

## GEOL 566: Geochemistry Seminar, Fall 2019 Syllabus (v. Aug 26<sup>th</sup>, 2019)

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(here much of the time)

### Class structure and background

This is a 2-unit class that will meet **T/Th 4:00-4:50pm** in ZHS B20 (time may change). Tuesdays will generally be lectures; Thursdays will be discussions of specific papers. Our first meeting will be on Tuesday, August 27<sup>th</sup>, at 4:00pm. If you cannot make that time and want to take the class, please contact Josh directly at the email address above.

Our main theme for this semester will be the application of geochemical tools to addressing major problems in catchment hydrology and hydrochemistry, with a particular emphasis on the use of solute chemistry to understand chemical weathering. We will also discuss implications for understanding the long-term carbon cycle. This class will suit graduate students with an interest in these topics and some coursework in geochemistry. GEOL 460 (Geochemistry) and GEOL 470 (Hydrogeology) are not prerequisites; some related knowledge will be helpful preparation, but will not be required as we will work through key concepts during the lectures and discussions. Students who have not taken GEOL 460 may wish do so simultaneously with this seminar, but that is not required.

*Your main responsibility for this class is to carefully read and critically assess the papers we have selected for each week (see below). You should come to each class having read the papers and having made written notes about them.* You should also have questions (usually at least 3 per paper) about what you don't understand as well as about where you see problems/gaps/etc. and potential for research opportunities. If you do not come prepared, you will not get the most out of the discussion. In addition to readings and discussion, you will be expected to complete a few specific assignments for this class, designed to get you engaging with the topic and developing and practicing core academic skills. These are detailed below.

### Learning objectives

The goals of this seminar include:

- Developing fundamental understanding of the major problems and open questions in catchment hydrology and hydrochemistry, and knowledge of the literature relevant to applying geochemical tools to these problems (including both recent literature and foundational papers).
- Gaining practice and developing skills in reading, critically evaluating, and discussing recent literature – including identifying gaps in current scientific understanding, and developing coherent ideas about how these could be addressed with future work.
- Building the capacity to identify gaps in our present understanding of major scientific problems, and developing coherent ideas about how these could be addressed with future work.

The learning objectives are woven into each of the class sessions and related homework.

## **Grading and specific assignments**

You will be expected to complete and turn in the following:

- 2 “manuscript reviews” (each worth 15% of your class grade), in which you write a critical review of one of the papers we have read in the class, as if you were asked to do so for a journal’s peer review process – the whole class will do this together, focusing on the same papers, and we will swap our reviews so we can all get a range of perspectives. The first of these will take place in Weeks 2-3. The second will be mid-way through the semester. We will provide detailed instructions at the time. The reviews are typically on the order of 1-3 pages long.
- 1 “mini-proposal” (worth 25% of your class grade), in which you present a 3-5 page written summary of a proposal for tackling one of the topics we cover – you can choose any topic during the semester, and this will be due on December 15<sup>th</sup> by midnight (i.e., during Week 16, which is finals week); this assignment represents the “final summative experience” for this class. You must submit a topic for this proposal (via email to both instructors) by Week 11 (November 6<sup>th</sup>); timely submission of a topic will count for 5% of your grade.

The assignments above reflect a total of 55% of your grade. Late work will be penalized 5% per week. In addition, you are all expected to lead one of our discussions, for 20% of your grade. The remaining 25% of your grade will be assigned on the basis of your active participation in the seminar discussions, evaluated on the basis of asking questions and actively commenting during each discussion. Please approach me (contact info above) with any questions or concerns. The sooner you contact me about any problems, the better. If you wait until the end of the semester, it may be too late to address them.

## **Statement for students with disabilities**

Any student requesting academic accommodations based on a disability is required to register with Disability Services and Programs (DSP) each semester. A letter of verification for approved accommodations can be obtained from DSP. Please be sure the letter is delivered to me as early in the semester as possible. DSP is located in STU 301 and is open 8:30 a.m.–5:00 p.m., Monday through Friday. Website for DSP and contact information: (213) 740-0776 (Phone), (213) 740-6948 (TDD only), (213) 740-8216 (FAX) [ability@usc.edu](mailto:ability@usc.edu).

## **Statement on academic integrity**

USC seeks to maintain an optimal learning environment. General principles of academic honesty include the concept of respect for the intellectual property of others, the expectation that individual work will be submitted unless otherwise allowed by an instructor, and the obligations both to protect one’s own academic work from misuse by others as well as to avoid using another’s work as one’s own. All students are expected to understand and abide by these principles. SCampus, the Student Guidebook, contains the University Student Conduct Code (see University Governance, Section 11.00), while the recommended sanctions are located in Appendix A.

## **Emergency preparedness/course continuity in a crisis**

In case of a declared emergency if travel to campus is not feasible, USC executive leadership will announce an electronic way for instructors to teach students in their residence halls or homes using a combination of Blackboard, teleconferencing, and other technologies. See the university’s website on Campus Safety and Emergency Preparedness.

## List of planned topics and schedule (subject to modification)

### Week 1 (Aug 27<sup>th</sup> and 29<sup>th</sup>)

Tuesday lecture (Hammond): Background on weathering & primer on hydrology – weathering & chemical cycles, Darcy's Law, reactive-transport, mass balance models, steady state vs. transient models, incongruent weathering,

Thursday Lecture/Discussion (West): Major open questions in catchment hydrology?

### Week 2 (Sept 3<sup>rd</sup> and 5<sup>th</sup>)

Tuesday lecture (West): Hydrograph separation, old vs. new water, multiple flow paths, concept of “water age”

Thursday discussion/readings (West): Water transit times and hydrograph separation with stable isotopes (H, O)

### Week 3 (Sept 10<sup>th</sup> and 12<sup>th</sup>)

Tuesday lecture (Hammond): Tracers, source functions, tracer losses, mixing and age dating with multiple flowpaths

Thursday discussion/readings (Hammond): Chemical tracers of groundwater transit times

### Week 4 (Sept 17<sup>th</sup> and 19<sup>th</sup>)

Tuesday lecture/discussion (West): Origins of solutes in natural waters

Thursday discussion/readings (**Abra Atwood**): Mass balance approaches to weathering budgets

### Week 5 (Sept 24<sup>th</sup> and 26<sup>th</sup> – JW away on Sept 26<sup>th</sup>)

Tuesday lecture (West/Hammond): Reactive-transport models

Thursday lecture (Hammond): Radioisotope balances, alpha-recoil, dissolution, weathering vs. particle transport

### Week 6 (Oct 1<sup>st</sup> and 3<sup>rd</sup> – JW away on Oct 1<sup>st</sup>)

Tuesday discussion/readings (**Le Li**): Applications of uranium series tracers to transit times

Thursday discussion/readings (West): Concentration-discharge relations and the problem of catchment chemostasis

Week 7 (Oct 8<sup>th</sup> and 10<sup>th</sup>)

Tuesday discussion/readings (West): Models for temporal dynamics

Computer Exercise (West): An introduction to using PHREEQC to model solute chemistry

Week 8 (Oct 15<sup>th</sup> and 17<sup>th</sup>)

Tuesday discussion/readings (**Emily Burt**): Large rivers, global fluxes, and inversion of solute sources

No discussion section (Fall Recess)

Week 9 (Oct 22<sup>nd</sup> and 24<sup>th</sup>)

Lecture (West): Primer on kinetics and thermodynamics; kinetic controls on mineral dissolution and incongruent weathering

Topic for discussion/readings (**Katie Denniston**): The trace calcite story, and how carbonate comes to dominate weathering fluxes

Week 10 (Oct 29<sup>th</sup> and 31<sup>st</sup> – JW may be away, TBC)

Tuesday lecture (Hammond): Adsorption/desorption

Topic for discussion/readings (**Jaclyn Pittman**): Ion exchange: An under-appreciated control on solute dynamics

Week 11 (Nov 5<sup>th</sup> and 7<sup>th</sup>)

Lecture (West): Reaction fronts

Topic for discussion/readings (West): Weathering fronts and solute acquisition

Week 12 (Nov 12<sup>th</sup> and 14<sup>th</sup> – JW not available on 14<sup>th</sup> so discussion of these papers will be on Tuesday Nov 19<sup>th</sup>)

Tuesday lecture (West): Weathering, global geochemical cycles, and climate feedbacks

No class on Thursday November 14<sup>th</sup>

Week 13 (Nov 19<sup>th</sup> and 21<sup>st</sup>)

Tuesday discussion/readings (**Abby Lunstrum**): Climate-dependence of weathering and global implications

Thursday lecture (West): The redox chemistry of hydrological systems and weathering interactions

Week 14 (Nov 26<sup>th</sup> and 28<sup>th</sup>)

Tuesday discussion/readings (**Xiaopeng Bian**): Sulfide oxidation and its carbon cycle implications

Thursday no class (Thanksgiving)

Week 15 (Dec 3<sup>rd</sup> and 5<sup>th</sup>)

Lecture (West): Seawater isotope records of weathering

Topic for discussion/readings (**Emily Tibbett**): Interpreting past records of weathering

**Readings for seminar discussions (subject to modification; we will keep you informed as the semester progresses!)**

**Required:** please make sure to read these *critically* as we will focus our discussion on them; **Background:** general papers, e.g., reviews, that may prove useful as general resources on the topic; **Optional/Recommended:** other specialized studies that may be of interest if you want to dig a bit deeper

Week 1 (for Tuesday)

**Required:** Frings and Buss. 2019. The central role of weathering in the geosciences. *Elements* v. 15, pp. 229-234.

**Background:** Other papers in the Elements issue available here:

<https://pubs.geoscienceworld.org/elements/issue/15/4>

Week 1 (for Thursday)

**Required:** Pfister and Kirchner. 2017. Debates – Hypothesis testing in hydrology: Theory and practice. *Water Resources Research* v. 53, pp. 1792-1798.

**Background:** Church, M.R. 1997. Hydrochemistry of forested catchments. *Annu. Rev. Earth Planet. Sci.* v. 25, pp. 23-59.

**Background:** Jenkins, Peters, and Rodhe. 1994. Hydrology. In *Biogeochemistry of Small Catchments*. Edited by B. Moldan and J Cerny. Scope 51. John Wiley and Sons. pp: 31-54.

Week 2 (for Thursday)

**Required:** Pearce, Stewart, and Sklash. 1986. Storm runoff generation in humid headwater catchments 1: Where does the water come from? *Water Resources Research* v. 22, pp. 1263-1272.

**Required:** Jasechko, Kirchner, Welker, and McDonnell. 2016. Substantial proportion of global streamflow less than three months old. *Nature Geoscience* v. 9, pp. 126–129. **[you should plan your first manuscript review assignment on the basis of this paper; that assignment will be due the Thursday of week 3, i.e., one week following this group discussion]**

**Background:** McGuire and McDonnell. 2006. A review and evaluation of catchment transit time modeling. *Journal of Hydrology* v. 330, pp. 543-563.

**Background:** McDonnell et al. 2010. How old is streamwater? Open questions in catchment transit time conceptualization, modelling and analysis. *Hydrological Processes* v. 24, pp. 1745-1754.

**Optional/Recommended:** Benettin, Rinaldo, and Botter. 2015. Tracking residence times in hydrological systems: forward and backward formulations. *Hydrological Processes* v. 29, pp. 5203-5213.

**Optional/Recommended:** Kirchner. 2016. Aggregation in environmental systems—Part 1: Seasonal tracer cycles quantify young water fractions, but not mean transit times, in spatially heterogeneous catchments. *Hydrol. Earth Syst. Sci.* v. 20, pp. 279–297.

#### Week 3 (for Thursday)

**Required:** Manning, Clark, Diaz, Rademacher, Earman, and Plummer. 2012. Evolution of groundwater age in a mountain watershed over a period of thirteen years. *Journal of Hydrology* v. 460-461, pp. 13-28.

**Required:** Jasechko. 2016. Partitioning young and old groundwater with geochemical tracers. *Chemical Geology* v. 427, pp. 35-42

**Background:** Phillips and Castro. 2003. Groundwater dating and residence time measurements. *Treatise of Geochemistry* v. 5, pp. 451-497.

**Optional/Recommended:** Budenburg and Plummer. 2000. Dating young groundwater with sulfur hexafluoride: Natural and anthropogenic sources of sulfur hexafluoride. *Water Resources Research* v. 36, pp. 3011-3030.

**Optional/Recommended:** Kagabu, Matsunaga, Ide, Momoshima, Shimada. 2017. Groundwater age determination using <sup>85</sup>Kr and multiple age tracers (SF<sub>6</sub>, CFCs, and <sup>3</sup>H) to elucidate regional groundwater flow systems. *Journal of Hydrology* v. 12, pp. 165-180.

#### Week 4 (for Tuesday)

**Required:** Garrels and Mackenzie. 1967. Origin of the Chemical Compositions of Some Springs and Lakes. In *Equilibrium Concepts in Natural Water Systems*, Ch. 10, pp 222-242.

#### Week 4 (for Thursday)

**Required:** Finley and Drever. 1997. Chemical mass balance and rates of mineral weathering in a high-elevation catchment, West Glacier Lake, Wyoming. *Hydrological Processes* v. 11, pp. 745-764.

**Required:** Bricker, Jones, and Bowser. 2003. Mass-balance Approach to Interpreting Weathering Reactions in Watershed Systems. *Treatise of Geochemistry* v. 5, pp 119-132.

#### Week 5 (for Tuesday)

**Required:** Maher and Mayer. 2019. Tracking Diverse Minerals, Hungry Organisms, and Dangerous Contaminants Using Reactive Transport Models. *Elements* v. 15, pp. 81-86.

#### Week 5 (for Thursday)

**Required:** Krishnaswami et al. 1982. Radium, Thorium and Radioactive Lead Isotopes in Groundwaters. *Water Resources Research* v. 18, pp. 1633-1675.

#### Week 6 (for Tuesday)

**Required:** Solomon, Hunt, and Poreda. 1996. Source of radiogenic helium 4 in shallow aquifers: Implications for dating young groundwaters. *Water Resources Research* v. 32, pp. 1805-1813.

**Required:** Vinson et al. 2018. Radium isotope response to aquifer storage and recovery in a sandstone aquifer. *Applied Geochemistry* v. 91, pp. 54-63.

**Background:** Porcelli and Swarzenski. 2003. The Behavior of U- and Th-series Nuclides in Groundwater. *Reviews in Mineralogy and Geochemistry* v. 52, pp. 317-361.

**(Very Optional) Background:** Osmond and Cowart. 2000. U-series nuclides as tracers in groundwater hydrology. In *Environmental Tracers in Subsurface Hydrology*. Cook and Herczeg, eds. Pp. 145-173.

#### Week 6 (for Thursday)

**Required:** Godsey, Kirchner, and Clow. 2009. Concentration–discharge relationships reflect chemostatic characteristics of US catchments *Hydrological Processes* v. 23, pp 1844-1864.

**Required:** Godsey, Hartmann, and Kircher. 2019. Catchment chemostasis revisited: water quality responds differently to variations in weather and climate. *Hydrological Processes* in press.

**Required:** Chorover, Derry, and McDowell. 2017. Concentration-Discharge Relations in the Critical Zone: Implications for Resolving Critical Zone Structure, Function, and Evolution. *Water Resources Research* v. 53, pp. 8654-8659 [this is a brief introduction to a special issue]

**Optional/Recommended:** Moatar, Abbott, Minaudo, Curie, and Pinay. 2017. Elemental properties, hydrology, and biology interact to shape concentration-discharge curves for carbon, nutrients, sediment, and major ions. *Water Resources Research* v. 53, pp. 1270-1287.

**Optional/Recommended:** Kim, Dietrich, Thurnhoffer, Bishop, and Fung. 2017. Controls on solute concentration-discharge relationships revealed by simultaneous hydrochemistry observations of hillslope runoff and stream flow: The importance of critical zone structure. *Water Resources Research* v. 53, pp. 1424-1443.

#### Week 7 (for Tuesday)

**Required:** Johnson, Likens, Bormann, Fisher, and Pierce. 1969. A Working Model the Variation in Stream Water Chemistry at the Hubbard Brook Experimental Forest, New Hampshire. *Water Resources Research*, v. 5, pp. 1353-1363.

**Required:** Maher. 2011. The role of fluid residence time and topographic scales in determining chemical fluxes from landscapes. *Earth and Planetary Science Letters* v. 312, pp. 48-58

**Required:** Ameli, Beven, Erlandsson, Creed, McDonnell, and Bishop. 2016. Primary weathering rates, water transit times, and concentration-discharge relations: A theoretical analysis for the critical zone *Water Resources Research* v. 53, pp. 942-960.

#### Week 8 (for Tuesday)

**Required:** Meybeck, M. 1987. Global chemical weathering of surficial rocks estimated from river dissolved loads. *American Journal of Science* v. 287, pp. 401-428.

**Required:** Gaillardet, Dupre, Louvat, and Lewin. 1999. Global silicate weathering and CO<sub>2</sub> consumption rates deduced from the chemistry of large rivers. *Chemical Geology* v. 159, pp. 3-30.

**Required:** Moon, Chamberlin, and Hilley. 2014. New estimates of silicate weathering rates and their uncertainties in global rivers. *Geochimica et Cosmochimica Acta* v. 134, pp. 257-274.

#### Week 9 (for Thursday)

**Required:** Blum, Gazis, Jacobson, and Chamberlain. 1998. Carbonate versus silicate weathering in the Raikhot watershed within the high Himalayan crystalline series. *Geology* v. 26, pp. 411-414.

**Required:** White, Bullen, Vivit, Schulz, and Clow. 1999. The role of disseminated calcite in the chemical weathering of granitoid rocks. *Geochimica et Cosmochimica Acta* v. 63, pp. 1939-1953 **[you should plan your second manuscript review assignment on the basis of this paper; that assignment will be due on Thursday before the discussion section]**

**Required:** Jacobson, Blum, Chamberlain, Craw, and Koons. 2003. Climatic and tectonic controls on chemical weathering in the New Zealand Southern Alps. *Geochimica et Cosmochimica Acta* v. 67, pp. 29-46.

#### Week 10 (for Thursday)

**Required:** Cerling, Pederson, and Von Damm. 1989. Sodium-calcium ion exchange in the weathering of shales: Implications for global weathering budgets. *Geology* v. 17, pp. 552-554.

**Required:** Neal et al. 1990. Limitations to the understanding of ion-exchange and solubility controls for acidic Welsh, Scottish and Norwegian sites. *Journal of Hydrology* v. 116, p. 11-23.

**Required:** Bluth and Kump. 1994. Lithologic and climatologic controls of river chemistry. *Geochimica et Cosmochimica Acta* v. 58, pp. 2341-2359.

**Optional/Recommended:** Herndon, Dere, Sullivan, Norris, Reynolds, and Brantley. 2015. Biotic controls on solute distribution and transport in headwater catchments, *Hydrol. Earth Syst. Sci.* v. 19, pp. 3333-3347.

#### Week 11 (for Thursday)

**Required:** Brantley et al. 2017. Toward a conceptual model relating chemical reaction fronts to water flow paths in hills. *Geomorphology* v. 277, pp. 100-117.



**Required:** Harman and Cosans. 2019. A low-dimensional model of bedrock weathering and lateral flow coevolution in hillslopes: 2. Controls on weathering and permeability profiles, drainage hydraulics, and solute export pathways. *Hydrological Processes* v. 33, pp. 1168-1190.

#### Week 13 (for Tuesday)

**Required:** White and Blum. 1995. Effects of climate on chemical weathering rates in watersheds. *Geochimica et Cosmochimica Acta* v. 59, pp. 1729-1747.

**Required:** West, Bickle, and Galy. 2005. Tectonic and climatic controls on silicate weathering. *Earth and Planetary Science Letters* v. 235, pp. 211-228.

**Required:** Winnick and Maher. 2018. Relationships between CO<sub>2</sub>, thermodynamic limits on silicate weathering, and the strength of the silicate weathering feedback. *Earth and Planetary Science Letters* v. 485, pp. 111-120

#### Week 14 (for Tuesday)

**Required:** Spence and Telmer. 2005. The role of sulfur in chemical weathering and atmospheric CO<sub>2</sub> fluxes: Evidence from major ions, <sup>13</sup>CDIC, and <sup>34</sup>SSO<sub>4</sub> in rivers of the Canadian Cordillera. *Geochimica et Cosmochimica Acta* v. 69, pp. 5441-5458.

**Required:** Calmels et al. 2007. Sustained sulfide oxidation by physical erosion processes in the Mackenzie River basin. *Geology* v. 35, pp. 1003-1006.

**Required:** Torres, West, Clark, Paris, Bouchez, Ponton, Feakins, Galy, and Atkins. 2016. The acid and alkalinity budgets of weathering in the Andes–Amazon system: insights into the erosional control of global biogeochemical cycles. *Earth Planet. Sci. Lett.* v. 450, pp. 381-391.

**Optional/Recommended:** Mayer, Shanley, Bailey, and Mitchell. 2010. Identifying sources of stream water sulfate after a summer drought in the Sleepers River watershed (Vermont, USA) using hydrological, chemical, and isotopic techniques. *Applied Geochemistry* v. 25, pp. 747-754.

**Optional/Recommended:** Turchyn et al. 2013. Isotope evidence for secondary sulfide precipitation along the Marsyandi River, Nepal, Himalayas. *Earth and Planetary Science Letters* v. 374, pp. 36-46.

**Optional/Recommended:** Killingsworth, Bao, and Kohl. 2018. Assessing Pyrite-Derived Sulfate in the Mississippi River with Four Years of Sulfur and Triple-Oxygen Isotope Data. *Environmental Science and Technology* v. 52, pp. 6126-6136.

**Optional/Recommended:** Sullivan et al. 2016. Oxidative dissolution under the channel leads geomorphological evolution at the Shale Hills catchment. *American Journal of Science* v. 316, pp. 981-1026

**Optional/Recommended:** Zoikos, Tank, and Kokeji. 2018. Mineral weathering and the permafrost carbon-climate feedback. *Geophysical Research Letters* v. 45, pp. 9623-9632.

#### Week 15

**Required:** Torres et al. 2014. Sulphide oxidation and carbonate dissolution as a source of CO<sub>2</sub> over geological timescales. *Nature* v. 507, pp. 346-349.

1-2 more papers to be added here

## **Additional Readings** (we may shift topics and address one of these, TBD)

### Ecohydrology

Berry et al. 2017. The two water worlds hypothesis: Addressing multiple working hypotheses and proposing a way forward. *Ecohydrology* v. 11, e1843.

Penna et al. 2018. Ideas and perspectives: Tracing terrestrial ecosystem water fluxes using hydrogen and oxygen stable isotopes – challenges and opportunities from an interdisciplinary perspective. *Biogeosciences* v. 15, pp. 6399-6415.

Allen, Kirchner, Braun, Siegwolf, and Goldsmith. 2019. Seasonal origins of soil water used by trees. *Hydrol. Earth Syst. Sci.* v. 23, pp. 1199-1210.

Evaristo et al. 2019. Characterizing the fluxes and age distribution of soil water, plant water and deep percolation in a model tropical ecosystem. *Water Resources Research* v. 55, pp. 3307-3327.

### Groundwater Springs

Rademacher, Clark, Hudson, Erman, and Erman. 2001. Chemical evolution of shallow groundwater as recorded by springs, Sagehen basin, Nevada County, California. *Chemical Geology* v. 179, pp. 37-51.

Manga. 2001. Using springs to study groundwater flow and active geologic processes. *Annual Reviews in Earth and Planetary Sciences* v. 29, pp. 201-208.

Ide, Hosono, Hossain, and Shimada. 2018. Estimating silicate weathering timescales from geochemical modeling and spring water residence time in the Kirishima volcanic area, southern Japan. *Chemical Geology* v. 488, pp. 44-55.

Luo and Jiao. 2019. Unraveling controlling factors of concentration discharge relationships in a T fractured aquifer dominant spring-shed: Evidence from mean transit time and radium reactive transport model. *Journal of Hydrology* v. 571, pp 528-544.

### U-series measurement of weathering timescales

**Background:** DePaolo, Lee, Christensen, and Maher. 2012. Uranium comminution ages: Sediment transport and deposition time scales. *Comptes Rendus Geoscience* v. 344, pp. 678-687.

**Background:** Dosseto. 2014. Chemical Weathering (U-Series). In *Encyclopedia of Scientific Dating Methods*.

DePaolo, Maher, Christensen, and McManus. 2006. Sediment transport time measured with U-series isotopes: results from ODP North Atlantic drift site 984. *Earth and Planetary Science Letters* v. 248, pp. 394-410.

Li et al. 2018. Weathering dynamics reflected by the response of riverine uranium isotope disequilibrium to changes in denudation rate. *Earth and Planetary Science Letters* v. 500, pp. 136-144.

Handley et al. 2013. Sediment residence times constrained by uranium-series isotopes: A critical appraisal of the comminution approach. *Geochimica et Cosmochimica Acta*, v. 103, pp. 245-262.