering model framework does not perform separate calculations for basalt weathering, for example the model proposed by Dessert et al. (2003), the results still highlight the significant climate dependence of basalt weathering. This dependency is distinguished from other rock types, and to some extent align with the relationship between basalt weathering flux and temperature/runoff identified by Dessert et al. (2003). At the same time, the MErSiM model framework can also depict the weathering-erosion relationship. A large portion of the basalt weathering basins are still limited by the supply of fresh material. Note that physical erosion is a factor that Dessert et al. (2003) did not discuss, perhaps because the basalt weathering regions lacked field-measured erosion rate data (as mentioned above, samples with beryllium isotope erosion rates from basalt weathering basins are very limited). If more field-measured basalt erosion data could be found, it would be very interesting to compare the erosion-weathering relationships between the continental basalts and island basalts, and of course, between basalts and silicates.

We have now revised the manuscript and supplemental materials to address the discussion above.

References

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