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Interactive comment on "Silicate weathering and CO₂ consumption within agricultural landscapes, the Ohio-Tennessee River Basin, USA" *by* S. K. Fortner et al.

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We thank the two reviewers for their time and consideration. Anonymous Referee #1 we have considered your suggestions and will make several changes to our manuscript that will improve it.

Reviewer Comment: 'mostly US-American studies are referenced, and studies from Europe or Asia showing similar effects are less addressed/referenced'.

Our Response: Firstly, we have included references on agricultural landuse affects on weathering outside of the United States:

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Guo et al., 2010 (China) Perrin et al., 2008 (France, Global) Pierson-Wickman, 2009 a&b (France) Struyf et al., 2010 (Europe, multi-country) West et al., 2002 (Hi-malaya/Asia)

But we agree that including additional references from outside of the United States will strengthen this manuscript. In the revised manuscript we will include additional citations from the following regions:

Asia: Jenkins et al. 1995, Stream chemistry in the middle hills and high mountains of the Himalayas, Nepal, Journal of Hydrology- They observe potential increases in silicate weathering associated with terraced farming and mention physical controls that might enhance weathering (flooding, ploughing etc.). They also observe elevated nutrients and acid anions from fertilizers, but these are not connected with enhanced silicate weathering rates.

Collins & Jenkins, 1996, The impact of agricultural land use on stream chemistry in the Middle Hills of the Himalayas, Nepal, Journal of Hydrology- They observe an increase in base anions and nutrients associated with agriculture and say greater chemical weathering rates occur in association with tillage (physical explanation similar to Jenkins et al., 1995)

Europe: Roy et al., 1999, Geochemistry of dissolved and suspended loads of the Seine River, France: anthropogenic impact, carbonate and silicate weathering, Geochimica et Cosmochimica Acta They identify agricultural sources of ions, but do not explicitly attribute agricultural practices to increased silicate weathering, primary findings are related to lithological differences between compared catchments

Conley et al., 2000, The Transport and Retention of Dissolved Silicate by Rivers in Sweden and Finland, Limnology and Oceanography They say that changes in water storage, plant uptake could affect DSi concentrations from agricultural lands and that DSi may have a relation to the amount of arable land.

Clymans et al., 2011, Anthropogenic impact on amorphous silica pools in temperate soils. Biogeosciences. They quantify changes in soil ASi resulting from human disturbance including agricultural landuse.

North America (Canada and U.S.): Moosdorf et al., 2011a, Atmospheric CO2 consumption by chemical weathering in North America, Geochimica et Cosmochimica Acta They observe the influence of liming on agricultural watersheds

We will also include the following references to further improve our interpretation throughout the manuscript.

Moosdorf et al., 2011b, Changes in dissolved silica mobilization into river systems draining North America until the period 2081-2100 Examines present day global dissolved silica fluxes and modeled future global fluxes.

Riebe et al., 2004, Erosional and climatic effects on long-term chemical weathering rates in granitic landscapes spanning diverse climate regimes. & White and Brantley, 2003. The effect of time on the weathering of silicate minerals: why do weathering rates differ in the laboratory and field? Chemical Geology

The combined suggest that landscape age, rather than the age of the lithology alone, and the duration of time in the weathering zone is important to the amount of chemical weathering. This will clarify our interpretation.

Reviewer Comment: 'The manuscript could be strengthened by comparing their results with results from other sites in the US or globally, affected or not affected by the analyzed effect. This would allow the reader to better understand the relevance of identified CO2-consumption-suppression rates or the rates of CO2 consumption. For example is the identified CO2-consumption regionally/globally high or low, or are the identified nitrification rates high or low, etc.' Our Response: We agree with the reviewer that comparing with more datasets would be ideal. We have given examples from the literature (e.g. Barnes and Raymond, 2009; Perrin et al., 2008, Pierson-Wickmann et

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al., 2009a, 2009b) that fertilizer applications enhance chemical weathering. However, these studies have focused on fertilizer effects on carbonate weathering/CO2. Our paper is the first that we are aware of that explicitly examines fertilizer effects on silicate weathering. While there may be other datasets that we could examine that might show these effects, this would involve a more extensive search as we feel we have cited all of the relevant literature. We also feel that comparing small watersheds in Ohio to the larger Ohio River Basin was an important first step to understanding the possible implications of fertilizer applications to silicate weathering and aids in the scaling-up perspective from small watersheds to a continental scale. In the revised manuscript we will strengthen our argument by stressing that our findings have global importance given that nitrogenous fertilizer use has increased globally (Pongratz et al., 2008), a point that we made in the introduction. We will suggest that future global increases in N-loading associated with fertilizer will affect silicate weathering globally and that the greatest effects will be observed in younger, more readily weathered soils (as per our Ohio River comparison). In our revised manuscript we will also suggest that other watersheds in distinct lithologies (globally) could be examined to determine which rocktypes are most reactive to fertilizer applications.

Reviewer Comment: 'Na and K, important cations are not measured, either. Because of this it is difficult to assess the quality of the samples by standard techniques like ion balance. The usual application of geochemical evaluation techniques is with this data set not possible. The missing of the named parameters does at the first glance not allow for the evaluation of a bias due to the missing ions.'

Our Response: Firstly, the quality of presented samples should be related to the methodology section. We feel that the presented QA/QC data are appropriate (pg. 9439) and that we can have confidence in our reported geochemical results. Secondly, in the revised manuscript we will have made note of why we have not provided other cation and anion data. The fertilized watersheds have also received potash applications that alter the amount of K+, and Cl- on the landscape. This would affect any ion

balance calculations in determining ion mobilization from silicate weathering. We feel that our approach of measuring DSi directly was enough to identify potential CO2 consumption. This approach was first utilized in a series of papers by Huh and Edmond (e.g. Edmund and Huh, 1997) and more recently by Goldsmith et al, 2008. These references will be included in our discussion. We have also noted that some DSi could be retained (Triplett et al., 2008), or taken up by diatoms or within plants, especially in the larger Ohio River Basin. However, only the corn kernels are removed from the NAEW corn watersheds and they contain negligible silica (as per corn analyses in Lanning et al., 1980). Unknown is whether the returned stover (corn parts) is readily mobilized by wind or water.

Edmund and Hugh, 1997, Chemical weathering yields from basement and orogenic terrains in hot and cold climates, in Ruddiman, W.F., ed., Tectonic uplift and climate change: New York, Springer. Goldsmith et al., 2008, Extreme storm events, landscape denudation, and carbon sequestration: Typhoon Mindulle, Choshui River, Taiwan, Geology Lanning et al., 1980, Silica and ash content and depositional patterns in tissues of mature Zea mays L. plants*, Annals of Botany Triplett et al., 2008. Silica fluxes and trapping in two contrasting natural impoundments of the upper Mississippi River, Biogeochemistry

Reviewer Comment: In addition it was not sufficiently tested if trace carbonate or even abundant carbonate would influence the analysis.

Our Response: This is an excellent point. NAEW soils (upper 60 cm) and they have a maximum carbonate concentration of 4.5 mg/g. The mean concentration for all sites is much lower (<1.0 mg/g). In our revised manuscript will include this information and spend more time discussing the specific mineralogy of NAEW soils (Eckstein et al., 2007). Additionally we will provide more detail on the lithologies of the Ohio River Basin using Moosdorf et al., 2010. Trace carbonates may be responsible for some of the DSi released even over large areas (Jansen et al., 2010).

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Eckstein et al., 2007, Chemical evolution of acid precipitation in the unsaturated zone of the Pennsylvanian siltstones and shale of central Ohio, Hydrogeology Journal.

Jansen et al., 2010, Dissolved silica mobilization in the conterminuous USA, Chemical Geology

Moosdorf et al., 2010, Lithological composition of the North American continent and implications of lithological map resolution for dissolved silica flux modeling, Geochemistry, Geophysics, Geosystems.

Reviewer Comment: ... the potential influence of atmospheric deposition is not sufficiently discussed.

Our Response: We have subtracted atmospheric deposition of NAEW analyzed constituents. We will expand our explanation by including NADP numbers we used for subtraction in our revised manuscript.

Reviewer Comment: The raw data and the regression results should be provided in the appendix or supplemental information to allow in the future for comparing with the presented findings.

Our Response: We plan to include this within supplemental information and appreciate this suggestion.

Reviewer Comment: I would suggest using rather km2 than ha. This allows easier comparison with the literature addressing CO2 consumption, and is the more modern unit to be used.

Our Response: We justify the continued use of ha as ha is also a common unit, and often the preferred unit for agricultural plots. Many of the references, including high-impact journals we cite use ha.

For example: Barnes and Raymond, 2009; Conley et al., 2008; Pierson-Wickmann 2009a, Jordan et al., 1997; Scanlon et al., 2001; Struyf et al., 2010.

Reviewer Comment: The text could be rearranged for improving readability.

Our Response: Specific suggestions to improve readability were not given, but we will thoroughly examine the article after all revisions are made.

Reviewer Comment: Considering the readership and scope of Biogeosciences, I recommend to extend the discussion beyond the Ohio River basin and/or to explain more straight "What is new" and "Why is it important". Our Response: We have made an effort to clarify the new and exciting aspects of this paper in our conclusions. As previously mentioned, this will includes discussing future global increases in N-fertilizer application and what they might mean for silicate weathering rates.

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Interactive comment on Biogeosciences Discuss., 8, 9431, 2011.