Answer to the reviewers

We thank both reviewers for their insightful and valuable comments. Below are the answer to their queries.

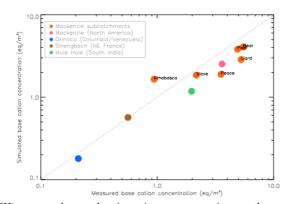
Reviewer 1:

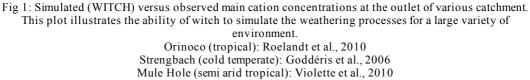
One overarching concern that is not addressed by the authors is the accuracy of the models that they are using for this region, particularly the climate and biospheric models. In previous work, the authors have established that the WITCH model reliably predicts weathering of North American loess over the past 10 kyr, but, as the authors acknowledge in Section 4, the results of their present study are highly dependent on the climatic and ecological predictions, and these come from the ARPEGE and CARAIB models, respectively. It is not clear how well these capture changes in central North America. The reference for the CARAIB model is related to forests in Europe, and the reference for ARPEGE the Mediterranean. Climate models in particular often capture variability in some regions much better than in others. Given the importance of variation in temperature, hydrology, and productivity for predicted weathering, it seems important to have some validation of the predictions that are being used to drive the weathering model for North American loess in this study. Alternatively, in the absence of such validation, it would be beneficial for the authors to acknowledge carefully in the manuscript that the current study is using one set of predictions as a "typical example" of how weathering might be expected to change under changing climate, rather than as definitive predictions.

The main objective of our contribution is indeed to stress that the response of the weathering of the Mississippi loess to changing climate might be complex, and potentially highly variable along the South-North transect that we simulated. The response of weathering to global warming depends on a complex interplay between mineralogical composition, temperature, drainage and continental vegetation. As such, our study can be seen as a "typical example". However, the models used have been validated previously. Three models have been used :, ARPEGE (climate), CARAIB (dynamic biosphere) and WITCH (weathering).

The climate model ARPEGE has been part of the international stretched-grid model intercomparison project (SGMIP). ARPEGE was able to reproduce the climate evolution over North America over 12 years (1987-1998 period). The ref is Fox-Rabinovitz et al. (2006). This will be mention in the revised version. The CARAIB model is a dynamic vegetation model initially developed for the global scale (Warnant et al., 1994). Thus it has been used and validated over all continents. The cited reference (Dury et al., 2011) refers to a specific study focusing on the projection of climate change impacts on European forests and using the latest version of the model. However, the carbon cycle of the model has been validated by Nemry et al. (1996) and Nemry et al. (1999), by using the net ecosystem productivity output fields as boundary conditions to an atmospheric model. This allowed calculating the seasonal cycle of atmospheric CO2 concentration and comparing it with observations at various stations of the atmospheric CO2 network at different latitudes and over all continents. Net primary productivity, biomass and soil carbon content have been validated within all large biomes of the world (Warnant et al., 1994; Otto et al., 2003). Model vegetation distribution maps have also been validated globally by Otto et al. (2003). Here, a new classification of plant functional types (PFT) is used (updated from François et al., 2011), which is valid globally, contrary to the one used by Dury et al. (2011). This classification has been built on the basis of plant species distributions from various parts of the world, including North America. In particular, subtropical PFTs were defined in this new classification on the basis of subtropical species from North America, and especially the Mississippi Valley.

Regarding WITCH, it was recently used to simulate the weathering processes in the Mississippi Valley over the last 10kyrs, and was able to reproduce the evolution of the mineralogical composition of the pedon (Goddéris et al., 2010). Furthermore, it has been used in various environments, from arctic to tropical settings, and was able to simulate the concentration in base cations at the outlet of the simulated catchments (Fig 1).





Mackenzie (cold): Beaulieu et al., 2011 and Beaulieu et al., 2012.

On a related note, the authors could usefully provide a bit more information about uncertainty in their results. It may be difficult to provide quantitative uncertainty estimates, but this deserves at least some consideration. For example, is the change in dolomite weathering rate in the southern pedon, from 1.0 mols/m2/yr to 0.9 mols/m2/yr actually a significant decrease? Or are these values for all intents and purposes the same? It would surprise me if, given the number of different factors that influence weathering, this 10% change is actually meaningful. Some treatment of this kind of question in the text would be helpful. One way to do this might be to run more thorough sensitivity tests than are presented in Section 4, where a comparative simulation is presented that involves no change in air temperature. This is certainly a valuable sensitivity tests in order to better assess the meaning of the projections.

Regarding the specific decrease from 1.0 to $0.9 \text{ mol/m}^2/\text{yr}$, we simply mention a slight decrease, compared to a marked decrease for the northern pedon. Of course, a 10% change might not be significant, the most important point is the difference in the weathering response for both sites, the northern site being characterized by a 60% decrease while the southern site slightly responds to the forcing.

More generally, over the length of the Mississippi Valley, the terrestrial lysocline may deepen at surprisingly various rates, which show that the link between weathering and drainage is not as simple as a linear correlation as often advocated in the literature. We agree that more sensitivity tests can be performed. We tested the temperature effect for two pedons, but these are not the only tests performed in the ms. Indeed, by running the cascade of models for 9 sites covering the whole Mississippi Valley with a uniform mineralogy, we tested the dependence on climate of our simulations. The comparison between these various pedons is the core of the contribution. In some ways, figure 8 (of the ms) illustrates the sensitivity of our main result (the terrestrial lysocline deepening in the future) to climate (the climate change for each site is also plotted on fig 8). We will add this discussion to the revised ms.

The methods could usefully be described in slightly more detail. In Section 2.1, the authors describe the model run by saying they project weathering into the future. To me, this implies starting somewhere around 2012, but the results they show start in 1950. Some clarification on this – and perhaps an explanation of why they decided to start in 1950 – might be helpful.

The reason why we started from 1950 is the following: weathering is not *a priori* expected to respond strongly to climate change. So the longer is the simulated time serie, the larger will be the weathering response. The longer ARPEGE/CARAIB simulations available cover the 1950-2100 period. We thus decided to run the model over the whole period, to maximize the weathering response. Instead of 88 years of simulation (2012-2100), we thus have a simulation period of 150 years, which is 70 % longer. This will be mentioned in the revised version.

Furthermore, each simulation is preceded by a 20-years spin off simulation to relax the initial conditions. This run must be performed under the less anthropogenically impacted conditions, so under the 1950 climatic conditions.

Moreover, I wonder whether there is there any interesting information to be gained from comparing the modeled values for the present day to any observed present-day weathering fluxes for this region (if these exist)?

The ARPEGE climate model is able to reproduce the time evolution of the climatic conditions over North America for the 1987-1998 period (Fox-Rabinovitz et al., 2006).

The geochemical output should be compared to non-polluted soil solutions. Unfortunately, such data are not available. This is a paradox: we have pretty good information on the mineralogy along the Mississippi transect (USGS data), allowing model simulations, but no soil solution data. This is why the model was previously calibrated on long term simulations (10 kyr, Goddéris et al. (2010) to check whether it was able to reproduce the evolution of the mineralogical composition. As the answer is yes, we might expect that weathering fluxes are correctly simulated. This will be mentioned in the revised version.

In terms of methodology, it would also be interesting to know why the authors chose 43% porosity, and what the basis is for assuming that all heterotrophic respiration and 1/3 of autotrophic respiration occur below ground (seems reasonable to me, but why 1/3?).

Bulk densities for peoria loess range from 1.33 to 1.53 g cm⁻³ (Bettis E.A. et al., 2003). Assuming a mean crustal rock density of 2.7 g cm⁻³, this gives a porosity ranging from 51 to 43%. We choose 43 %. The ref is now mentionned in the revised text.

We assume that 1/3 of autotrophic respiration is occurring below ground. This number is arbitrarily choosen. We have no reffor that, this number should depend on many parameters, including the plant species. It seems to us reasonable.

Are there any potential artifacts introduced to the comparison in Section 5 by fixing the initial depth of the dolomite front at 2.8 m? This seems like a reasonable initial assumption, but it would be interesting to know if the results would be significantly different if the initial depth were at 2.0 or 3.5 m, for example, especially since there are poor empirical constraints on the present-day depth to dolomite. The authors argue that this should not be relevant, but without clear evidence. Sensitivity tests might be useful for this.

Results of the simulations are not significantly dependent on the dolomite front depth (it was tested, but not shown). Once the vertical draining waters reach the dolomite front, they rapidly reach saturation with respect to dolomite, whatever the dolomite front depth. Below the dolomite front, waters keep an almost constant chemical composition, fully controlled by dolomite occurrence. Changing the dolomite front position slightly modifies the retreat rate of dolomite (increase if dolomite front is shallower, or decrease if the dolomite front is located deeper, because of pH increase). But this is only marginal. Indeed, most of the pH increase with depth occurs between the surface and 1 m depth, where it increases by about 1 unit (fig. 6 of the submitted paper). Our results might be dependent on the dolomite front depth only if the dolomite front is located above 1 m depth, where pH is highly dependent on depth, but this is not the case (field data from Williams et al., 2010) showing that dolomite occurs well below 1m depth, and from table 1 of our contribution).

The silicate dissolution will be impacted by the location of the dolomite front. However, the contribution of silicate to the atmospheric CO_2 consumption is marginal, as discussed in section 3.2.4 of the ms. We will add a short discussion in the revised version.

In Section 3.2.2, it was not clear to me increasing occurrence of dry events in the south should lead to increasing Na export (end of the first paragraph). It makes sense that the overall greater drainage should increase elemental fluxes, including for Na (end of third paragraph), but the way the first paragraph is written, it sounds like higher Na export is due to the dry periods when albite may stop dissolving. This seems confusing, though I may be missing something here. Either way, it might help to clarify the text.

There is a mistake in the manuscript. Na export is increasing for the Southern pedon from 380 mol/ha/yr (1950) to 470 mol/ha/yr (2100), and not from 85 to 1000. We apologize for this mistake. This will be corrected in the revised version. Despite this mistake, we maintain that Na export is increasing. We will

expand the end of the first paragraph to make this clear, in agreement with the reviewer's query. Despite the increasing occurrence of dry events, drainage is slightly increasing in the South over the period 1950-2100, promoting albite dissolution. At the same time, overall temperature rise forces albite dissolution rate to increase. For these two reasons, Na export is increasing. This will be explicitly mentionned in the revised ms.

At the end of Section 3.2.3, the authors comment that the changing weathering under changing climate may also change the chemistry of soils and soil solutions, so plants have access to different nutrients. Is this considered in the CARAIB biospheric model, or is the information only passed "one way," i.e. from the biospheric model to the weathering model. This might be worth a sentence to clarify. There could be an interesting associated feedback.

There is no feedback from the weathering model on the biospheric model, as mentioned in the abstract and the conclusion (cascade of models). CARAIB is only forced by climate (air temperature, precipitation, sunshine hours, air relative humidity and wind speed), no nutrients are involved in the biospheric model. Indeed, we fully agree that future developments should include a real feedback. We will clarify this point at the end of section 3.2.3 of the revised ms.

Reviewer 2:

While in the introduction the global context is established (e.g. global rates of CO2-consumption rates are mentioned, which is one of the highest one considering recent publications), is it possible to set the results a bit more into the context of global change and the Earth system? I am not sure if this can be done easily. Thus this comment should be seen as a suggestion.

We understand that this may increase the impact of our paper, but we do not see precisely how we can do it better. The context of global change is settled in the introduction, and the conclusion states that our results might be important in terms of ocean acidification, which is one of the most important issue in the global change and Earth system context. So we prefer to keep the text as it is.

You state that continental weathering is besides other factors a function of physical erosion. However, the reference West et al., shows this for felsic lithologies. In the MS carbonate dissolution seems to be in the focus. Has physical erosion an impact on your results? How well are surface hydrological processes covered by the model framework?

Dolomite dissolution is at least 6 order of magnitude faster than plagioclase dissolution. For that reason, it is expected that dissolution of dolomite should not be limited by physical erosion. Dolomite weathering is thus not dependent on physical erosion.

The soil hydrological budget is calculated by the CARAIB global dynamic vegetation model. The soil hydrology model included in CARAIB has been validated (Hubert et al., 1998). Appendix A2 describes how we use the predicted hydrology as an input of the WITCH model. Although still coarse, this method allows accounting physically for the role of land plants and evapotranspiration on the vertical drainage.

It is stated that the multi-parameter dependence of continental weathering makes it difficult to assess the response of continental weathering to climate change. Could you explain why? For example some researchers use simple functions depending on the named parameters to estimate exactly this. Simple functions are also used in the Geoclim Earth system model, if I am not wrong? So it might be useful, and this would shed more light onto the innovations used here, to explain what gaps in knowledge your presented approach closes. For which scales (time and space) are the results relevant or can the outcome of the models be of relevance?

This is an important point. Several compilations, such as Oliva et al. (2003) and Dessert et al. (2003) propose simple phenomenological laws linking runoff and air temperature to weathering rate of silicate minerals. The same method has been largely improved by Hartman et al. (2009). As discussed in Goddéris et al. (2009), the phenomenological laws (transfer functions) are based on spatially distributed data. As such, they can be seen as a snapshot of the weathering system at a given time. If these data are coming from weathering systems more or less at steady-state, they can be used to predict the multi million year evolution of the Earth system, as done in the GEOCLIM model. But at the century timescale and under global change conditions, the weathering system is forced out of steady-state, because the characteristic time of the forcing (century) is much shorter than the response time of the weathering system (typically 10³ to 10⁵ years). In

this case, phenomelogical laws cannot be used because they do not give any insight into the dynamics of the system. This is the reason why complex mechanistic models are required. Furthermore, phenomenological laws only account for runoff, mean annual temperature. In addition, our modeling accounts for the change in vegetation cover, in the soil CO₂ production and vertical diffusion, in evapotranspiration and drainage... Finally, the fine mineralogical composition can be considered in our simulations, while only large lithological classes are considered in phenomenological models. A short discussion about this point will be added.

Loess minerals often have "fresh" unweathered surfaces (P10850, L3-4). However, the loess considered here has weathered already a few thousand years. It may be appropriate to discuss why the term fresh is relevant for this study, or better, why carbonates are less affected by aging than igneous felsic minerals?

The word "fresh" has been used because loess material comes from rocks grinded by Quaternary glaciers. Even if they have been weathered for thousands of years since their deposition, they represent a weathering system much younger than tropical lateritic profiles, which are million of years old. However, we agree that the word "fresh" is confusing. We will replace it by the concept of young weathering profile in the revised ms.

Carbonates are in fact much more affected by aging than felsic minerals, because within 10 kyr, the dolomite front deepened by several meters, while felsic minerals are only slightly depleted at the surface (Goddéris et al., 2010).

Model settings (P10851, L9 to 18): Are geomorphological settings considered, or terrain characteristics? I guess the loess-areas are relatively flat; therefore surface runoff should be of no significance? What would be the role of slope influencing the percolation patterns assumed here?

We do not consider the geomorphological setting. This is beyond the capabilities of our model. We assume flat surface, with only a vertical drainage. This will be clarified in the appendix A2 of the revised ms.

Could a more, steep environment bias the general findings outlined here, or a do changes in strong rain fall events (strength or frequency) lead to a significant change of the proportion of surface runoff or a decreased proportion of water percolating to the relevant weathering zones discussed?

As stressed in the study, weathering of the loessic profile is heavily dependent on vertical drainage. If the proportion of surficial runoff increases at the expense of vertical drainage, weathering will decrease. Surficial runoff is rather constant for the Southern pedon, while it decreases for the Northern pedon. In both cases, it is 3 to 6 times below the actual evapotranspiration, having a limited impact on the vertical drainage.

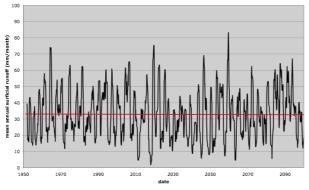


Fig 2: Mean annual surficial runoff (black line) and long term linear trend (red line), calculated for pedon South.

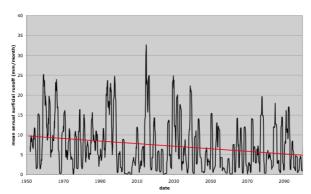


Fig. 3: Mean annual surficial runoff (black line) and long term linear trend (red line), calculated for pedon North.

The CO2 consumption for dolomite is reported for the south to be about 1 mol m-2 a-1. This is about six times the world average CO2-consumption rate. Are there regional studies to compare these results with? What is the regional CO2-consumption for the continent or carbonates in this area on average? It would be useful to compare the outcome with some reported values.

Since most of this CO₂ consumption is related to dolomite dissolution, these numbers must be compared to the CO₂ consumption by carbonate outcrops. Carbonate outcrops (including marls, dolomites, limestones...) cover about 13.4% of the continental surface ($20.1 \times 10^6 \text{ km}^2$) (Amiotte-Suchet et al., 2003). They consume 12.3×10^{12} mol of atmospheric CO₂/yr (Gaillardet et al., 1999). This gives a mean CO₂ consumption of 0.62 mol/m²/yr, close to our estimate.

Results can be compared with the modelled values (parametric laws calibrated on field data) from Moosdorf et al. (2011). For unconsolidated sediments of North America, including marginally loess, CO_2 consumption should be 0.2 mol $CO_2/m^2/yr$ for the Southern pedon, and 0.05 mol $CO_2/m^2/yr$ for the Northern pedon. For carbonate rocks of North America, these numbers rise to 0.8 and 0.25 mol $CO_2/m^2/yr$ respectively in Moosdorf et al. (2011). These numbers are of the same order of magnitude than our results (1 mol $CO_2/m^2/yr$ for the South and 0.3-0.7 mol $CO_2/m^2/yr$ for the North). Given the various type of sediments included in the unconsolidated sediment class of Moosdorf et al. (2011), it is not surprising that we got a better agreement with the carbonate class weathering. This discussion will be included in the revised version.

P10862, L 15: Can the finding, CO2-diffusion increases with temperature, be generalized to other soil-rock systems? I think this is an important point.

That is a good question, but it can only be solved through a careful modeling of other rock system. We feel that this is beyond the scope of the present study.

Conlusions: Considering the high complexity of the analysis approach, how close to reality is this model set up now? Should it be improved, and if yes how? Or in other words, how, if, can it be applied as a setup for global applications? This type of information would be useful for scientists following comparable or different approaches.

Required improvements are discussed in the limitation section. This concerns the hydrological behaviour of the weathering profile, and the modelling of the dissolution/precipitation of carbonate phases. Because dissolution is controlled by transport, vertical drainage must be carefully modelled. Carbonate precipitation modelling requires an accurate knowledge of the precipitation rate kinetics close to equilibrium. Given their high reactivity, we should worry about carbonate rocks in the context of global climate change. This was also the conclusion of Beaulieu et al. for another large North American watershed (Beaulieu et al., 2012). We will slightly expand the limitation section.

One very last point: if possible at this stage, we would like to add Marie Dury (University of Liège) to the co-authors, for her contribution to the CARAIB simulations.

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