2020

Using X-ray diffraction of stream sediment to better understand the geology and weathering environment of central Cuba

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Senior Honors Thesis

Defended April 29, 2020

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Acknowledgements

Thank you to everyone who worked on this project and who graciously provided me with advice and support. This research would not have been possible without the interdisciplinary, collaborative efforts of many talented individuals. To our collaborators in Cuba, thank you for making this project possible. John Hughes and Nico Perdrial: your enthusiasm and commitment in helping me learn how to perform quantitative X-ray Diffraction is greatly appreciated. I want to thank my advisor, Paul Bierman, for getting me involved in this project and patiently providing me with thoughtful guidance. Thank you to Gillian Galford for graciously stepping up as committee chair. I'd also like to thank Mae Kate Campbell and Amanda Schmidt for their help in providing additional data concerning the watersheds studied here such as mapped catchment areas and erosion rates.

Sources of funding that made this research possible: UVM FOUR Summer Undergraduate Research Fellowship (SURF); Hawley Undergraduate Research Award; APLE Summer Stipend Award (declined for SURF)

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I. Introduction

Largely due to political reasons, there is not much data available to U.S. scientists on the geology of Cuba because of restricted access to the country, and thus our understanding of the local stream geochemistry, water quality, mineralogy, and complex rock formations is limited (Iturralde-Vinent et al. 2016; Schmidt et al. 2016). However, considering the improving political situation, the US Department of Agriculture and the Cuban Ministry of Agriculture signed a Memorandum of understanding on March 21, 2016 which contains eight areas of cooperation, including "agricultural research and management techniques for soil and forest conservation." (USDA, 2016). The research project I am part of (funded by the US National Science Foundation to UVM and Oberlin) in collaboration with other American universities and Centro de Estudios Avanzados de Cuba (CEAC) is focused on quantifying the effects on the landscape of industrialized agriculture followed by country-wide soil conservation efforts. The aim of my specific project is to better contextualize the work that the 2016 memorandum makes possible by providing a better understanding of the stream sediment of Cuba. Our study is the first of its kind in Cuba after nearly 60 years of isolation from U.S. scientists (Schmidt et al. 2016).

In a globalized world, where environmentally-impactful industrial agriculture is common, understanding the mechanisms and benefits of transitioning towards organic, conservation agricultural practices is vital to sustaining soil health, water quality, wildlife, and Earth's rapidly growing human population (Schmidt et al. 2016). Cuba is a uniquely useful place to study the effects of conservation agriculture practices on erosion due to its history of industrial sugarcane agriculture (1959-1991) followed by 25 years of small scale, organic conservation agriculture during and after the special period (1991-present); thus Cuba provides a model to test for the landscape scale results of implementing conservation agriculture practices (Schmidt et al. 2016). Such research is time-sensitive as agricultural imports and exports in Cuba have increased in the last decade and demand for local, sustainable agriculture has decreased (Zahniser et al. 2015). Because of this, data on the effects of conservation agriculture must be collected soon before industrial agriculture dominates Cuba again.

As agricultural impacts on the landscape are quantified, contextualizing these impacts with data concerning the nature of actively weathering detritus is crucial to a comprehensive and mechanistic understanding of how human land use influences water quality, erosion rates, and overall landscape change. This project is the first detailed study of central Cuban stream sediment mineralogy and rock weathering processes, providing critical insight into how Cuban rocks weather into soil, thereby influencing water chemistry as well as soil and water quality in central Cuba.

By determining the composition of river sediment samples, I provide a comprehensive understanding of the actively weathering detritus for all the basins we are studying in Cuba. Using this understanding, I aim to put into context the erosion rates, water chemistry, and water quality data from each basin, allowing for a more mechanistic understanding of these data as well as both human impacts and natural rock weathering processes. More specifically, my results of local sediment composition improve our understanding of the impacts of conservation agriculture on sediment and soil composition, erosion, and water chemistry in Cuba through its control on the nature and rate of rock weathering and soil formation. Implications include what rock and mineral types are being weathered, how resistant these phases are to erosion, and how the weathering of these phases influences water quality, soil health, and wildlife, therefore providing crucial insight into human impacts on Cuba's dynamic landscape. As worldwide erosion rates increase with the use of industrial agriculture, providing this mechanistic understanding of rock-weathering in Cuba is crucial as it could lead to more effective solutions mitigating the environmental impacts of agriculture on landscapes both locally and on a global scale.

Chapter II is a paper drafted with the intention of being submitted to Catena for review. This paper focuses less on the impacts of land-use changes and more on the specific implications derived from mineralogic data determined using X-ray diffractometry. Raw X-ray diffraction data and quantification results are currently attached to this thesis as an appendix, and will eventually be available in an online EarthChem data repository.

II. Using X-ray diffraction of stream sediment to better understand the geology and weathering environment of central Cuba

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Abstract

In humid, tropical regions, where chemical weathering rates are high, the relationship between bedrock lithology, stream sediment mineralogy, and stream water chemistry is neither straightforward nor well known. For a country such as Cuba, which has been isolated from much of the world's geologic community for decades, data are sparse; yet, such data are important for understanding landscape change over time, including deciphering trends in stream water geochemistry and rates of erosion, both natural and human-caused. Here, as part of a collaborative Cuban-American project focused on understanding mass transfer from the island to the ocean, we used quantitative X-ray diffraction to determine the bulk mineral composition of river sediment in two grain sizes. We used this data to determine the relationship between sediment mineralogy and water chemistry, weathering rates, sediment elemental composition, and mapped bedrock geology in 26 central Cuban drainage basins.

Diffraction data show that quartz, feldspar, and calcite are present in most stream sediment samples reflecting the relative abundance of igneous, metamorphic, and carbonate lithologies in central Cuba. Although quartz dominated stream sediment composition across all sampled watersheds (N=26), significant amounts of calcite, swelling clays, feldspars, and amphiboles suggest that the mineralogy of the stream sediments is useful both as a marker for watershed geologic characteristics and as a proxy

for both physical and chemical weathering intensity. We find that water quality indicators such as Ca, Mg, DOC, K, and Cl can be deciphered through quantification of weathering-prone minerals such as calcite and weathering products such as swelling clays present in transport-limited streams. Additionally, we suggest that sediment mineralogy may provide insight into water quality parameters over longer time periods than traditional water samples. In combination with other data, mineralogical analysis of stream sediments can be used to determine the lithologic influence on local sediment and water composition in individual watersheds.

Introduction

Accurate description of the composition and distribution of bedrock underlying watersheds is crucial to understanding weathering processes, erosion, nutrient inputs, and geologic histories both regionally and globally (Johnsson, 1993; Mage and Porder, 2013; Keen-Zebert et al., 2017). In heavily vegetated regions such as Cuba, unweathered outcrops are rare and field access is limited, making traditional bedrock mapping difficult. In areas like this, quantifying the composition of stream sediments allows for the indirect sampling of an entire drainage basin from a single sample while simultaneously providing insight into the weathering processes affecting minerals as they travel from their bedrock source to, and down, stream networks (Johnsson, 1993). However, quantifying source rock composition using stream sediment mineralogy is difficult and uncertain because the composition and grain size distribution of stream sediment as it moves downstream is influenced by authigenesis, chemical and physical weathering, hydrodynamic sorting, and diagenesis, each of which are intimately interrelated and controlled by many interacting drivers (Schock and Steidtmann, 1976; Johnsson, 1993; Clark et al., 2017).

Here, we use quantitative powder x-ray diffraction to determine the mineralogical composition of 26 stream sediment samples from Central Cuba. We use these data to infer the mineral composition of rock formations underlying drainage basins upstream. We compare XRD-based mineralogic data to stream water chemistry, sediment elemental composition, and ^{10}Be-derived erosion rate data to infer the type and rate of regional weathering processes.

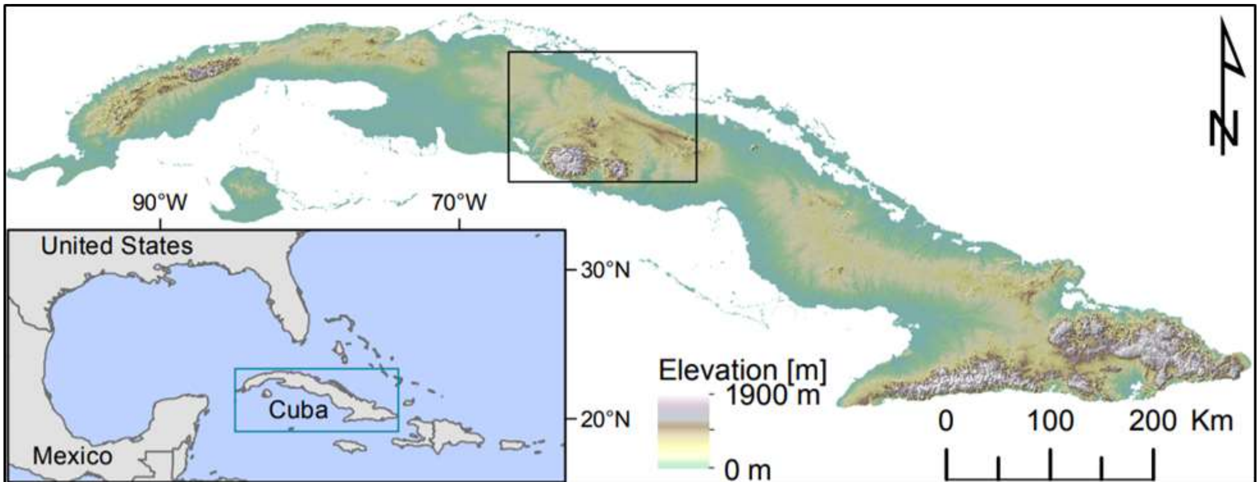


Fig. 1, Elevation map of Cuba. Inset shows location of Cuba relative to surrounding countries, black outline shows location of Fig. 3. Map from Bierman et al., (2020).

Background

1. Geology –

Cuba's bedrock geology is a complex mixture of young volcanic rock, older marine sedimentary rock, younger sedimentary rock from the weathering of basement rocks, and metamorphic forms of most of these rock types resulting from plate collision (Case and Holcombe, 1980; Schmidt et al., 2016). Cuba emerged after the breakup of Pangea ~135 ma as the North American Plate and the Caribbean plate converged, initiating subduction of the North American Plate. Over millions of years, this process produced magma that rose to the surface, erupted, and solidified, forming the backbone of Cuba as we know it today (Iturralde-Vinent et al., 2016). Due to its volcanic history, Cuba is largely underlain by igneous rock with high silica content, but also contains localized igneous rock with high Mg and Fe content (Pardo, 1975; French and Schenk, 2004; Iturralde-Vinent et al., 2016). Additionally, Cuba's position in an ocean basin resulted in the deposition of marine sedimentary rock with high calcite content (Iturralde-Vinent et al., 2016; French and Schenk, 2004). Most of these rock types also exist in forms that have been altered by the heat and pressure caused by past volcanic and metamorphic processes and the faulting of Cuba's active plate boundary (Case and Holcombe, 1980). Though the general geologic history of Cuba is established, analysis of local rock type and mineralogy will help properly contextualize landscape analyses from a geochemical perspective.

2. Physical geography -

Cuba is the largest Caribbean island (105,000 km^2). The terrain is primarily low-lying with uplands where streams head before flowing across plains towards the coasts. Pico Turquino (1,999 m), the highest peak, is located in the Sierra Maestra mountains on the southeast coast. Central Cuba, the focus of this study, contains the Escambray mountains to the south with abundant central/northern plains and hills throughout.

3. Weather, climate, and vegetation -

Cuba's climate is warm and tropical with dry winter months and wet summer and fall months. Villa Clara, central Cuba, has an average temperature of 24.8°C and experiences an annual rainfall of 1570 mm (Climate-data.org). Under the Koppen-Geiger classification, central Cuba is classified as almost entirely tropical savanna with tropical monsoon in the mountainous areas and small temperate regions at the highest peaks (Beck et al., 2018). Farmed plains, diverse three-canopy layer rainforests, two-canopy seasonal evergreen forests, one- to two-canopy semi-deciduous forests, single-canopy tropical karstic forests, one- to two-canopy dry forests, freshwater swamps, savannas, and grasslands are all present in central Cuba with mangroves and scrublands occupying the coasts (Prance and Borhidi, 1993).

4. Landscape history (humans) -

Before Spanish occupation began in 1492, Cuba was 92-95% covered by forests and woodlands and indigenous peoples grew cassava, yucca, and maize (Borhidi and Muñiz, 1980; Cosculluela, 1946). Woodland and forest cover plummeted to 8-13% from the start of the 19^{th} century through the 20^{th} century as woodland areas were cleared for agriculture including sugar-cane production, cattle breeding, and tobacco production (Borhidi and Muñiz, 1980; Zepeda, 2003). Cuba's history of large-scale, industrial sugarcane agriculture during a period of heavy dependence on industrial chemicals and petroleum (1959-1991) was followed by 25 years of small scale, organic conservation agriculture during and after the special period (1991-present). Since the beginning of the special period, forest cover has recovered to 36% (Galford et al., 2018).

Methods

Sediment samples were collected from central Cuban streams in summer 2018 (Bierman et al., 2020). We used X-ray powder diffraction for mineralogic analysis and scanned 26 stream sediment samples, each from a different watershed, in two grain sizes (fine: <63 µm, and coarse: 250-850 µm). Samples were 1) wet sieved to isolate two grain sizes and thus normalize for hydrodynamic sorting, which can obscure comparisons between unconsolidated samples (Johnsson, 1993), 2) oven dried at 65˚ C, 3) ground to a powder with an agate mortar and pestle, 4) scanned on a standard glass slide, 5) rinsed with tap water in a ceramic dish to isolate heavy minerals, 6) air dried, 7) re-ground, and 8) scanned on a zero-background plate (washed scan). Scans were performed from 5-55˚ (2Θ) at 2˚/minute using a Rigaku MiniFlex II diffractometer equipped with a Cu Kα X-ray tube. Diffractograms were analyzed and quantified for mineral abundances using the Rietveld module in Rigaku PDXL (v. 1.6.0.0). All Rietveld simulations were performed using a consistent set of reference minerals included in the PDF-2 database.

To analyze the clay fraction and identify swelling clays as well as specific clay species, 12 samples were placed in ethanol, shaken vigorously, and then sonicated for 1 minute before the suspended material was pipetted off to isolate the <50 µm fraction. These extracted fractions were pipetted onto a glass side (in preferred orientation), air dried, scanned from 3-25° (2Θ) at 1°/minute, heated to 400° C, scanned again, then heated to 550° C before being scanned a final time. For 3 of 12 samples, the final scan was not possible since the slide was damaged during heating and there was no extra sample material.

Mineral abundances calculated from XRD were compared to percent area in the drainage basin of each underlying lithology based on Case and Holcombe, 1980), water chemistry data (Bierman et al., 2020), XRF data for sediment elemental composition (See data repository for XRF table, personal communication, Yoelvis Bolanos), and in situ ^{10}Be data (Campbell et al., 2019) using scatter plots and ternary plots to test correlations between mineral abundance and other metrics.

Results

Most samples are dominated by quartz (coarse: 1.4 to 69.7%; mean = 33.2%, fine: 3.1 to 68%; mean = 18.6%) and feldspar (coarse: 0 to 74%; mean = 30.8%, fine: 0 to 89.0%; mean = 29.6%) with lesser amounts of calcite (coarse: 0 to 34.9%; mean = 10.6%, fine: 0 to 52.2%; mean = 12.5%), mica (coarse: 0 to 80.0%; mean = 3.8%, fine: 0 to 92%; mean = 13.8%), amphibole (coarse: 0 to 35.1%; mean = 5.9%, fine: 0 to 31.9%, mean = 5.9%), pyroxene (coarse: 0 to 59%; mean = 5.5%, fine: 0 to 34%; mean = 4.7%), oxide minerals (coarse: 0 to 15.7%; mean = 2.1%, fine: 0 to 33.3%, mean = 4.5%), and chlorite minerals (coarse: 0 to 6%, mean = 0.4%, fine: 0 to 24%; mean = 2.8%). Serpentine and zeolite minerals

Table 1: Average mineralogic composition of samples across watersheds (wt. %, N=26)*

	feldspar	quartz	calcite	10Å clay	mica	amphibole	pyroxene	oxide	chlorite	zeolite	serpentine
250-850 µm	30.8	33.2	10.6	7.0	3.8	5.9	5.5	2.1	0.4	0.1	0.8
<63 µm	29.6	18.6	12.5	9.0	13.8	5.4	4.7	4.5	2.8	0.2	0.0

* quantified based on XRD data using the Rietveld module in Rigaku PDXL software (v. 1.6.0.0)

are detected but scarce, and serpentine minerals are only detected in the coarse grain size (Table 1). Feldspars, amphiboles, pyroxenes, and zeolites show little to no content variation between grain sizes. Quartz is almost twice as abundant in the coarse fraction and micas are more than four times as abundant in the fine fraction than in the coarse fraction. Calcite, clay, chlorite, and oxide are all slightly more abundant in the fine fraction than in the coarse fraction. Heat treatment tests show 10Å clay peaks collapsed after heating to 400° C and no change after subsequently heating to 550° C in every tested sample with usable data (n=9), indicating the presence of swelling clays (Fig. 2).

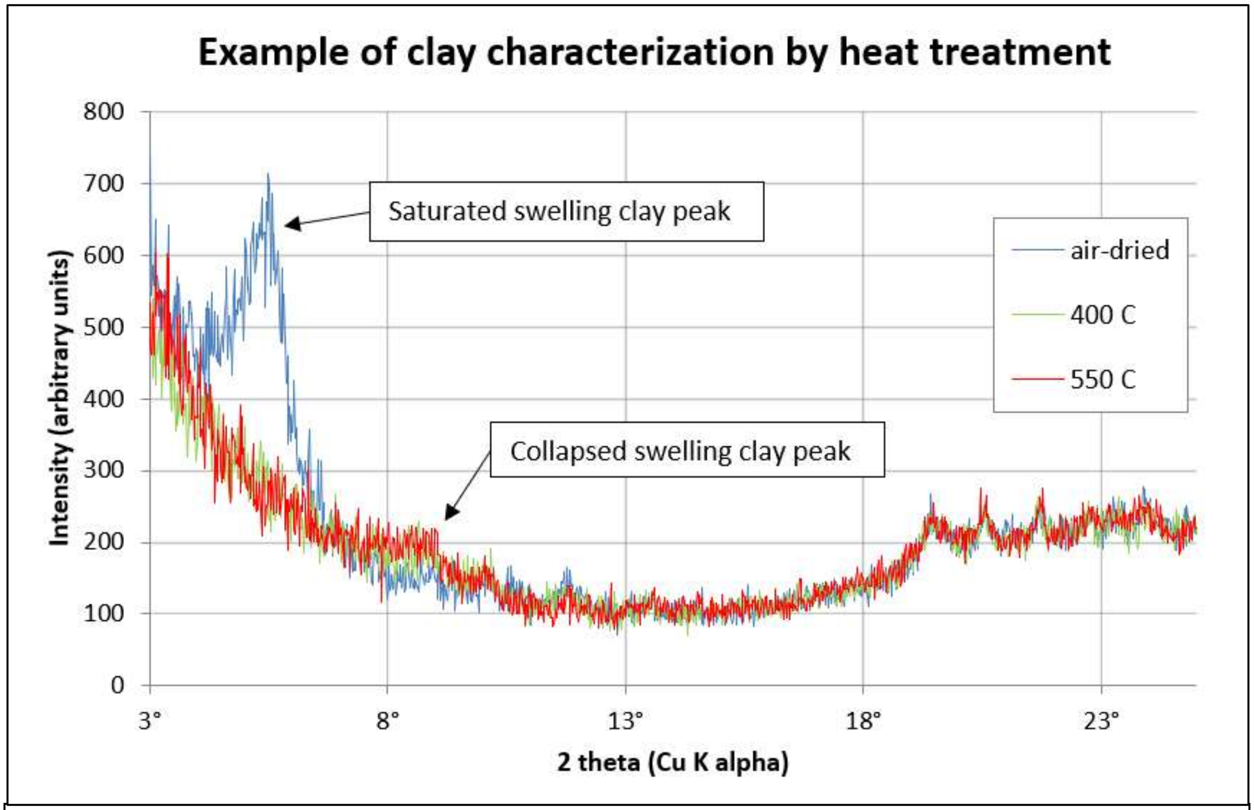


Fig. 2, Heat treatment test performed using XRD on sample CU-15 showing a 10Å clay peak that collapses after being heated to 400° C for 30 minutes.

1. Mineralogical comparison between stream sediments and bedrock -

In many cases, samples from watersheds underlain by similar bedrock (Case and Holcombe, 1980) contain similar minerals, but have significant variability in the quantity of each mineral (Fig. 3, 4). Watersheds where 100% of the underlying bedrock is metamorphic contain large amounts of quartz. Calcite is the second-most dominant mineral in streams dominated by metamorphic bedrock except in sample CU-101 where feldspars are equally abundant and sample CU-110 where feldspars are more abundant. Samples from watersheds underlain entirely by marine carbonate all contain quartz and calcite. 75% of 100% metamorphic samples contain feldspars, and half contain pyroxenes. The 100% marine carbonate basins can be divided into two distinct groups: 1) the larger watersheds, CU-120 and CU-121 (both ~50 km^2) which both contain large amounts of clay minerals, and 2) the smaller watersheds, CU-122 (~2 km^2) and CU-132 (~23 km^2) which both contain no detectable clay.

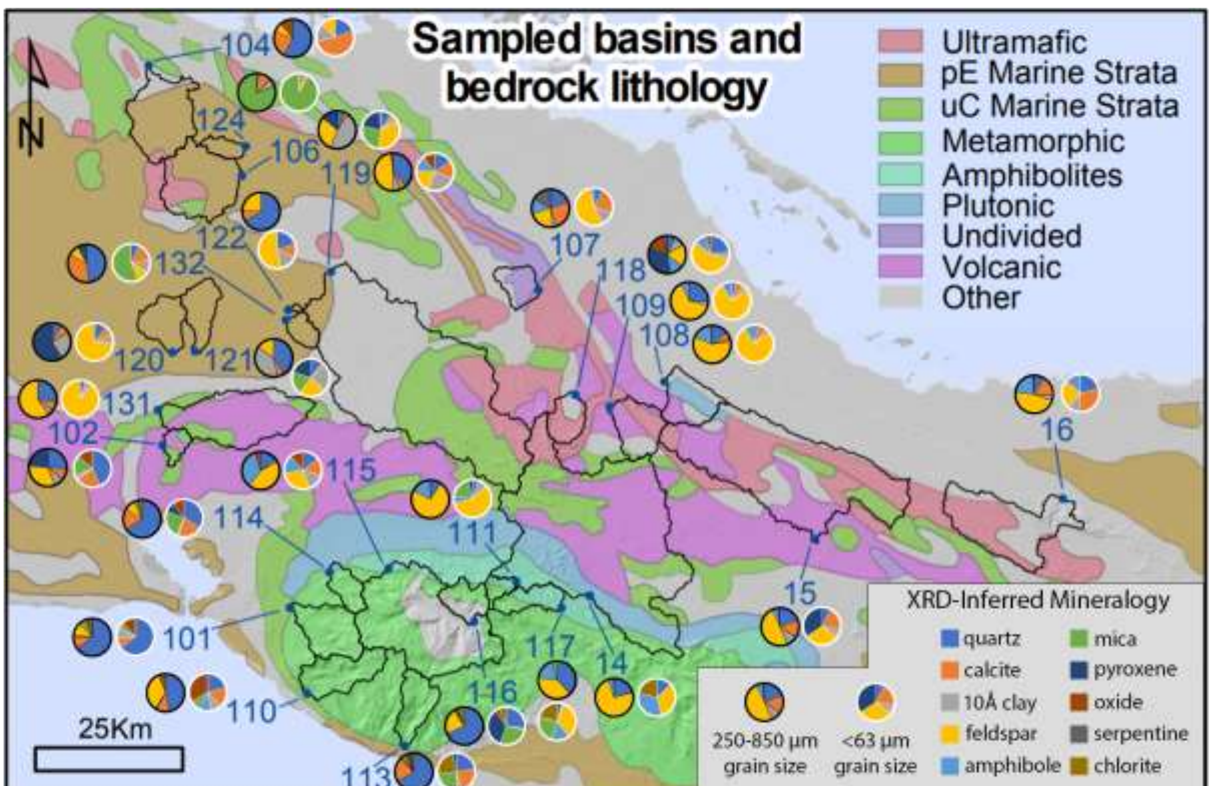


Fig. 3, Map showing sampling locations and watershed area, underlying lithology, and XRD-derived mineralogy data in two grain sizes. Modified from Bierman et al., (2020).

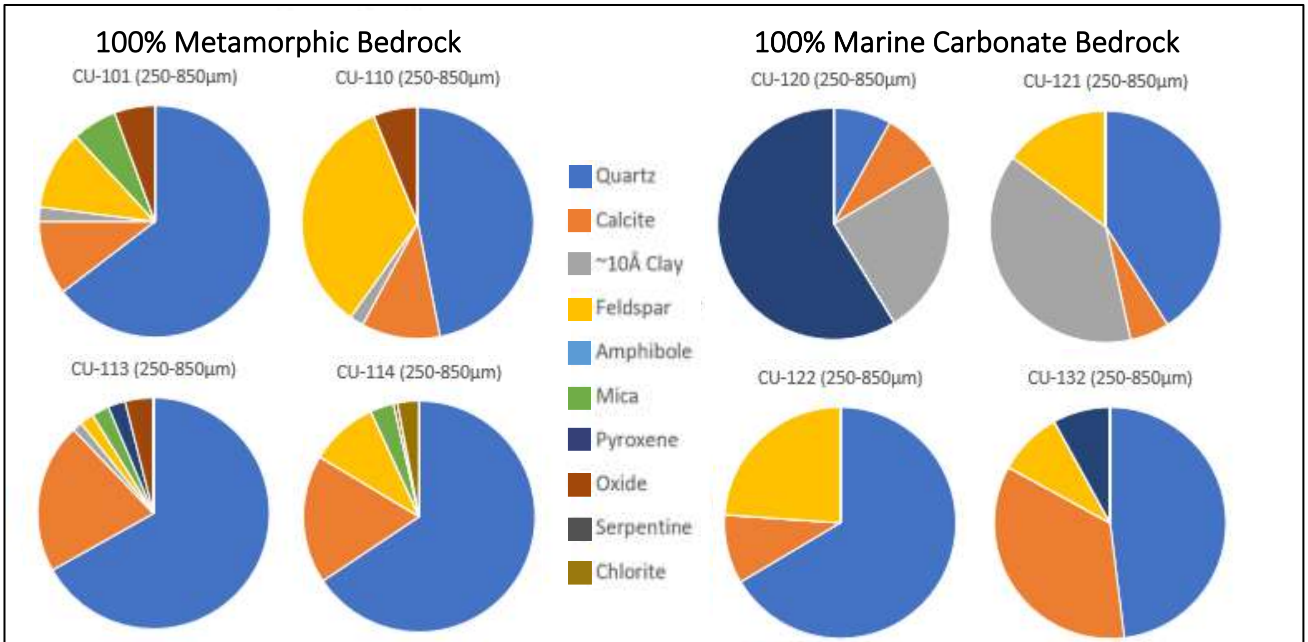


Fig. 4, Pie charts showing the mineralogic composition of samples (measured using XRD) grouped into watersheds with the same underlying bedrock units. Grain size and sample ID are shown above each pie chart.

Correlations between watershed lithologic percent coverage and sediment mineral percentages are significant ($p < 0.05$) in many cases with correlation coefficients reaching 0.89 (silicic plutons vs. feldspar in the fine grain size; Table 2). Notable correlations between percent lithologic coverage and percent mineral composition include: 1) Marine carbonate lithologic units show no correlation with calcite; are negatively correlated with amphibole; and are positively correlated with clay and mica (Fig. 5); 2) Metamorphic lithologic units are positively correlated with quartz, mica, and oxide, and are negatively correlated with felspar; 3) Mafic lithologic units are positively correlated with feldspar, pyroxene, and amphibole and are negatively correlated with calcite, clay, and mica; 4) Silicic plutons are negatively correlated with quartz, calcite, mica, clay, and pyroxene, and are positively correlated with feldspar and amphibole (Table 2).

Mineralogic ternary plots (Fig. 6A-B) show a similar distribution of minerals between grain sizes with most samples tending towards mafic-dominated and calcite-dominated with clay-dominated being the least abundant (clay proportion is <30% in 20/26 coarse grain samples (A), and 16/26 fine grain samples (B)). There are greater relative amounts of calcite in the coarse grain size (average of 38.4% in the coarse grain size, average of 29.1% in the fine grain size) and greater relative amounts of clay in the fine grain size (average of 18.4% in the coarse grain size, average of 24.6% in the fine grain size). The lithologic ternary plot based on the map from Case and Holcombe, 1980) indicates that most basins (17/26) are dominated by marine carbonate and mafic lithologies with only 9 watersheds dominated by metamorphic lithologies (Fig. 6C).

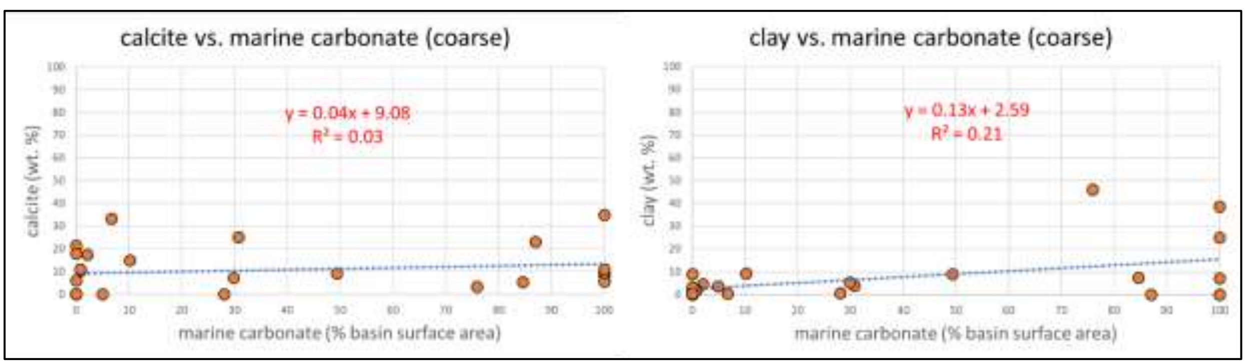
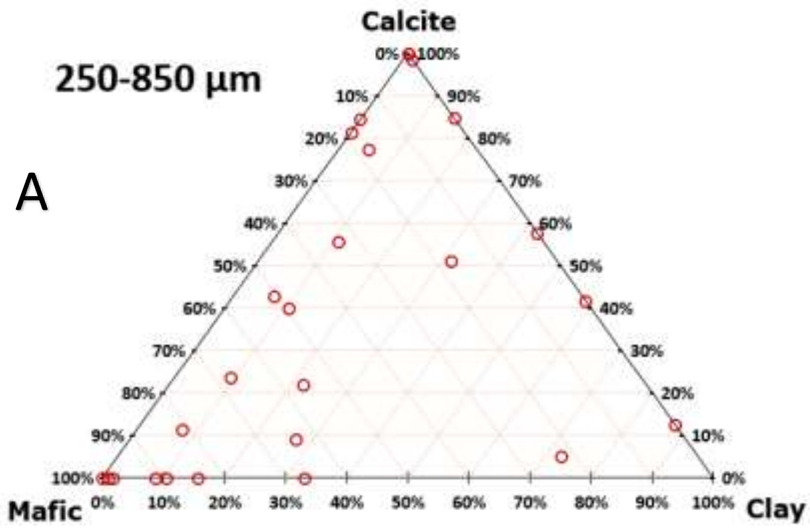
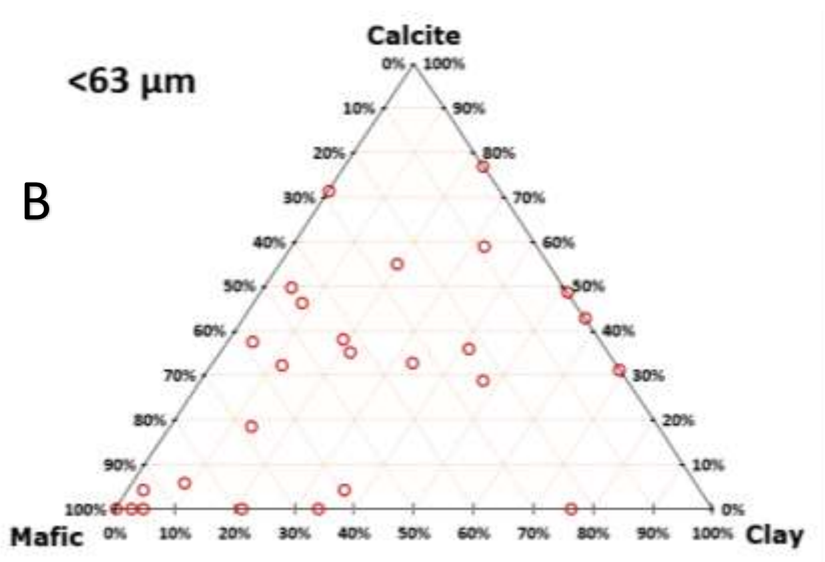


Fig. 5, regressions between marine carbonate lithologic units (Case and Holcombe, 1980) and mineralogy (this study) measured using XRD.

Table 2: Correlation coefficient values for XRD-derived mineralogy (wt. %) vs. bedrock (area %) in two grain sizes

	Marine Carbonate	Metamorphic	Mafic	Silicic Plutons
Quartz (250-850µm)	0.01	0.64	-0.43	-0.48
Quartz (<63µm)	-0.40	0.41	0.07	-0.50
Calcite (250-850µm)	-0.02	0.02	-0.41	-0.64
Calcite (<63µm)	-0.04	-0.08	-0.42	-0.61
Clay (250-850µm)	0.39	-0.61	-0.15	-0.38
Clay (<63µm)	0.28	0.03	-0.13	-0.07
Feldspar (250-850µm)	-0.51	-0.59	0.33	0.49
Feldspar (<63µm)	0.09	-0.47	0.28	0.89
Amphibole (250-850µm)	-0.47	-0.19	0.30	0.89
Amphibole (<63µm)	-0.33	-0.41	0.05	0.36
Mica (250-850µm)	0.27	0.49	-0.09	-0.41
Mica (<63µm)	0.47	0.71	-0.24	-0.40
Pyroxene (250-850µm)	0.35	0.31	0.29	
Pyroxene (<63µm)	0.08	-0.12	0.10	-0.74
Oxide (250-850µm)	-0.15	0.39	0.15	0.80
Oxide (<63µm)	-0.39	0.45	-0.05	-0.40
Serpentine (250-850µm)	-0.12		-0.34	
Serpentine (<63µm)				
Chlorite (250-850µm)	-0.26	0.32		-0.40
Chlorite (<63µm)	-0.13	0.04	-0.09	0.08
Zeolite (250-850µm)	-0.23	-0.49	0.28	-0.58
Zeolite (<63µm)	-0.26	0.28		
significant correlation (p-value < 0.05)				
significant correlation in both grain sizes (p-value < 0.05, same sign)				
not enough data to calculate				





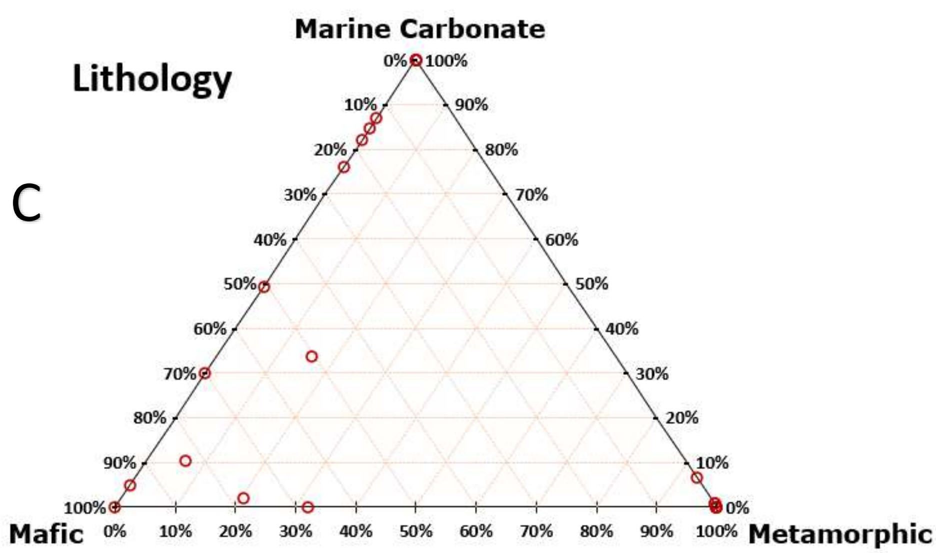


Fig. 6, A and B) ternary plots showing the spread of mineralogy in both grain sizes in terms of calcium, clay, and mafic mineral content. Values are adjusted so the sum of these minerals is 100% for each sample. Here, mafic minerals include amphibole, mica, pyroxene, and serpentine groups. C) ternary plot showing the spread of the three most prevalent lithologies based on Case and Holcombe, , 1980), also adjusted to sum to 100%.

2. Comparison between stream sediment mineralogy and stream water chemistry –

For most elements and compounds (N=17), stream water and sediment mineral composition are not correlated. Out of a total of 357 regressions, only 35 yield p values <0.05. Notable correlations comparing XRD data to water chemistry include: calcite with dissolved Ca; clay with dissolved Mg, DOC, dissolved Cl, dissolved K, and dissolved Na; mica with TDN; feldspar with dissolved SiO_2, and quartz inversely with dissolved SiO_2 (Fig. 7; table 3).

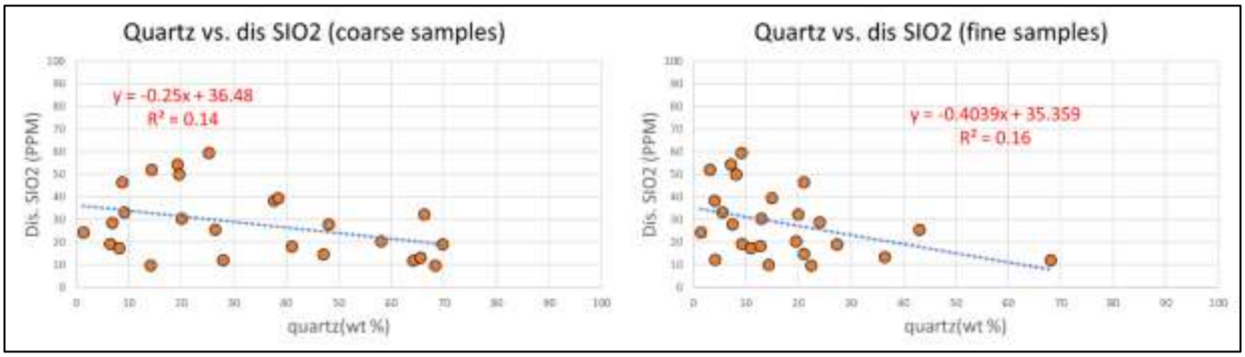


Fig. 7, Regressions between dissolved SiO_2 in stream water (Bierman et al., 2020) measured in the field and mineralogy in two grain sizes (<63 µm and 250-850 µm) (this study) measured using XRD.

Table 3: Correlation coefficient values for XRD-derived mineralogy (wt. %) vs. dissolved material in two grain sizes

	Cl	N	P	Na	Mg	Si	SiO2	K	Ca	Ti	V	Rb	Sr86	Sr88	Ba	DOC	TDN	Area
Quartz (250-850µm)	0.07	-0.48	-0.18	0.10	-0.34	-0.38	-0.38	-0.15	0.29	0.28	-0.58	0.02	0.24	0.24	0.18	-0.17	-0.27	-0.17
Quartz (<63µm)	-0.01	-0.45	-0.38	-0.19	-0.37	-0.40	-0.40	-0.16	-0.02	-0.01	-0.39	-0.20	-0.10	-0.10	-0.08	-0.16	-0.32	-0.13
Calcite (250-850µm)	0.05	0.20	-0.17	0.18	-0.09	-0.02	-0.02	-0.23	0.30	0.31	-0.17	0.10	0.28	0.27	0.17	-0.06	0.22	-0.21
Calcite (<63µm)	0.00	0.08	-0.14	-0.06	-0.09	-0.06	-0.06	-0.24	0.22	0.27	-0.12	0.03	-0.03	-0.04	0.01	-0.20	0.15	0.01
Clay (250-850µm)	0.52	-0.16	-0.15	0.30	0.07	-0.18	-0.18	0.59	0.04	0.02	0.02	0.63	0.26	0.27	0.25	0.57	-0.14	-0.05
Clay (<63µm)	0.48	0.00	0.26	0.44	0.46	0.18	0.18	0.44	0.29	0.26	-0.01	0.52	0.44	0.45	0.40	0.16	0.24	0.12
Feldspar (250-850µm)	-0.22	0.20	0.61	-0.28	0.21	0.50	0.50	0.07	-0.51	-0.49	0.68	-0.28	-0.47	-0.47	-0.21	-0.01	0.11	0.49
Feldspar (<63µm)	0.10	0.06	0.31	0.24	0.41	0.36	0.36	0.32	-0.14	-0.15	0.37	0.21	-0.01	-0.01	0.18	0.45	0.04	-0.02
Amphibole (250-850µm)	-0.32	0.23	0.08	-0.26	0.06	0.32	0.32	-0.23	-0.30	-0.27	0.33	-0.41	-0.37	-0.36	-0.28	-0.36	-0.03	0.08
Amphibole (<63µm)	-0.20	0.23	0.25	-0.25	0.01	0.25	0.25	-0.03	-0.37	-0.37	0.33	-0.37	-0.34	-0.34	-0.23	-0.27	0.01	0.61
Mica (250-850µm)	-0.17	0.43	-0.20	-0.09	0.04	-0.09	-0.09	-0.30	0.33	0.35	-0.14	-0.13	0.09	0.09	-0.15	-0.23	0.41	-0.15
Mica (<63µm)	-0.02	0.25	-0.28	0.03	-0.21	-0.24	-0.24	-0.22	0.22	0.21	-0.24	0.02	0.28	0.28	-0.07	-0.12	0.15	-0.34
Pyroxene (250-850µm)	0.38	-0.20	-0.23	0.30	0.07	-0.18	-0.18	0.41	-0.02	-0.09	-0.07	0.34	0.20	0.20	0.11	0.44	-0.26	-0.19
Pyroxene (<63µm)	0.01	-0.22	0.06	0.04	0.01	-0.02	-0.02	0.20	0.05	0.05	0.02	0.19	0.07	0.07	0.07	0.04	-0.20	0.04
Oxide (250-850µm)	-0.25	-0.33	-0.32	-0.28	-0.09	-0.24	-0.24	-0.28	-0.15	-0.11	-0.30	-0.12	-0.26	-0.26	-0.31	-0.13	-0.14	-0.04
Oxide (<63µm)	-0.11	-0.35	-0.20	-0.31	-0.32	-0.33	-0.33	-0.09	-0.14	-0.14	-0.27	-0.24	-0.18	-0.19	-0.22	-0.21	-0.36	0.03
Serpentine (250-850µm)	-0.13	0.42	0.11	-0.07	0.26	0.30	0.30	-0.19	0.08	0.10	-0.03	-0.17	-0.05	-0.06	0.23	-0.11	0.33	-0.13
Serpentine (<63µm)																		
Chlorite (250-850µm)	-0.18	-0.20	-0.15	-0.19	-0.29	-0.21	-0.21	-0.23	0.02	0.03	-0.35	-0.24	-0.08	-0.08	-0.10	-0.27	-0.28	-0.15
Chlorite (<63µm)	-0.25	0.00	0.04	-0.19	-0.30	-0.09	-0.09	-0.15	-0.17	-0.19	-0.06	-0.32	-0.19	-0.19	-0.20	-0.29	-0.05	0.21
Zeolite (250-850µm)	-0.01	0.03	0.33	-0.03	0.29	0.33	0.33	0.11	-0.05	-0.06	0.48	-0.01	-0.13	-0.13	-0.03	-0.01	0.06	0.27
Zeolite (<63µm)	-0.14	-0.20	-0.18	-0.13	-0.28	-0.26	-0.26	-0.15	-0.05	-0.05	-0.26	-0.12	-0.09	-0.10	-0.18	-0.16	-0.06	-0.09

significant correlation (p-value < 0.05)

significant correlation in both grain sizes (p-value < 0.05, same sign)

not enough data to calculate

3. Comparison between stream sediment mineralogy and elemental composition -

Out of 588 regressions comparing XRD-derived mineralogy and XRF-derived elemental content, 103 have p-values of <0.05. Correlations were generally weak with an average correlation coefficient (r) of 0.22. Notable correlations include calcite with Ca, feldspar with Na, and amphibole with Fe (Table 4, see data repository).

4. Comparison between stream sediment mineralogy, erosion data, chemical weathering, and slope -

When comparing ^{10}Be-derived erosion data (Campbell et al., 2019) to XRD-derived mineralogy data (this study), calcite and clay are both negatively correlated with erosion rates while feldspar, amphibole, and chlorite are both positively correlated with erosion rates. Additionally, clay is positively correlated with chemical lowering rates (Bierman et al., 2020). All other correlations had p values >0.05 (Table 5). Although quartz content is not associated with weathering rates, it is worth noting that quartz content determined by XRD is positively correlated with mean basin slope values (p values: 0.036 in the fine fraction, 0.055 in the coarse fraction). Additionally, amphibole is positively correlated with watershed area (Table 3).

Table 5: Correlation coefficient values for XRD-derived mineralogy (wt. %) vs. erosion and chemical lowering rates in two grain sizes

	10-Be Erosion rate (m/Myr)	Chemical lowering (m/Myr)
Quartz (250-850µm)	-0.25	0.02
Quartz (<63µm)	-0.12	-0.27
Calcite (250-850µm)	-0.51	0.17
Calcite (<63µm)	-0.46	0.00
Clay (250-850µm)	-0.37	0.03
Clay (<63µm)	-0.28	0.58
Feldspar (250-850µm)	0.51	0.04
Feldspar (<63µm)	0.14	0.19
Amphibole (250-850µm)	0.49	-0.04
Amphibole (<63µm)	0.57	-0.08
Mica (250-850µm)	0.26	-0.31
Mica (<63µm)	-0.12	-0.03
Pyroxene (250-850µm)	-0.10	-0.21
Pyroxene (<63µm)	-0.10	0.12
Oxide (250-850µm)	0.09	-0.32
Oxide (<63µm)	-0.05	-0.37
Serpentine (250-850µm)	-0.14	0.12
Serpentine (<63µm)		
Chlorite (250-850µm)	0.04	-0.11
Chlorite (<63µm)	0.41	-0.26
Zeolite (250-850µm)	-0.02	0.14
Zeolite (<63µm)	-0.10	-0.28
significant correlation (p-value < 0.05)		
significant correlation in both grain sizes (p-value < 0.05, same sign)		
not enough data to calculate		

Discussion

XRD analysis of sediment provides insight into how the presence of minerals in stream sediment can be used as a marker for the presence of specific lithologic units in tropical climates with high weathering rates. Overall, we find that sediment mineralogic composition does not quantitatively predict the exposed area of lithologic units due to variations in the capacity of each lithologic unit to produce sediment and for that sediment to be transported intact, downstream. In central Cuba, we find that calcite, amphibole, and swelling clay content can be used to infer weathering intensity and water chemistry.

The presence of swelling clays (smectites and vermiculites) in every tested sample (N = 9) indicates significant chemical weathering, as expected in wet, tropical environment such as Cuba (Reynolds, 1971; Liu et. al., 2009; Figure 3). Swelling clays with peaks between 9.5 - 10.5 Å that collapse between 400-500˚ C, as observed in this study, can be identified more specifically as either montmorillonite or vermiculite (Brindley, 1952). Additional testing is required to distinguish between these phases. Because swelling clays were identified in every sample tested with heat treatment across watersheds with diverse lithologies, we infer that all clays identified and quantified through Rietveld analysis are swelling clays.

Variations in composition between grain sizes are a result of preferential weathering as is common and expected in studies of unconsolidated stream sediment (Johnsson, 1993). Based on mineralogy quantification from XRD (Table 1), minerals containing resistant elements (Si) are more abundant in the coarse fraction (quartz), minerals containing less-resistant elements (Ca, Na, K) are more abundant in the fine fraction (mica, clay, and calcite; Roser, 2000). It is also likely that hydrodynamic sorting contributes to grain size variations, although this is not directly shown by our data.

1. XRD determined composition vs mapped lithology -

Although our correlations between XRD-derived mineralogy and mapped lithology are too weak to quantify bedrock types from stream sediment composition, the presence of certain minerals is useful as an indicator that lithologic units are present in a given watershed (ex: feldspar indicates igneous, mica and clay indicate non-igneous, Table 2).This notion is especially useful for inferring basin lithology in areas such as central Cuba where unweathered outcrops are uncommon.

We find that stream sediment composition reflects source geology with some discrepancies that can be attributed to stream dynamics. Our results show that quartz in sediment is more prevalent than suggested by lithologic units identified by Case and Holcombe (1980). This discrepancy could be due to: 1) broadly-applied mapping techniques that do not identify smaller, quartz-rich lithologies that could heavily influence stream sediment composition and/or 2) weathering processes resulting in a higher presence of quartz in stream beds than in bedrock, because is highly resistant to weathering (Folk, 1980; Johnsson, 1993). It is possible that quartz would dominate stream sediment even where bedrock is quartz-poor because Cuba has high weathering rates that would dissolve most other minerals more quickly (Rad et al., 2013). Our data confirm that quantifying basin scale geology from sediment composition can be unreliable since each lithologic unit varies in its capacity to produce debris based on a multitude of factors such as crystal size, rock isotropy, chemical stability, and microfabric (Palomares and Arribas, 1993).

Ternary plots (Fig. 6) suggest that calcite is associated with marine carbonate lithologic units, mafic minerals are associated with mafic lithologic units, and swelling clays are a measure of weathering intensity. The lesser amounts of clay content in the coarse grain size is likely a result of hydrodynamic sorting, while the lesser amounts of calcite content in the fine grain size could be due to small grains of calcite being easily dissolved, keeping calcite weight content low. Although stream sediment composition is less uniform than lithologic composition, the dominance of calcite and mafic minerals (in wt %) in both grain sizes (Fig. 6A-B) can be associated with the watersheds being dominantly marine carbonate and dominantly mafic in the lithologic ternary plot (Fig. 6C).

2. XRD-determined composition vs water chemistry -

Water chemistry data support the results of stream sediment mineralogy quantification while also providing insight into how mineralogy can be used to infer weathering processes. Regression analyses suggest that dissolved silica is derived from feldspar, with little to no contribution from quartz as indicated by the correlations of dissolved SiO_2 with feldspar (positively) and quartz (inversely) coupled with the observation that feldspar and quartz are commonly opposite in their lithologic correlations (Table 2; Table 3). This makes sense in an environment with high weathering rates because quartz is significantly more durable and chemically stable than feldspar (Folk, 1980). The correlations of clay content with dissolved Mg and dissolved Na are consistent with the presence of smectite minerals, further supporting the presence of swelling clays. Additionally, the correlation between clay and DOC shows the usability of clay content as a marker for significant weathering since higher DOC levels, especially in transport-limited catchments, are commonly associated with higher weathering rates (McCracken, 1998). The correlation between clay and both dissolved K and dissolved Cl suggests that swelling clays may also be useful for predicting the presence of these elements in stream water. The strong correlation between calcite and dissolved calcium supports the notion that calcite is highly susceptible to weathering, making its presence useful for predicting water chemistry.

3. XRD-determined composition vs XRF -

Correlations between XRF and XRD data from the same stream sediment samples show agreement in the presence of calcite (from Ca) and amphibole (from Fe), with XRF data suggesting that the feldspars identified using XRD are mainly sodic due to the correlation between feldspar content and elemental Na content (See data repository). However, we find that most mineralogy data do not correlate with elemental data. This could be a result of highly impure or heterogenous sample material or highly complex mineralogic compositions that cannot be precisely quantified within the scale of this study.

4. XRD-determined composition vs. erosion and chemical weathering rate estimates -

Our results show that calcite, swelling clay, feldspar, and amphibole content in stream sediments can be used to indirectly infer weathering rates as well as the relative intensities of physical and chemical weathering. The correlation of feldspar and amphibole with erosion rates and the inverse correlations of calcite and clay with erosion rates show that varying amounts of these minerals in watersheds with similar lithologies can be used to infer physical weathering intensity across watersheds. Samples with less clay and calcite content and/or more feldspar and amphibole content demonstrate more intense weathering. These findings also allow for predictions of landscape-scale mineral distribution in areas with high weathering rates, where upland, "source" sediment reservoirs will be enriched in amphibole and feldspar but depleted in clay and lowland, "sink" reservoirs will be depleted in calcite but enriched in clay content. This agrees with the low durability of calcite relative to other minerals as well as the easily transportable nature of fine clays and the moderate durability of amphibole and feldspar (Folk, 1980). In agreement with Thorpe et al. (2019), Clay mineral content is also positively correlated with chemical lowering rates, suggesting that chemical weathering rates can be inferred by assessing the amount of clay in a watershed. Swelling clays are therefore a highly useful, although complex, marker of weathering intensities as their quantity is indicative of the intensity of physical and chemical weathering as well as their comparative rates within a watershed, especially when observed in combination with feldspar, calcite, and amphibole contents.

We find that the central Cuban landscape is transport-limited as opposed to weathering-limited as is expected in a low-relief environment with intense weathering. Although quartz dominates our samples, we find no association between quartz content and weathering rates as would be expected due to the ultra-stable nature of quartz, which is generally expected to cause a relative increase in quartz content with increasing weathering rates (Folk, 1980). However, because chemical weathering rates in Cuba are very high on average, it is likely that all the sampled watersheds are transport limited as opposed to being weathering limited (Fig. 8; Rad et al., 2013). This would cause an increase in quartz content in sediments across all watersheds as shown by Johnsson (1993), and would mean that variations in quartz content across watersheds are determined by transport capacity, not weathering rates. This notion is supported by the positive correlation between watershed slopes and quartz.

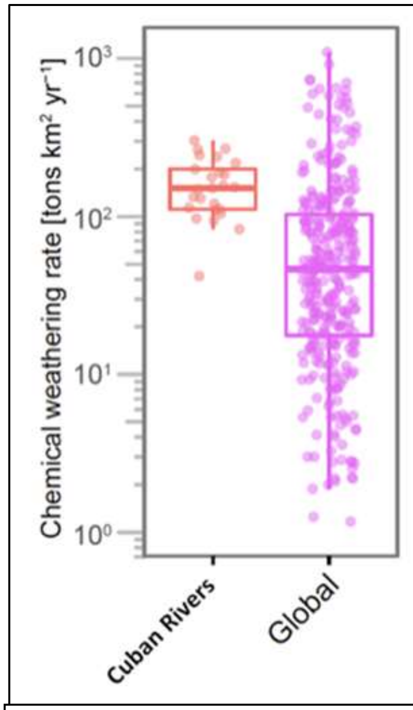


Fig. 8, Adapted from: Bierman et. al. (2020), data are from Rad et. al. (2013).

Our findings indicate that stream sediment mineralogy data is useful for predicting certain water quality parameters over long periods of time from a single sample, which is not possible with water samples. This relies on the notions that 1) water quality can be inferred from stream sediment mineralogy, and that 2) central Cuban streams are transport limited. Because the residence time of sediment in stream channels is significantly longer than that of travelling water and dissolved material, this sediment provides a record of the minerals that are present and influencing water chemistry such as calcite and swelling clays.

5. Summary -

We find XRD-derived stream sediment mineralogy to be a useful metric by which to infer landscape characteristics and processes such as erosion, chemical weathering, water quality, and exposed lithology in warm, tropical, densely vegetated environments such as central Cuba. In agreement with Palomares and Arribas (1993) and Johnsson (1993), our results show that the composition and exposed area of bedrock lithology cannot be quantified based on stream sediment mineralogy alone, but less durable and stable phases can be used as markers for the presence of certain lithologic units within a watershed: calcite marks carbonate units; mafic minerals mark mafic igneous units; feldspar marks igneous units.

We show that both feldspar and amphibole abundances are greater in watersheds with higher ^{10}Be-determined erosion rates, indicating that they can be used to infer relative weathering rates. Additionally, debris in areas with especially high erosion rates may become enriched with amphibole and feldspar minerals. We also show that calcite and swelling clays are more abundant in areas with lower erosion rates, and thus can also be used to infer weathering rates. Soils in areas with especially high erosion rates may become depleted of calcite and clay minerals. The opposite is true towards the watershed sink, where deposits will be depleted of amphibole and feldspar minerals but high in calcite and swelling clay minerals.

Clays and calcite minerals quantified in this study are not only useful as markers for physical weathering, but also as proxies for chemical weathering rates and water chemistry. The correlation between chemical weathering rates and clay shows that the relative amounts of chemical weathering that occur across watersheds can be inferred from the swelling clay content of stream sediments. The correlations between 1) swelling clay content, dissolved Mg and Na, and 2) calcite content and dissolved Ca indicate that these minerals have a significant impact on water chemistry. Swelling clays may also be useful for predicting dissolved K, dissolved Cl, and DOC. Additionally, our findings suggest that stream sediment samples from transport-limited regions such as central Cuba provide insight into water chemistry over longer periods of time than traditional water samples through the quantification of minerals such as swelling clays and calcite.

III. Future Work

I show that X-ray diffraction of stream sediments from central Cuba provides insight into landscape weathering processes, allowing for a more mechanistic understanding of watershed dynamics as rock is weathered, transported, dissolved, and deposited. A more detailed analysis of the specific mineral phases present in these samples could reveal more about the relationship between stream sediments and the landscape mechanisms of stream systems in tropical areas. This could be achieved through methods such as: 1) isolating certain minerals magnetically, through density separation, or by other means and scanning them in their pure form to reduce noise in the resulting scans, 2) performing single-crystal X-ray diffraction on select grains, 3) performing additional tests on clay phases, such as ethylene glycol treatment, and 4) observing sediment microscopically. Microscopic mineral point-counting would also provide insight into the structure of the grains which is useful for discerning weathering processes. It could also be useful to perform scans on stream sediments from other watersheds in Cuba, providing a more complete study of the island.

Future work can therefore be done in multiple ways, 1) performing more in-depth tests on the stream sediments studied here, and/or 2) expanding this study to include more watersheds. I suggest expanding the study by studying additional watersheds to see if the trends observed in the basins studies here are reproducible in similar areas. After observing the results from these additional watersheds, more in-depth analyses of certain minerals may be useful for more precisely gauging how the weathering processes of these minerals are being influenced and unfolding across Cuba and other tropical islands.

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