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Carbon Flux and Weathering Processes in Icelandic Glacial-Fed Rivers

Allison Quiroga
Western Kentucky University, allisonquiroga353@topper.wku.edu

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CARBON FLUX AND WEATHERING PROCESSES IN ICELANDIC GLACIAL-FED RIVERS

A Thesis
Presented to
The Faculty of the Department of Geography and Geology
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In Partial Fulfillment
Of the Requirements for the Degree
Master of Science

By
Allison Quiroga

May 2018

CARBON FLUX AND WEATHERING PROCESSES IN ICELANDIC GLACIAL-FED RIVERS

Date Recommended 17 APRIL 2018

Dr. Jason Polk, Director of Thesis

Dr. Royhan Gani

Thorsteinn Thorsteinsson

Dr. Thorsteinn Thorsteinsson

5/11/18

Dean, Graduate School Date

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Two years ago, at the beginning of this journey, writing an acknowledgements section seemed like a lifetime away. Today, after having completed the journey and reflecting on that time two years ago seems yet again, like a previous lifetime. There's an unexplainable and very conceptual aspect to graduate school that I didn't fully comprehend until much later I would have liked. This reason, and for many others, this graduate experience challenged me academically, emotionally, and professionally. Despite and due to this struggle, I feel like a more informed and educated Geoscientist. I hope to continue to build and develop the skills I've learned throughout this process and applies those skills to be a better and more thorough researcher and professional. My journey has had many lows and many highs, and I was very lucky to have professional and personal support and assistance from many people.

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CARBON FLUX AND WEATHERING PROCESSES IN ICELANDIC GLACIAL-FED RIVERS

Allison Quiroga May 2018 95 Pages

Directed by: Drs. Jason Polk, M. Royhan Gani, Þorsteinn Þorsteinsson

Department of Geography and Geology Western Kentucky University

An investigation into the carbon dynamics and weathering processes occurring in Icelandic glacial-fed streams was conducted during the spring to summer seasonal transition in June of 2017. Four major outlet rives were sampled from the glaciers of Gígjökull, Steinsholtsjökull, Sólheimajökull, and Falljökull. Markarfljót, the major river that Gígjökull, Steinsholtsjökull, and many other glaciers drain into, was also sampled. Longitudinal sampling occurred at all sites to capture downstream trends in the hydrogeochemistry and carbon dynamics. Distinct differences in geochemistry between glacier surface meltwater, sub-glacial waters, pro-glacial lake water, and post-mixed downstream samples were evident in the data. Glacier surface streams were characterized by relatively colder water temperatures, lower specific conductivity, lower total dissolved solids (TDS) and ion concentrations, and more enriched $\delta^{13}C_{DIC}$ values than downstream samples. The THINCARB model was used to calculate the total dissolved inorganic carbon (TDIC), excess partial pressure of carbon dioxide ($EpCO_2$), and percent contribution to TDIC by bicarbonate (HCO_3), carbonate (CO_3), and carbonic acid and dissolved CO_2 (H_2CO_3). All sites showed a slight decreasing trend in DIC and $EpCO_2$ downstream. The calculated CO_2 flux ranged from 1.14×10^7 g/yr to 2.80×10^9 g/yr. The DIC flux ranged from 6.81×10^7 g/yr to 8.44×10^9 g/yr. The average carbon within the CO_2 fluxing in these rivers accounts for 0.0004% of the annual, global flux of carbon. The $\delta^{13}C$ values were the most variable throughout the study and indicate there are

multiple sources influencing the river downstream. This study suggests that, despite previous assumptions and estimations, these glacial-fed rivers act as sources of CO_2; however, the samples from this study only provide a snapshot into the carbon flux dynamics during the Spring to Summer seasonal transition. In most samples, HCO_3 was the dominant species contributing to DIC content within the rivers, suggesting that DIC is being transported to the ocean as HCO_3 but sourced to the atmosphere as CO_2. By acting as sources of CO_2 to the atmosphere, the process of glaciers melting, which drive geochemical processes within the rivers, are contributing to a positive feedback loop with respect to global warming.

CHAPTER ONE: INTRODUCTION

1.1 Research Overview

Consensus within the scientific community about the reality and impact of anthropogenic CO_2 influence on global climate makes climate change an important and relevant environmental issue (Barnett et al. 2001; Matthews et al. 2004); however, climate change investigations in the Arctic are increasingly urgent, as there are gaps in understanding how continued warming from increasing atmospheric carbon dioxide (CO_2) levels will impact the region (Striegl et al. 2005; McGuire et al. 2009). There is agreement that glaciers and ice sheets, or caps, (GICs) are melting in response to climate change (Bøggild et al. 2010; Stibal et al. 2012; IPCC 2013), which is one reason GICs are included in the global climate change discussion. The glacier melting process contribution to atmospheric CO_2 remains understudied and is still not well understood or quantified fully within the carbon budget.

GICs, particularly temperate ones like those found in Iceland, are sensitive and dynamic systems. Because of the massive size of GICs, even minor changes in melting rates can have a significant impact on regional and global environmental systems, due to the large volume of frozen water stored in glaciers. CO_2 is of particular concern in climate change discussions, because it exists in the highest concentrations among greenhouse gases (GHG) and is directly linked to temperature increases (Houghton et al. 1990; Nakicenovic et al. 2000; IPCC 2013). Rivers, including glacial meltwater-fed rivers, serve as an important transport mechanism for various ions, including forms of carbon (Thomas and Raisewell 1984; Nikanorov and Brazhnikova 2009; Jarvie et al. 2017). CO_2 concentrations can increase from various mechanisms and processes, such as volcanism, plant respiration, and combustion of fossil fuels; these processes are referred to as carbon sources. Other mechanisms, such as photosynthesis, silicate and carbonate rock weathering, and carbonate accretion, are examples of carbon sinks, which remove carbon from active systems and the atmosphere. The storage capacity of glaciers, described above, typically makes them a sink; however, as glaciers melt, CO_2 trapped in ice is released and, through atmospheric exchange and weathering from glacial runoff, impacts carbon stored in the ice and bedrock, which can result in glacial environments acting as a complex carbon and GHG source/sink system (Oechel et al. 1993; Stibal et al. 2012). Understanding carbon flux mechanisms in the Arctic, and Iceland in particular, is important to refine the carbon flux estimates that exist for the region, because there is a lack of studies on this topic (Sarin et al. 2002; Finlay et al. 2006). Furthermore, quantifying the carbon flux and understanding how the various processes influence the carbon dynamics occurring within Iceland's glacial outlet rivers is paramount to anticipating, with accuracy, how future changes in climate may affect the country and surrounding region.

The purpose of this study was to examine the source, transport, and flux of dissolved inorganic carbon (DIC) in Icelandic glacial rivers by analyzing the DIC in glacial meltwater-red rivers. An additional objective of this study was to evaluate the downstream, or longitudinal, changes in carbon within the various rivers. This study focused on the main drainage rivers from Gígjökull, Steinsholtsjökull, and Sólheimajökull glaciers in the southern region and Falljökull in the southeastern region of Iceland. Carbon flux data were extrapolated to determine a conservative estimate of total DIC flux from various glaciers, as measured from glacial meltwater-fed rivers.

This research produced a quantitative investigation of the transport and flux of carbon stored within snow and ice in Iceland by analyzing the dissolved CO_2 in river runoff from those components of the cryosphere. These data, when combined with discharge, meteorological fluctuations, and isotopic analysis to source the carbon, provide a holistic dataset for quantifying and characterizing the CO_2 that may enter the environment from rapidly melting GICs. This research answered the following questions:

- How do different weathering processes affect the carbon dynamics in Icelandic glacier-fed rivers?
    - What is the dominant weathering process contributing to the dissolved inorganic carbon flux within the study rivers?
- What is the flux of dissolved inorganic carbon and carbon dioxide at each of the study sites?

1.2 Glaciers and Ice Caps

GICs are significant features on the planet that cover approximately 10% of the world's land surface (USGS 2016). Glacial environments, including glaciers, ice sheets, ice caps, and permafrost store approximately 75% of the planet's fresh water (Lutz et al. 2015; USGS 2016) and serve as important proxies for examining various global and regional issues (Blunier and Brook 2001; Mayewski et al. 2004). In response to rising temperatures, the melting of glaciers and ice sheets increases (Meier et al. 2007; Stibal et al. 2012). An increase in the magnitude and seasonality of glacier runoff is among the most significant hydrological consequences anticipated from future changes in climate (Aðalgeirsdóttir et al. 2006). Iceland's glaciers are especially sensitive to impacts from climate change, such as extended melting seasons and increased air temperature. A report published by Jóhannesson et al. (2007) estimates that within 150-200 years, Iceland will be without glaciers; the glacial melt will raise global sea level by nearly one centimeter.

During glacial melting, water and CO_2 stored in the glaciers are transported to the world's oceans within the meltwater. This process has a possibility of acting as a positive feedback loop with respect to global warming. Positive feedback loops occur when a small disturbance in a system increases the magnitude of the disturbance. The feedback loop described above is characterized by the release of CO_2 from melting glaciers, which increases atmospheric CO_2 concentrations and, in turn, increases global temperatures, which subsequently melt glaciers more rapidly. Alternatively, glacial meltwater rivers can act as a carbon sink, as glacial meltwater is a driving force behind chemical and mechanical weathering that occurs in the meltwater rivers; for this reason, rivers are often considered carbon sinks (Dessert et al. 2003), as the process of carbonate and silicate rock weathering uses CO_2 within the weathering reactions. Understanding the relationship between carbon in glaciers and hydrogeochemical processes in their associated rivers will help develop a more quantified understanding of how carbon is being transformed from source to sink within the environment of Iceland.

1.3 Rivers in the Global Carbon Cycle

According to Meybeck (1982), riverine transport of organic carbon from drainage basins to the world's oceans is a major component of the global carbon cycle; yet, few studies exist for Arctic rivers for DIC. The drainage basins of the entire Arctic region (19 million km^2) process about 11% of the global runoff (Lammers et al. 2001; Köhler et al. 2003) and the corresponding carbon flux in rivers from the watersheds to the Arctic ocean is one key connection between the terrestrial and marine components of the

Arctic's carbon budget. Globally, rivers transport organic, inorganic, and particulate carbon from their sources to the oceans, which are the world's largest carbon reservoirs; however, the ocean's ability to buffer global air temperature is diminished as more CO_2 is absorbed and introduced. Reduced buffering and absorption of CO_2 from the world's oceans promotes further global temperature rise and subsequently more rapid melting of GICs.

1.4 Iceland

There are several aspects that make Iceland sensitive and responsive to climate change and, therefore, of interest in carbon flux studies. Foremost, Iceland is in a unique geographic position in the North Atlantic Ocean just along the southern boundary of the Arctic Circle. Iceland's position between the warmer Irminger ocean current and colder east Greenland and east Iceland ocean currents has led to a historical sensitivity to changes in ocean circulation (Geirsdóttir and Eiríksson 1994). The Gulf Stream directs warm ocean currents from lower latitudes to the country's southern border, while cold water from the Arctic comes from the north. These influences are responsible for Iceland's glacial development and the temperate climate.

The rivers of Iceland are of economic concern as well. Hydroelectric energy, drawn from rivers, produces approximately 80% of the energy in Iceland. Not only is that energy used to provide service to residential consumers, but more importantly to provide cheap energy to industrial users. To meet these needs, various rivers have been diverted and altered to meet the rising demand. In addition, global interest continues to grow as Iceland becomes a more desirable tourist destination. The rivers, or more specifically the waterfalls, are some of the well-known landscape features that attract tourists. With over two million tourists a year and growing, the tourism industry in Iceland is quickly becoming prolific and could be impacted detrimentally by climate change as glaciers continue to melt away (Hamilton et al. 2005; Compton et al. 2015).

In Iceland, a third of the runoff to rivers is contributed from glacial melting (Björnsson and Pálsson 2008); these rivers are the dominant transport mechanism of carbon. While there has been considerable research on runoff and modelling, as well as discharge measurements, studies relating to carbon flux and transport in glacial-fed rivers in Iceland remain relatively understudied; however, several recent studies focused on the delineation of DOC in glacial environments (Anesio et al. 2010; Bhatia et al. 2013; Spencer et al. 2014; Hood et al. 2015), but studies of DIC are lacking in comparison. Several studies have established the importance of DIC in riverine systems by studying the dominance of DIC on the total dissolved carbon content in various rivers (Brunet et al. 2005; Huang et al. 2012; Jarvie et al. 2017). Based on existing studies, controls for riverine DIC are primarily associated with watershed inputs and in-stream geochemical and biologic processes (Tamooh et al. 2013). CO_2 exchange from the river with the atmosphere, carbonate and silicate weathering, primary production, and community respiration are all processes that contribute to DIC in rivers and need to be examined within the context of glacial-fed rivers in Iceland (McConnaughey et al. 1994; Amiotte-Suchet et al. 1999; Abril et al. 2005; Finlay and Kendall 2007).

CHAPTER TWO: LITERATURE REVIEW

2.1 Climate Change

Climate change is a controversial societal topic, especially when discussing global warming. A report in the 2001 IPCC assessment determined most of the observed warming over the last 50 years is likely due to the increase in GHGs, which agrees with recent assessments by the Intergovernmental Panel on Climate Change (IPCC 2013). There is evidence of a warming trend in climate that has been measured in different natural systems; some of the most serious implications involve water and its availability (Barnett et al. 2005; Milky et al. 2005). Climate change investigations in the Arctic are increasingly important, as there are gaps in the understanding of warming effects in the region (Striegl et al. 2005).

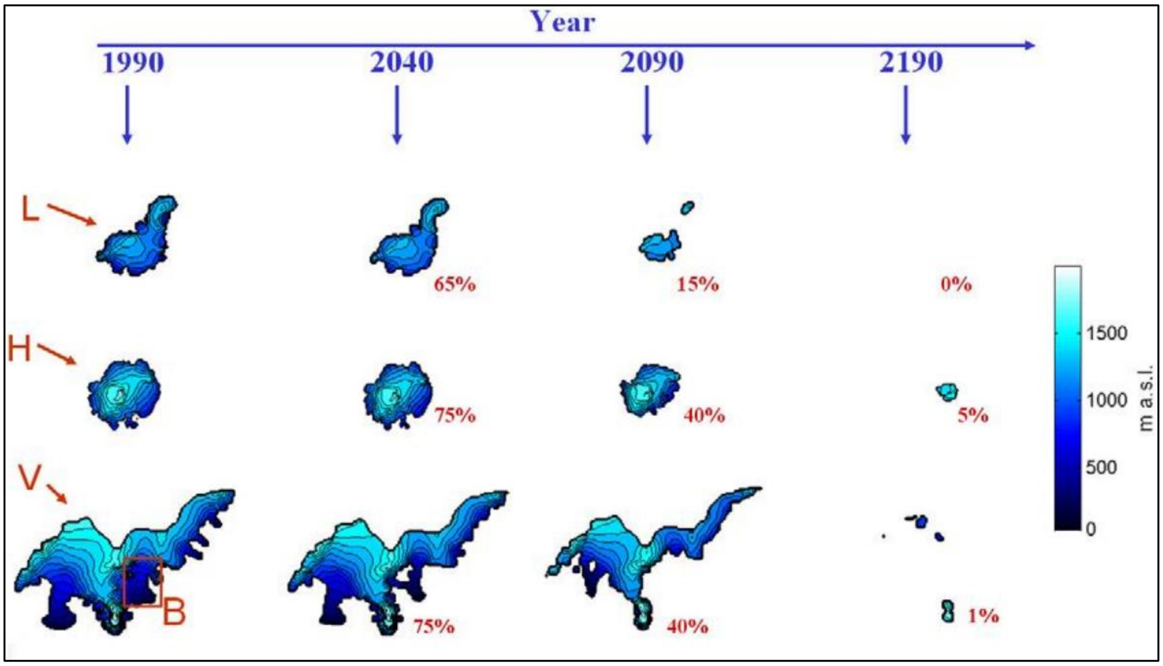


Figure 2.1: Anticipated loss of glacier extent in Langjökull (L), Hofsjökull (H), and Vatnajökull (V) by 2190 (Source: Aðalgeirsdóttir et al. 2006).

2.2 Glaciers

Globally, glaciers and ice caps cover approximately 10% of Earth's surface (Hood et al. 2015) and in Iceland, specifically, cover 10% of the land area (Björnsson and Pálsson 2008). Glaciers are defined as consistent, large bodies of ice and snow that form in cold environments, typically in the Arctic and Antarctic, and are big and thick enough to sink and move under their own weight (Benn and Evans 2014). Glaciers also form in high latitude regions and high elevation mountainous regions, such as the Himalayas, where precipitation rates are high. Glaciers form when the precipitation and compaction of snow in an area, termed accumulation, exceeds the removal of snow through melting and sublimation, termed ablation (Benn and Evans 2014). Over centuries, the consistent accumulation on a terrain leads to the formation of glaciers and ice caps. Glaciers are smaller than ice caps but form in the same conditions. GICs form primarily in the polar and sub-polar regions in geographic areas that are relatively flat and higher in elevation (Benn and Evans 2014). Glaciers can be categorized by size, shape, and behavior as well as thermal state. There are different types of glaciers in Iceland, including mountain, or valley, glaciers that form within steep, walled valleys and piedmont glaciers, which occur when valley glaciers spill into adjacent flatter areas and create "lobes." The thermal category of Iceland's glaciers is considered temperate, or "wet," as the GICs are at or near their melting point throughout the year, regardless of season.

Glaciers, temperate or polar, are sensitive and dynamic landforms (Oerlemans 1994). Melting occurs not only from air temperature variation, but also from basal melting from pressure and frictional movement of the glacier and short-wave radiation (Bøggild et al. 2010; Stibal et al. 2012). Because of their massive size, minor changes in melting rates and accumulation/ablation rates can have a significant impact on regional and global environments, due to the large volume of frozen water stored in glaciers. Some countries, such as Iceland, are reliant on glaciers to stimulate different aspects of their economy, such as hydropower, and tourism; however, there are larger-scale, worldwide implications from glacier responsiveness and sensitivity to climate change. To reference the basic life cycle of a glacier, it is formed through the accumulation of snow and ice and undergoes ablation frequently throughout the formation process; this relationship is referred to as mass balance (Braithwaite 1981; Dowdeswell et al. 1997). Influences on a glacier, such as temperature or precipitation changes, can result in the ablation rates overcoming the accumulation rate. When ablation exceeds accumulation, the result is increased removal of glacial material in a way that creates an imbalance in the system and overall net loss of snow and ice. This removal of material occurs from two different processes: the melting of snow and ice and sublimation (the transition of ice to water vapor).

Increases in surface temperature have significant consequences on the hydrological cycle. Warmer global temperatures have other implications in glacial environments, such as less precipitation during winter and earlier seasonal melting of winter accumulation (Barnett et al. 2005). Recent studies have also established that precipitation in glacial environments may increase in warmer months in response to increased global air temperatures (IPCC 2013). Melting glaciers are still a concern for regions that do not rely on glacier melt for aquifer recharge; elevated sea level and ocean salinity are hazards to all coastal areas.

The primary source of warming is a result of increased concentrations of greenhouses gases (GHG) in the atmosphere. CO_2, water vapor, methane, and nitrous oxide are the major GHGs (Rodhe 1990; Houghton et al. 1990; IPCC 2013). GHGs cause positive radiative forcing of the climate system and subsequent warming of surface temperatures (IPCC 2013). Results of this effect includes the thermal expansion of the shallow warmer ocean and melting of GICs, which both contribute to sea level rise and severely impacts coastal communities, which are threatened by overdevelopment and coastal erosion in response to global sea-level rise (Feagin et al. 2005). CO_2 is one of the most impactful GHGs and often is the focus of most climate change and global warming discussions, because it exists, in comparison to the other GHGs, in the highest volume in the atmosphere. In addition, CO_2 is a useful GHG to discuss, since multiple studies have determined historical trends and its association with changes in surface temperature.

An additional issue is the negative impact that increasing CO_2 concentrations have on coral reefs. Global warming and ocean acidification, which results in coral bleaching, compromise carbonate accretion, which influences the building mechanism of reefs (Hoegh-Guldberg et al. 2007). Since the early 1980s, episodes of coral reef bleaching and subsequent reef mortality, due primarily to climate-induced ocean warming, have occurred almost consistently on a yearly basis in one or more of the world's tropical or subtropical seas (Baker et al. 2008). As atmospheric CO_2 concentrations continue to rise and the DIC reservoir in the oceans increase, the effect on coastal reefs will continue to occur.

Glacial melting has equally impactful effects on the glaciers themselves. Glacier meltwater forms various features, such as moulins and surface streams, which also are involved in carbon storage and transport. In addition to those features, glaciers sequester carbon from autochthonous and allochthonous sources and release it to downstream environments (Hood et al. 2015). There is a substantial input of inorganic carbon from biologic sources beneath a glacier, where the bacteria must rely on limited remaining substrates for energy production (Christner et al. 2003; Christner et al. 2008). Smaller-scale changes occur within the life cycle of a glacier that can typically be observed in separate zones within it. Changes to the surface zone of a glacier, otherwise known as “supraglacial” zone, can typically be observed in different surface expressions. Englacial features form as water moves within the glacier and forms karst-like features. Subglacial lakes and river systems are features that form beneath the bulk of a glacier, in the subglacial zone, that can become dammed beneath the glacier. Subglacial dams, or even minor melting events, can result in a violent release of water and cause major downstream destruction. These events are known as jökulhlaups, where a melting event causes a mass expulsion of water. In Iceland, it is not uncommon for jökulhlaups to occur in response to a sub-glacial volcanic eruption.

Various biological and geochemical processes stimulate feature development in glaciers. There are small-scale, supraglacial features called cryoconite holes. Cryoconite, the amalgamation of sediment and debris within a surface depression, and cryoconite holes form from biological processes. Cryoconite holes form in the area of the glacier that is actively losing material, otherwise known as ablation zone of glaciers (Wharton Jr. et al. 1985). Cryoconite holes form when windblown sediment and debris are caught and accumulate on the surface of a glacier or ice sheet. The concentration of dark material promotes the melting of the underlying ice by reducing the surface albedo, absorbing more solar energy, and forms a depression (Wharton Jr. et al. 1985). The cryoconite holes develop and deepen by the continued absorption of solar radiation. The water also transports nutrients and bacteria. Cryoconite holes provide a habitable zone of growth for various microorganisms (Cook et al. 2015). There are extremophiles, microbes that live in extreme environments, which live on the surface of glaciers and impact surface morphology and glacier melting rates. Microorganisms produce a sticky substance, which is why sediment and debris get caught on the surface of a glacier and form cryoconite holes. Also, these extremophiles are recognized to utilize atmospheric CO_2 as a carbon source, while using light as an energy source (Stibal et al. 2012). The bacteria store the carbon within self-generated, cohesive extracellular polymeric substances (EPS), which allows the microorganisms to later utilize it as a carbon source during periods of nutrient shortage (Sheng et al. 2010).

On glaciers, periods of nutrient shortage can occur quite frequently, since the only medium in which nutrient transport can occur is meltwater; a transport mechanism dependent on melt events. An investigation of the direct and indirect relationships supraglacial microorganisms have with their environment is important when investigating inorganic and organic carbon sources and sinks on glaciers and glacier melting rates. In anaerobic environments, the bacteria must take oxygen from any remaining substrates within the nutrient depleted melt water. As a result, downstream anaerobes produce methane, an effective GHG and biogenic source of inorganic carbon (Skidmore et al. 2000; Stibal et al. 2012).

Englacial features, features within a glacier, are more difficult to measure and quantify, because of the difficulty to access them. Various englacial features can be categorized as “pseudokarst.” Pseudokarst landforms are karst-like features in non-typical environments, such as glaciers, but form as a result of similar processes to that of carbonate rock dissolution. Pseudokarst development follows the same general mode of feature formation as in traditional karst environments. As water flows through and under glaciers, it creates various dissolutional features, which include moulins, shafts, cascades, vadose galleries, phreatic channels, siphons, griphons, water filled dolines, and glacier caves (Mavlyudov 2006). Moulins are also a part of the drainage system in glaciers and form as a result of melting (Fountain and Walder 1998). Moulins form a direct conduit from the surface of a glacier to beneath it, allowing water to flows to its base. Once beneath the glacier, water acts a lubricant and allows the glacier to move more easily, resulting in greater subglacial melting (Fountain and Walder 1998).

During melting events, small and large streams will flow on the surface and within glaciers. Melt events typically occur during warm seasons, but water-carved channels also exist during accumulation seasons (Isenko et al. 2005). Channels vary in characteristics based on the thermal category of the glacier: thermal, poly-thermal, or polar. Some of the earliest scientific observations in the mid-19th century found that subglacial streams formed as meltwater penetrates through crevasse features in the glacier and, from there, flow beneath the glacier (Hopkins 1862). The structure of the drainage system of a glacier can help indicate the glacier’s dynamics, inner ablation, and other hydrological processes (Isenko at al. 2004). Once beneath the glacier, the water can accumulate in subglacial lakes; however, the transport of water off a glacier typically occurs through glacial streams and rivers. These conduits remain active throughout the year on temperate and, more commonly, on polar glaciers as global temperature continues to increase.

2.3 Carbon Cycling

As described previously, carbon plays a major role in climate change. Continuous measurements of atmospheric CO_2 at the Mauna Loa Observatory in Hawaii since 1958 have recorded atmospheric CO_2 concentrations over decades (Keeling 1989). Post et al. (1990) explain how the carbon cycle is quantified by measuring three different types of carbon: inorganic carbon, organic carbon, and particulate organic carbon. Inorganic carbon is comprised of carbon within bicarbonate and carbonate ions from weathered rock material, as well as dissolved CO_2; organic carbon is derived from organic molecules (Post et al. 1990; Lloret et al. 2011). Post et al. (1990) describes carbon derived from live organisms and leftover decayed plant and animal matter as particulate organic carbon. Modern populations have contributed to the increasing concentration of atmospheric CO_2 through the combustion of fossil fuels, carbon-based remains of prehistoric organisms, usually in the form of oil or gas. Since 1860, carbon emissions from burning fossil fuels increased at a rate of approximately 4.3% per year until 1973; however, this study excluded global events, such as the Great Depression and the World Wars. The trend ceased in 1973, due to the oil embargo; however, since 1988, the emissions increased again (Post et al. 1990).

The ocean is the main storage reservoir for carbon, although there are two other major storage reservoirs: the atmosphere and terrestrial systems (Post et al. 1990; Schimel 1995; IPCC 2005), which include carbon storage in rock and sediment. The atmosphere still plays a significant role in carbon storage, although it stores the smallest reserve, and primarily acts as a flux between the ocean and terrestrial reservoirs. Glaciers are of interest in global carbon storage, transport, and sequestration. Since the 19th century, global surface air temperature has increased substantially with a warming trend that has been strong for the past three decades (IPCC 2013). The Arctic terrestrial system covers approximately 25% of the Earth's vegetated land surface and contains about one third of the total global terrestrial carbon (McGuire et al. 1995; Zimov et al. 2009).

Studies indicate that during the 1990s, the Arctic region was, on average, a modest CO_2 sink that contributed less than 0.5 Pg C/yr (McGuire et al. 2009), but recent estimates are still being calculated. In contrast, studies indicate a net positive budget of CO_2 for Iceland from measuring volcanic emissions of CO_2, anthropogenic emissions of CO_2, and fixation from chemical weathering and vegetation (Stein and Macdonald 2004; Gíslason 2005); however, this may change as the climate continues to warm and additional carbon is released from GICs and their associated weathering processes.

Determining the carbon flux of rivers is an especially important component of the carbon cycle (Finlay et al. 2006). Rivers are a significant transport mechanism for water from land to oceans and, subsequently, transport carbon as well (Lyons et al. 2013). Although riverine carbon flux is a smaller component in the global carbon cycle, the transport of carbon in the river system is significant, but lacking in studies in Arctic areas (Sarin et al. 2002; Finlay et al. 2006). DIC is an important component of riverine carbon flux (Tamooh et al. 2013) and is the dominant species of carbon within many riverine systems (Meybeck 1993; Jarvie et al. 2017). In Iceland, the meltwater from nearby glaciers is a major source of inorganic carbon; however, the weathering of bedrock is also an important source of DIC, mostly as HCO_3 (Gislason 1990; Sarin et al. 2002; Louvat et al. 2008). The effects of a warming climate on the carbon flux in Arctic environments have not been adequately studied to develop a robust knowledge of the climate warming and carbon cycling feedback effects in those regions (Striegl et al. 2005). Of the nearly 230 rivers in Iceland, the ten largest are glacial fed or influenced (Gudjonsson 1990; Adalsteinsson et al. 2000) and play some role in the carbon budget; see Figure 2.2.

Figure 2.2: Map of Iceland with GICs and rivers (Source: Max Naylor 2007).

Controls for riverine DIC are primarily associated with watershed inputs and in-stream geochemical, biologic processes, and exchange with the atmosphere (Raymond et al. 2013; Brunet et al. 2005; Tamooh et al. 2013). CO_2 exchange from the river with the atmosphere, weathering or dissolution of rock, primary production, and community respiration are all processes that contribute DIC to rivers (McConnaughey et al. 1994;

Amiotte-Suchet et al. 1999; Abril et al. 2005; Finlay and Kendall 2007). The chemical and mechanical erosion of siliciclastic material that occurs within rivers contributes significantly to CO_2 consumption rates (Dessert et al. 2003; Louvat et al. 2008). The average CO_2 consumption rate in Iceland's rivers is 0.74×10^6 mol/km^2/yr, which is higher than the global average for rivers draining siliciclastic rocks (Louvat et al. 2008). The chemical weathering of basalt, a Ca and Mg silicate, results in a significant DIC presence in Icelandic streams (Louvat et al. 2008; Stefánsson et al. 2017) by using CO_2 to generate other forms of DIC, such as bicarbonate (HCO_3) and carbonate (CO_3).

Studies have determined that geochemical weathering reactions are generally independent of bedrock lithology and, instead, dominantly controlled by runoff (White and Blum 1995; Gíslason et al. 1996; Gaillardet et al. 1999; Skidmore et al. 2004). Furthermore, when calcium carbonate exists in quantities as low as 0.03% of the bedrock volume, carbonate weathering will still dominate in a largely non-carbonate environment (White et al. 1999). This is especially relevant to a region like Iceland, which is dominated by siliciclastic bedrock rich in carbonate minerals, Ca and Mg. The presence of Iceland spar, a well-known, transparent variety of pure calcite, also contributes to the complexity between carbonate and silicate weathering relationships in Iceland. In Iceland, approximately 2,000 m^3/s of glacial meltwater-fed, or influenced, rivers contributes to about a third of the total runoff (Aðalsteinsson et al. 2000; Björnsson and Pálsson 2008). Non-glacial rivers are classified as direct run-off or spring-fed rivers (Kjartansson 1945; Gudjonsson 1990). The discharge of direct runoff streams is variable and largely controlled by daily fluctuation in precipitation, with the greatest peaks in discharge occurring in spring and fall (Jónsdóttir and Uvo 2009); however, with a warming climate there are observable shifts in historical peak run-off from summer and autumn to winter and spring (Barnett et al. 2005).

Dessert et al. (2003) suggest an Arrhenius-type relationship between temperature and CO_2 consumption from silicate weathering, based off observations from research indicating elevated weathering rates with increasing temperature (White and Blum 1995); however, results from these studies are in conflict regarding the importance of temperature in chemical weathering, suggesting run off and denudation may be more dominant (Gaillardet et al. 1999; Dessert et al. 2003). The presence of volcanic rocks and geothermal activity also provides insight into Iceland's above-average chemical weathering rates. Estimates from 2003 indicate the weathering of volcanic rocks is responsible for 30% of global CO_2 consumption (Dessert et al. 2003). Previous studies of riverine carbon flux in volcanically active regions indicate surface weathering, atmospheric inputs and high temperature water-rock interactions to be the primary sources of carbon in those regimes (Dessert et al. 2003).

A temperate climate, high precipitation and runoff, volcanic and siliciclastic lithology, and drastic relief contribute to the elevated chemical weathering rates which occur in the rivers of Iceland; therefore, it is important to determine the carbon flux from rivers in high runoff islands, such as Iceland, which could contribute a large quantity of dissolved carbon to the ocean (Louvat et al. 2008). A 2001 study by Stefánsson and Gíslason determined that all dissolved Si, Fe, and Al in sample waters from their study site in southwest Iceland were sourced from the weathering of rock. The study also determined that increased weathering results in higher Na concentrations since the secondary mineralization of zeolites have not begun to form (Stefánsson and Gíslason

2001); this suggests low Na concentrations are a result of negligent water-rock interactions or secondary phases of mineralization have occurred and consumed the Na in the solution.

2.3.1 Hydrothermal Influences

Cold water in Iceland can be geochemically distinguished from geothermal water by observing lower chlorine concentrations, pH ranges of 6-7, comparable Na:K ratios with that of the associated rock, and undersaturation with respect to calcite (Arnórsson 1983). Generally, groundwater in Iceland is relatively pure and low in dissolved ions, thus making it distinct from hydrothermally influence groundwater inputs. Studies have shown that rock weathering does not significantly contribute to the concentration of chloride in surface or ground water; therefore, increased concentrations of chloride is an indicator of geothermal influence, and potentially marine influence (Sigurðsson and Einarsson 1988; Arnórsson and Andrésdóttir 1995). Silica and sulfate ions have also been determined to be geochemical indicators of geothermally-influenced groundwaters (Sigurðsson and Einarsson 1988).

2.4 Carbon Isotopes

Isotope geochemistry can be used to identify and isolate the different sources of carbon in the environment. Isotopes are elements with variable number of neutrons. Isotopes have both stable and unstable species. Stable isotopes are studied in climate studies because they do not undergo radioactive decay, but experience fractionation, the basis for modern analyses (Urey 1947; Bigeleisen and Mayer 1947). The differences in mass give the isotopes different physical and chemical properties, which can be measured through various natural processes dependent upon fractionation acting upon the isotopes.

Stable isotope analysis measures the ratio between the heaviest isotope to the lightest isotope. This ratio is expressed as δ, which is then compared to known standards for reference. The accepted standard reference for carbon is the Vienna Pee Dee Belemnite (VPDB). Since stable isotope ratios are typically small values, the ratio is multiplied by 1000 to better express concentration differences and expressed as per mil notation (‰). Equation 1 expresses the ratio as (Clark and Fritz 1995):

$$\delta^{13}C_{sample} = \frac{m(^{13}C/^{12}C)_{sample}}{m(^{13}C/^{12}C)_{reference}} x\ 1000\ VPDB \qquad \text{(Equation 1)}$$

Isotope fractionation describes the tendency of isotopes to separate during naturally occurring processes, such as evaporation or melting, as the result of the mass differences. The two most common processes that result in isotope fractionation are equilibrium and kinetic (non-equilibrium) (Clark and Fritz 1995). In equilibrium isotope-exchange reactions, the heavier isotope becomes enriched (Kendall and Caldwell 1998). In equilibrium, reactions rates are the same in the forward and reverse direction and the isotope ratios stay constant; however, the isotopic concentrations are not identical. Due to these properties, the heavier isotope preferentially accumulates in the compound with the higher state of energy (Clark and Fritz 1995; Kendall and Caldwell 1998). In glacial ice, the lighter fractions of carbon are represented, as they preferentially accumulate in water during air-gas exchange processes.

In kinetic, or non-equilibrium, isotopic fractionations the forward and backward reaction rates are not identical. Kinetic fractionation is more heavily influenced by ratios of the masses of isotopes and the vibrational energies (Clark and Fritz 1995; Kendall and Caldwell 1998). Generally, the bonds within lighter isotopes tend to break more easily than the bonds in heavier isotopes. Lighter isotopes react more readily and thus exist in higher concentrations in the reaction's product, while the opposite is true for heavier isotopes. Photosynthesis and other biological processes favor this form of fractionation.

Stable isotope values provide source data for DIC within water samples. Substances can be characterized by different relative isotropic abundances of various elements, which provide an isotopic signature (Muccio and Jackson 2009). Source determination is possible, because of enrichment or depletion of elements in response to kinetic and thermodynamic factors, such as changes in climate and volcanic eruptions.

Stable isotope analysis studies of $\delta^{13}C_{DIC}$ in groundwater, surface water, and geothermal water have been conducted in Iceland with similar results: $^{13}C_{DIC}$ ranges are large and are variable (Hilton et al. 1998; Sveinbjörnsdóttir et al. 2005; Stefánsson et al. 2017). Major sources in higher temperature groundwater, specifically, have been identified as DIC derived from the dissolution of basalt, atmospheric CO_2, and mantle CO_2 (Stefánsson et al. 2016). Sources of DIC in lower temperature waters are similar, consisting of water-rock interaction with basalt, atmospheric CO_2, and sometimes marine groundwater (Sveinbjörnsdóttir et al. 1995). Surface waters in Iceland have similar isotopic compositions to cold, shallow subsurface waters; however, surface vegetation cover has a measurable influence over $\delta^{13}C$ in surface streams, unlike sub-surface groundwater (Sveinbjörnsdóttir et al. 2010). Low temperature waters extracted from shallow wells have been associated with lesser reactions from water-rock interactions (Sveinbjörnsdóttir et al. 2010). The waters in Icelandic rivers, characterized by high turbulence and mixing, are expected to have $\delta^{13}C_{DIC}$ in equilibrium with atmospheric CO_2, due to increased gas exchange (Lyons et al. 2013).

CHAPTER THREE: STUDY AREA

3.1 Introduction

Iceland is a volcanically active island country approximately 103,000 km^2 in area and located in the North Atlantic Ocean along the edge of the Arctic Circle (Figure 3.1),. This location qualifies Iceland’s climate as subarctic in the southern coastal regions, but tundra inland and in the north. The coordinates also put Iceland in the path of the North Atlantic Current and Gulf Stream, which brings warm, temperate waters northward from the Gulf of Mexico. The Irminger current pulls the warm waters to the south and west coasts of Iceland while cold waters from the east of Greenland are drawn toward the western coast (Figure 3.1).

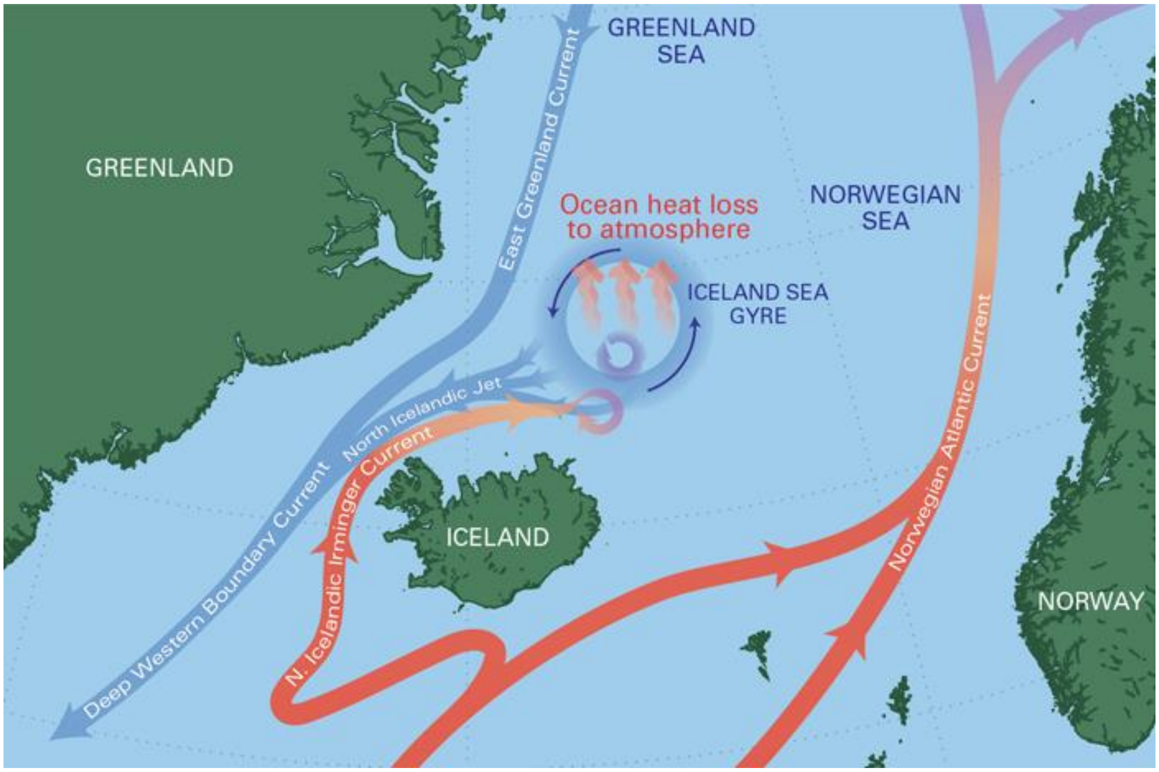


Figure 3.1: Illustration of the various currents impacting Iceland, not to scale (Source: Woods Hole Oceanographic Institute 2012).

3.1.1 Cultural Setting

The economy of Iceland is dependent on factors that are controlled by regional climate, geologic, and glacial processes. The geographic location of Iceland and climatic influences make the region a difficult zone for natural vegetation to grow. The turbid, cold water in the glacial rivers are uninhabitable to traditional river cyanobacteria and plant life, though extremophiles are not uncommon in the more extreme habitats across Iceland, such as geothermal features and glacier surfaces. Birch trees were the only trees native in Iceland and not suitable for timber production; however, the timer was overharvested for use as firewood in the first centuries of settlement. Overharvesting, in combination with the winds in the highlands, which rapidly removes soil, contribute to the ongoing and wide scale prevalence of soil erosion on the island (Blöndal 1993). Historically, the land was cleared to provide grazing land for sheep farming. Grazing practices in Iceland have been determined to have severe impacts on erosion and run off rates (Blöndal 1993; Arnalds 2003; Boardman et al. 2003). The modern expanse of the Icelandic landscape is mostly characterized by low-lying shrubs, moss, and grass.

3.1.2 Geologic Setting

Iceland is situated along the Mid-Atlantic Ridge, a divergent plate boundary that trends roughly N-S along the middle of the Atlantic Ocean. The feature is an active spreading center that generates hot, crustal material as the Eurasian and North American tectonic plates pull apart from each other (Einarsson 2008). Unlike other areas along the ridge, Iceland sits atop what is called the Iceland Plume, a mantle hotspot. Hotspots are considered to form from either the movement of buoyant magma through thin areas in the crust or from the presence of anomalously hot magma. In either situation, the magma rapidly cools upon contact with cold ocean water and accumulates. The sustained accumulation of basaltic material through continuous volcanic eruptions resulted the origin and build-up of Iceland. Today, Ca-Mg rich basalt still dominates the bedrock; however, rhyolite, another igneous rock, and Holocene-aged sediments also characterize the landscape. The lithology, along with the high precipitation and drastic relief, results in an environment extremely sensitive to chemical weathering, and, thus, may contribute to a long-term CO_2 sink within the global carbon cycle (Stefánsson and Gíslason 2001).

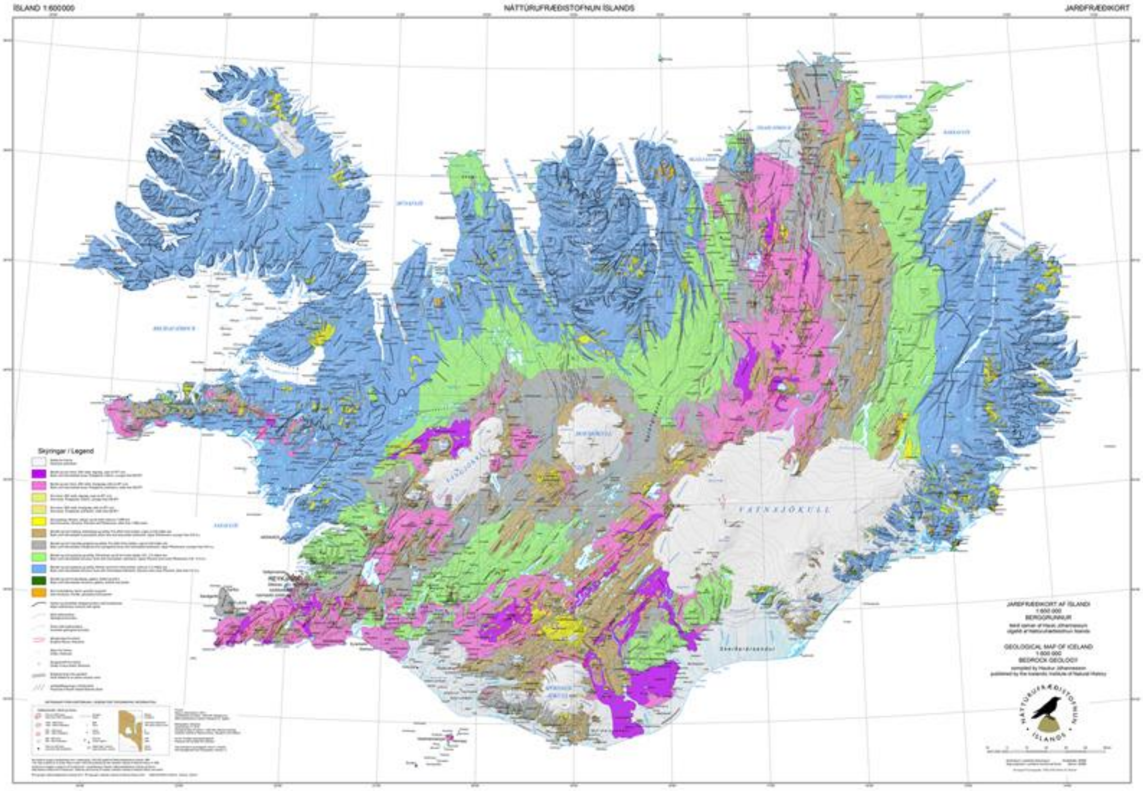


Figure 3.2: Geologic map of Iceland (Source: Haukur Jóhannesson 2014).

The glaciers and rivers of interest in this study are located in the Eastern Volcanic Zone (EVZ), where the landscape is dominated by volcanic fissures and long hyaloclastite ridges (Einarsson 2008). Hyaloclastites, a pyroclastic breccia with glassy clasts, are the product of a basaltic eruption under ice, or cold water, which is evidence not only of past eruptions, but also the glacial extent at the time of the hyaloclastite deposition. While hyaloclastites may have similar mineralogical compositions to basalts generated from the same magma, they differ in deposition and rate of weathering. In laboratory simulations, hyaloclastite dissolution enhances the fluxes of Na, Si, Ca, F, and S at constant runoff, vegetation cover, and basaltic glass content (Gíslason and Eugster 1987). In addition, hyaloclastite dissolves faster than crystalline basalt and depending on runoff will experience elevated elemental fluxes (Gíslason and Eugster 1987).

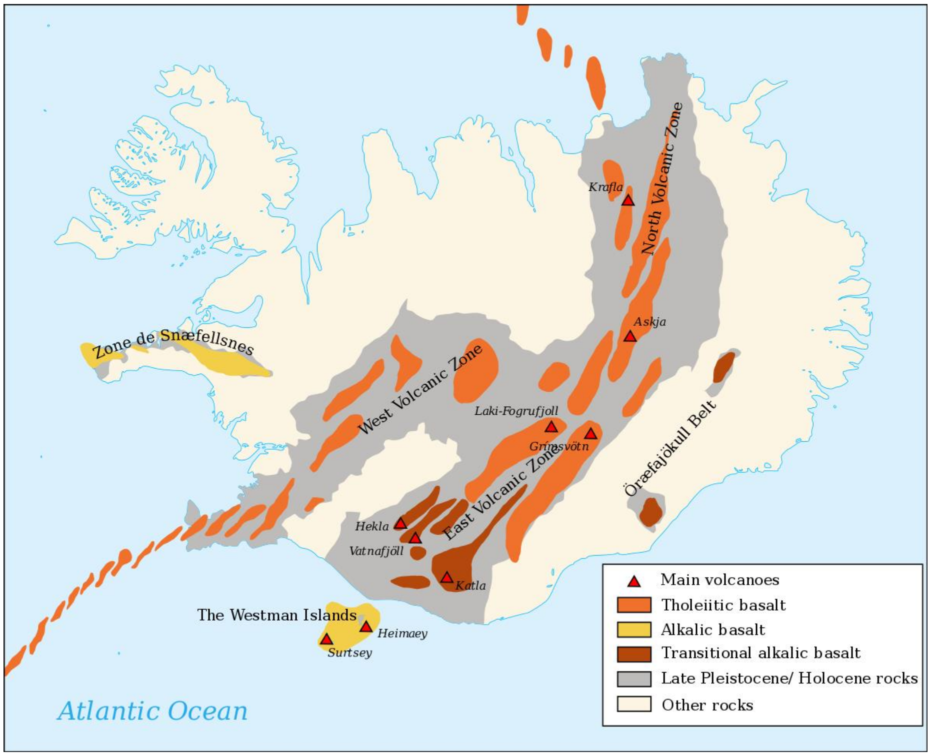


Figure 3.3: Volcanic zones of Iceland (Source: Chandra X-Ray Observatory 2012).

A geologic map is provided in Figure 3.2 that also illustrates the various volcanic zones. Hyaloclastite ridges are topographic features and are found frequently throughout the Icelandic landscape. Hyaloclastites have been determined to dissolve faster than basalt, due to the dominant presence of volatile glass within the rock's matrix (Stefánsson and Gíslason 2001). Associated with the volcanic zones are proximal areas characterized by geothermal activity. Regions of geothermal activity in Iceland have been classified into low-temperature (LT) and high-temperature (HT) zones based on geological context and temperature data from drill hole studies (Bödvarsson 1961). The areas of interest in this study are within proximity to the Eastern Rift Zone (ERZ). A previous study at Sólheimajökull presented data that suggest the hydrogeochemistry of the river is measurably influenced by accessory hydrothermal calcites and geothermal proton supply (Burns 2016).

Tuyas are flat topped, steep sided mountains composed of different types of volcanic deposits often found in glaciated volcanic areas. They are formed primarily from subglacial, single-vent volcanic eruptions, where the lava solidifies quickly after melting through a portion of the glacier. When the remaining volume of ice has melted, the tuya formation is exposed. They form in a similar fashion to hyaloclastite ridges but are distinguished from them topographically, due to the difference in shapes.

3.1.3 Climatic Setting

The influence of the Gulf Stream on Iceland’s southern coast creates a temperate climate that results in the formation and development of “wet,” or “temperate,” glaciers. In Iceland, 60% of the glaciers and ice caps are suspected to be sitting atop active volcanoes (Björnsson and Pálsson 2008), which also enhances melting and weathering from subglacial heating and geothermal fluids flowing from below.

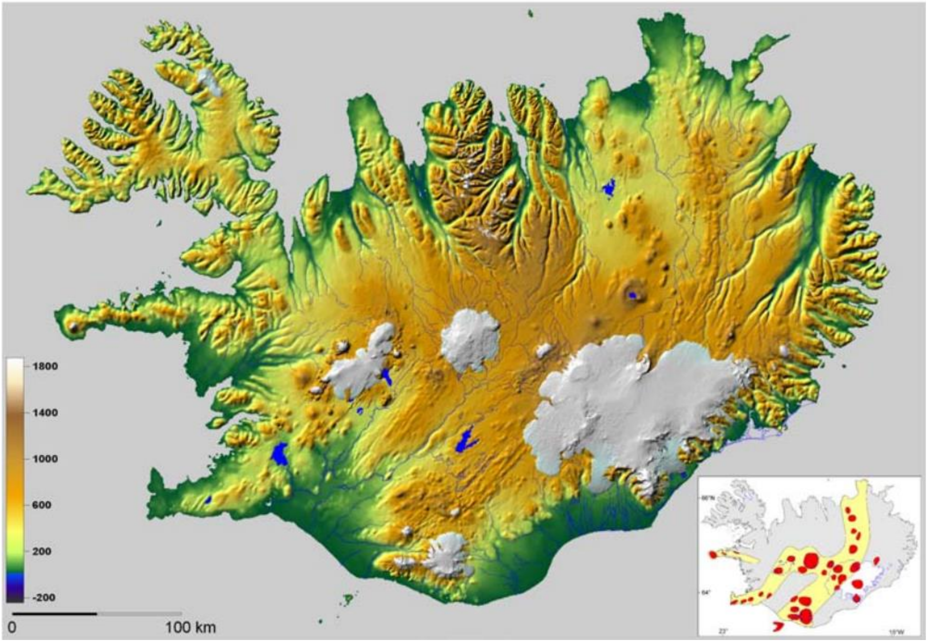


Figure 3.4: Glacial cover and elevation in Iceland (Source: Björnsson and Pálsson 2008).

Precipitation models indicate that most precipitation falls in the southern region of Iceland (Figure 3.3). Approximately 20% of the precipitation that falls is deposited on the glaciers and ice caps in Iceland (Jóhannesson et al. 2006). Piedmont glaciers and mountain glaciers exist in Iceland and catch and store precipitation. Piedmont glaciers are lobe-like glaciers that form when steep valley glaciers spill over the rock confinement into an adjacent, flatter area. Mountain glaciers form in valleys at higher elevations with higher relief. These types of glaciers are known to respond relatively rapidly to regional climate changes, less than the timeframe of decades (Sigurðsson and Jónsson 2006). Icelandic glaciers especially, due to geographic and climatic factors, respond to climate change rapidly. Furthermore, glaciers significantly affect the quantity, variability, and timing of streamflow (Jóhannesson 2006). Runoff from Iceland’s glaciers is expected to peak within the next 50 years and be followed by a decreasing trend, due to reduced volume of glacial ice (Björnsson and Pálsson 2008).

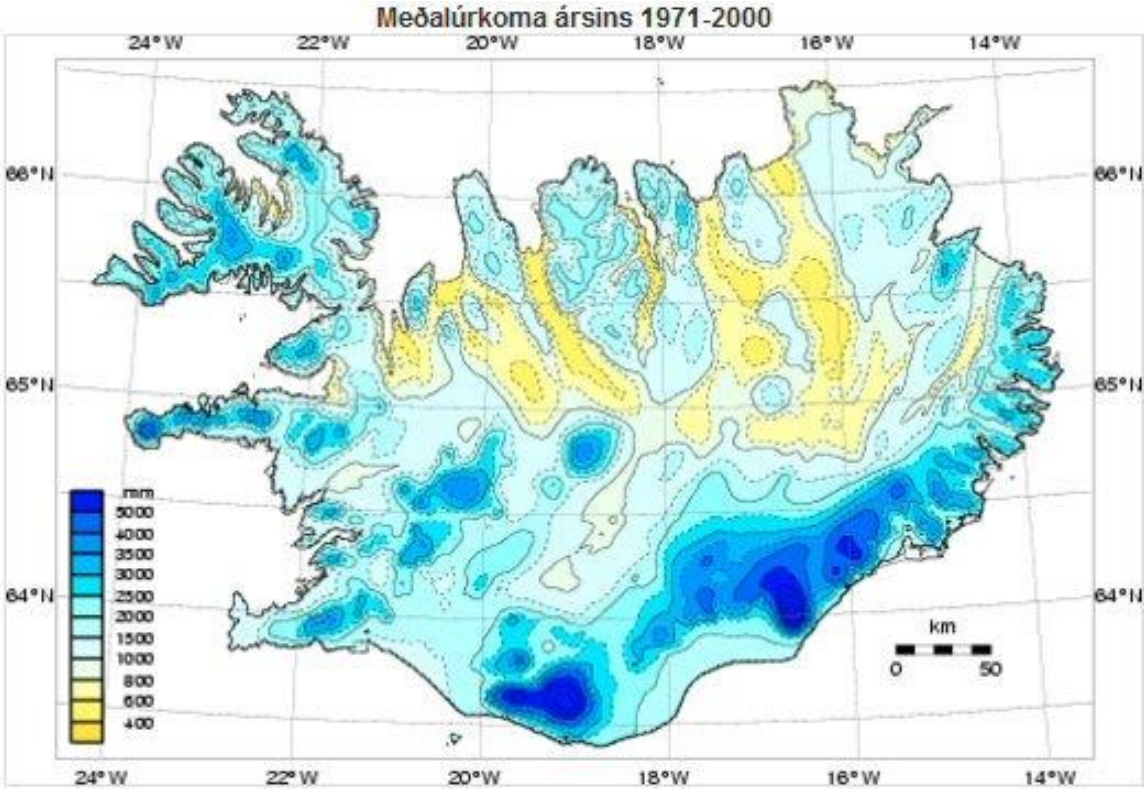


Figure 3.5: Precipitation map of Iceland (Source: Crochet et al. 2007).

GHGs trap warm air within the atmosphere and are subsequently responsible for the progressive and accelerated melting of glaciers from increasing temperatures. There is concern with the recent, unprecedented atmospheric CO_2 concentrations, because CO_2 is a particularly harmful greenhouse gas. Meltwater transports various substances and materials from on top, within, and beneath the glacier.

3.2 Study Sites

3.2.1 Gígjökull

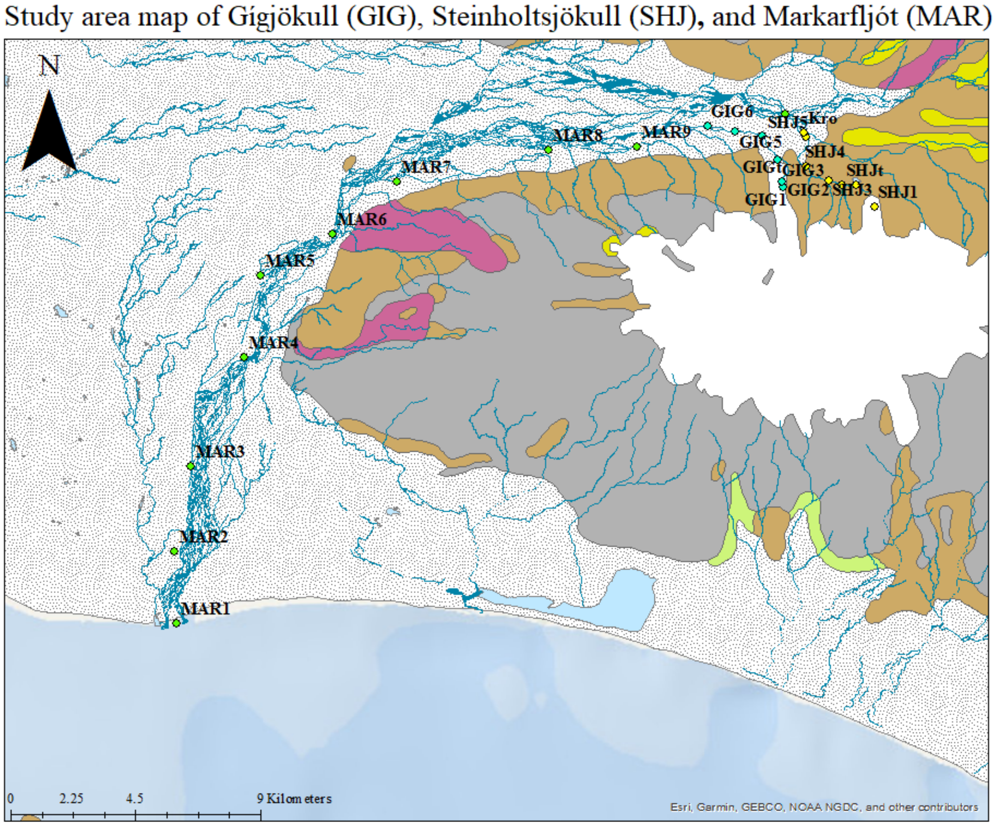
Gígjökull is a retreating glacier that originates at approximately 1,600 m above sea level on the Eyjafjallajökull ice cap, which sits atop the Eyjafjallajökull stratovolcano. The ice cap has an area of 81 km^2 its outlet glaciers are rapidly retreating. A contributing factor of the rapid retreat is the presence and frequency of volcanic activity and the associated geothermal heat beneath the ice. Unlike the other glacier sites visited during this study, Gígjökull currently lacks a pro-glacial lake. The 2010 eruption of Eyjafjallajökull resulted in the destruction of an established, moraine dammed pro-glacial lake and subsequent changes in the proximal geomorphology.

The drainage river of Gígjökull, Gígjökulsá, eventually feeds into a much larger river, Markarfljót. Markarfljót is a large river that drains both the Eyjafjallajökull and Mýrdalsjökull ice caps. Its origin is east of the volcano, Hekla, and it continues southward where the landscape eventually flattens. The river becomes extremely braided where the velocity drops significantly, until it flows into the ocean.

3.2.2 Steinsholtsjökull

Steinsholtsjökull is another outlet glacier of the Eyjafjallajökull ice cap and is located slightly east of Gígjökull. Unlike Gígjökull, Steinsholtsjökull is located several kilometers back into a narrow canyon carved from its own retreat and has little tourist visitation. The glacier is difficult to get to and has no trails, roads, or vehicle access, which is the likely reason for very little tourist visitation. Steinsholtsjökull has a pro-glacial lake present below and past the glacier's terminus. Moraines line the edges of the canyon and have formed into tall, steep features that assist in channelizing and directing the hydrology. The glacier has many active ice falls along the edge of the mountain; however, the glacier is still mostly intact despite the icefalls, unlike Gígjökull. The most recent event to alter the landscape and dynamics at Steinsholtsjökull was a massive landslide that occurred in 1967, which extremely altered the terminus of the glacier and the portion of the glacier valley (Dugmore 1989), and subsequently resulted in a major jökulhlaup.

Steinsholtsjökull’s river, Steinholtsá, drains into the river Krossá, which is a drainage river of Mýrdalsjökull and runs through Þórsmörk, a popular tourist and hiker destination. Krossá quickly empties into Markarfljót, slightly upstream of the Gígjökull confluence. The sampling locations of both Gígjökull and Steinsholtsjökull are indicated in Figure 3.4.



Study area map of Gígjökull (GIG), Steinholtsjökull (SHJ), and Markarfljót (MAR)

Figure 3.6: Sampling locations along the drainage rivers of Gígjökull (GIG) and Steinsholtsjökull (SHJ) as well as the major river, Markarfljót (MAR), in which both glaciers ultimately converge (Source: created by Author).

3.2.3 Sólheimajökull

Sólheimajökull is an outlet glacier of Mýrdalsjökull, an ice cap in southern Iceland. Mýrdalsjökull covers the Katla volcano, the sister volcano to Eyjafjallajökull. Both volcanoes are relatively active and historically have a tendency to erupt concurrently (Sturkell et al. 2003). Katla has erupted 21 times since 870 AD (Óladóttir et al. 2005), with its most recent eruption in 1918. In comparison, Eyjafjallajökull's most recent eruption was in April of 2010 and had major global impact. Sólheimajökull comprises over half the area of the entire Eyjafjallajökull ice cap at 44 km^2. The glacier is 11 km long and drains the southwestern part of the ice cap (Schomacker et al. 2012). Like the two previously mentioned study glaciers, Sólheimajökull is currently in a state of retreat. The main river draining Sólheimajökull, Jökulsá á Sólheimasandi, was sampled from the terminus of the glacier to the ocean, approximately 10 km to south.

Study area map of Sólheimajökull (SOL, SOLA, SOLt)

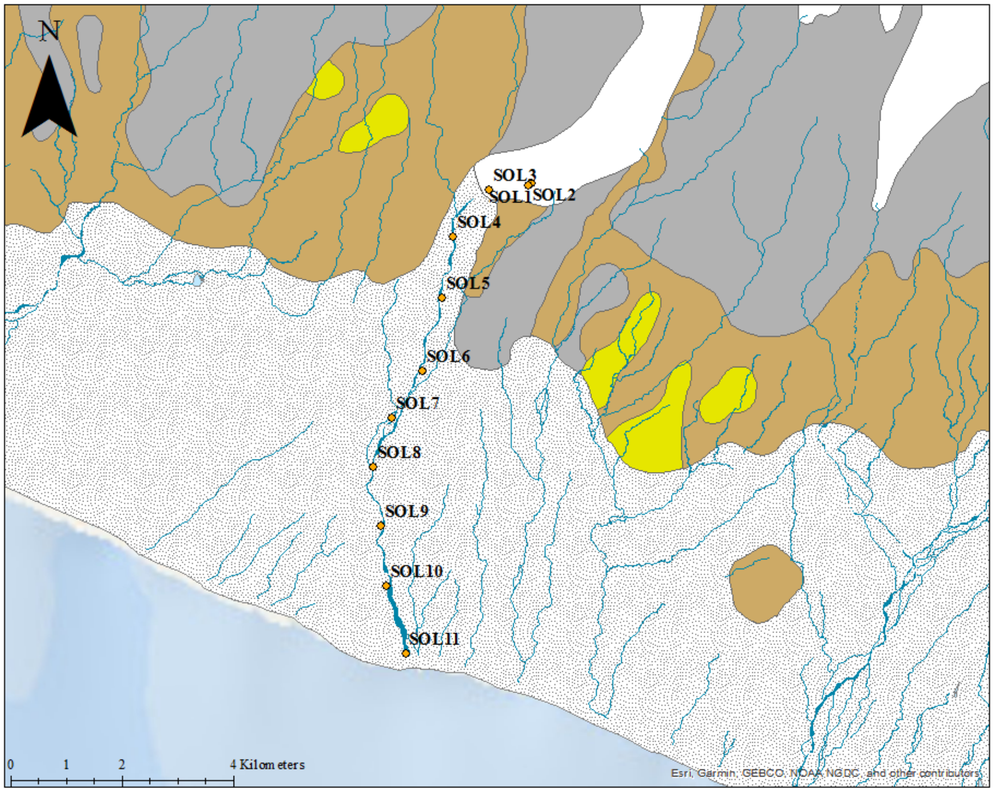


Figure 3.7: Sampling locations along Sólheimajökull's drainage river Not shown are the sample identifiers for the second sampling session along this river (SOLA), as they are identical to the locations of SOL and SOLt, which was at the same location as SOL2 (Source: created by Author).

A recent study identified three sources of waters contributing to Jökulsá á Sólheimasandi: supraglacial runoff, sub-glacial waters, and tributaries contributing external mixed waters (Burns 2016). Unfortunately, due to inaccessibility, neither of the tributaries, Jökulsárgil and Fjallgilsá, could be sampled. Both tributaries enter the river from the western side and contribute measurable volumes of water; however, the input of these tributaries was identifiable in the data.

3.2.4 Falljökull

Falljökull is an outlet glacier of Vatnajökull, which is the largest glacier in Iceland and Europe with a previously established area of 8,100 km^2; Vatnajökull dominates the landscape in the eastern region of Iceland. Beneath the ice, which peaks at 2,109 m, are several volcanoes. Among the most well-known and active are Bárðarbunga, an ice-filled and ice-covered caldera, and Grímsvötn. Grímsvötn and Bárðarbunga are extensively monitored by various agencies within Iceland due to their frequent volcanic activity. Both volcanoes are a part of active volcanic systems and experience multiple earthquakes every month. As typical for many volcanoes that are covered by ice, there is a high risk of jökulhlaups occurring during or after eruptions. Longitudinal sampling plans could not be applied to this study location, due to the relatively short distance from source to ocean and inaccessibility of some upstream and downstream sites.

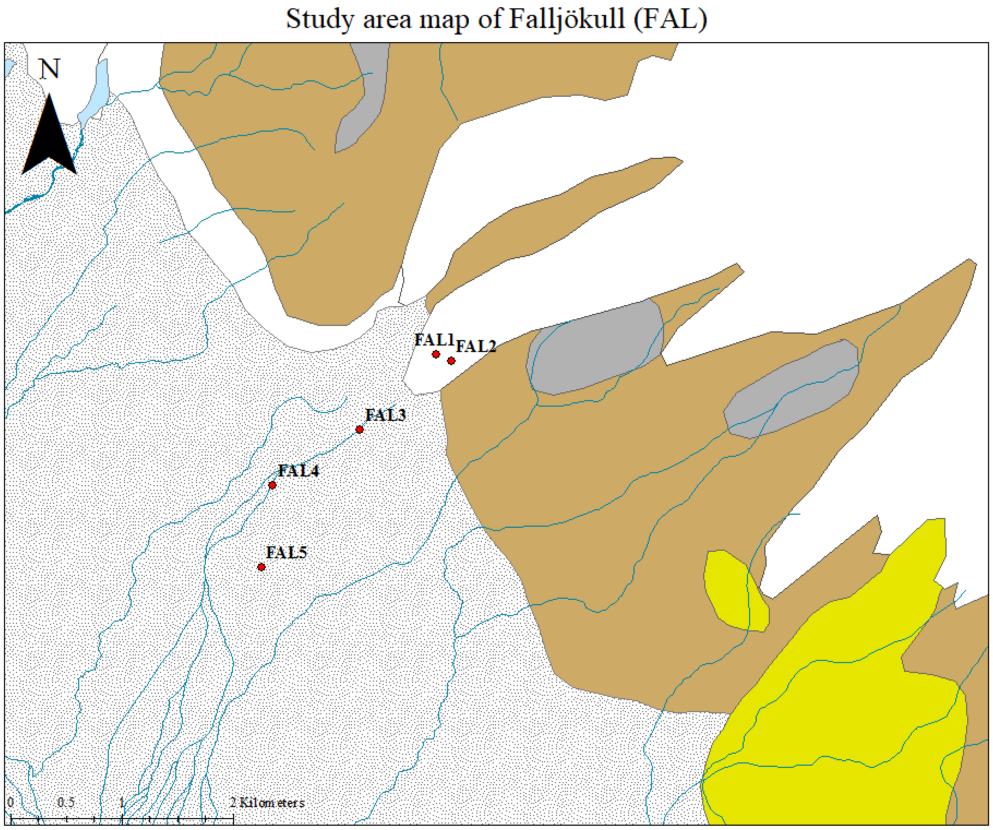


Figure 3.8: Sampling locations along Falljökull's drainage river (Source: created by Author).

CHAPTER FOUR: METHODS

4.1 Field Methods

4.1.1 General Sampling Description

Analysis and grab water samples were collected from various sites along the primary drainage rivers and where appropriate, glacial surface streams and pro-glacial lakes, from Gígjökull, Steinsholtsjökull, Sólheimajökull, and Falljökull. The large river Markarfljót was sampled independently, without a source sample, to serve as an end member for both Gígjökull and Steinsholtsjökull, since both rivers converge with Markarfljót before reaching the ocean. Where access allowed, samples were collected longitudinally to quantify the hydrochemical alteration and carbon evolution of the water from source to sink. All samples were given a unique sample identifier in the format of SiteID_Sample number, the analysis, and site location. Field notes were taken to document field observations, weather conditions, and sample collection notes. Sampling occurred during the 2017 field season from June 6th to June 11th to capture the meltwater increase during the spring to summer transition.

Sampling began at various glaciers along the Eyjafjallajökull ice cap, specifically from the two named outlet glaciers Gígjökull and Steinsholtsjökull. The main drainage river of Gígjökull was sampled approximately every kilometer from the outlet of the river beneath the glacier to the confluence with Markarfljót, which totaled seven samples with the addition of a tributary sample. An example of the sampling process at the source of the river is included in Figure 4.1. The drainage river of Steinsholtsjökull was sampled approximately each kilometer from the glacier to the confluence with Krossá and once from one of the tributaries for a total of seven samples. The sampling distance of one kilometer was extended for Markarfljót, due to the suspected equilibrium and stability of in-stream processes and distance to cover versus time available. Markarfljót was sampled approximately every 2.5 km, with a total of 10 samples, from the river's mouth into the ocean upstream to its confluence with Krossá.

Figure 4.1: An image from Gígjökull showing an example of the surface stream sampled (red) and sub-glacial output stream (yellow), with the 2010 eruption material circled in

green. At most glaciers, the first sample of the set was often extremely close to the second sample, in order the capture the geochemical characteristics of the water as soon as it exited beneath the glacier. Scale of yellow circle is three meters (Source: Author).

Sólheimajökull was sampled 11 times, approximately each one km, from the glacier's terminus to the river's outlet into the ocean. On a separate day, two types of sampling occurred: reverse sampling from ocean to glacier and hourly sampling on the main glacial stream outlet from the glacier before the stream's entrance to Sólheimajökull's pro-glacial lake. The final portion of sampling occurred at Falljökull, an outlet glacier of the Vatnajökull ice cap.

Rock samples were gathered from bedrock outcrops near the glacier's outflow and sampling locations. Hand-sized samples of approximately 150 g were collected and stored in airtight, re-sealable bags. Upon return to WKU, rock samples were powdered using a handheld Dremel using a coarse, abrasive head. Samples were shipped to the SIRFER lab at the University of Utah in approximately 50 mg-sized aliquots for carbon isotope analysis to establish a baseline signature for the subglacial rocks and subsequent chemical weathering. Samples were analyzed using Cavity Ring Down Spectroscopy (CRDS), using a Picarro CRDS, which measures a molecule's unique, near-infrared absorption spectrum.

4.1.2 Grab Samples

Identical sampling procedures were applied at all sample locations. Grab water samples were collected at each site for hydrochemical parameters using a YSI ProDSS multiparameter meter: temperature, pH, specific conductivity (SpC), dissolved oxygen (DO), nitrate (NO_3), total dissolved solids (TDS), and pressure. A Hach DR-900 colorimeter was used for measuring turbidity and total suspended solids (TSS) of grab samples in the field.

4.1.3 Analyzed Samples

Water samples were collected and filtered using 0.45 µm nitrocellulose filters for alkalinity, which was analyzed upon completion of the field day using a field-appropriate version of the Gran titration method. Additional water samples were collected and filtered for anion, cation, and stable carbon isotope analysis (Table 4.1). Cation samples were pre-preserved with sulfuric acid to ensure a final sample pH of less than two and reduce the occurrence of post-sampling alteration. In addition, small bedrock samples, and glacial stream samples were collected for stable carbon isotope analysis. All samples kept in coolers during the sampling portion of the field day, but transferred to refrigerators upon completion of the sampling.

Table 4.1. Parameters and analytes for sampling (Source: created by Author).

Test	Equipment/Sample Bottle Size	Analytical Method
Temp.	Pro-DSS	Grab
pH	Pro-DSS	Grab
Nitrate	Pro-DSS	Grab
SpC	Pro-DSS	Grab
DO	Pro-DSS	Grab
Pressure	Pro-DSS	Grab
TDS	Pro-DSS	Grab
Turbidity	DR-900	Grab
TSS	DR-900	Grab
Alkalinity	125 mL	Field Titration
Anion	60 mL	Ion Chromatograph
Cation	60 mL	Inductively Coupled Plasma Mass Spectrometry (ICP-AES)
Carbon Stable Isotope	20 mL	SIRFER Lab

4.2 Secondary Data Collection

4.2.1 Discharge

Discharge was taken on the glacial rivers of Gígjökull, Steinsholtsjökull, and Sólheimajökull from a moving boat with an attached acoustic Doppler current profiler (ADCP). Three measurements were averaged from each river to determine a final discharge measurement for the river. ADCPs are commonly used to take streamflow measurements in water as shallow as a 0.3 m deep (Mueller and Wagner 2009). The deepest river was measured at just under one meter at Sólheimajökull with the shallowest depth measured from Gígjökull at 0.38 m. An ADCP was chosen for discharge determination primarily for its availability from the Icelandic Meteorological Office (IMO), but also for the unique characteristics of Icelandic rivers that can make alternative methods of discharge determination difficult. The ADCP is an ideal method for measuring discharge or stream velocity in streams characterized by unsteady, bidirectional, and nonstandard flow (Mueller and Wagner 2009).

Discharge was measured in a single setting along the three of the studied rivers at various distances from sources. The discharge measurement for Steinsholtsjökull was taken the day before sampling occurred and two days before sampling occurred at Sólheimajökull. These data were included in the study, due to the absence of precipitation and comparable air temperatures, or changes in other parameters, which could affect downstream discharge. Though not ideal, these discharge values are representative of base flow in the studied rivers and, therefore, allow for a conservative discussion based on the results from other data.

4.2.2 Watershed Information

Watershed data, image files, and watershed size, were provided by the IMO for use in flux determination and conservative total flux estimates. Watershed data are indicated in Table 4.2. Images of the watersheds are included in Figures 4.2, 4.3, and 4.4.

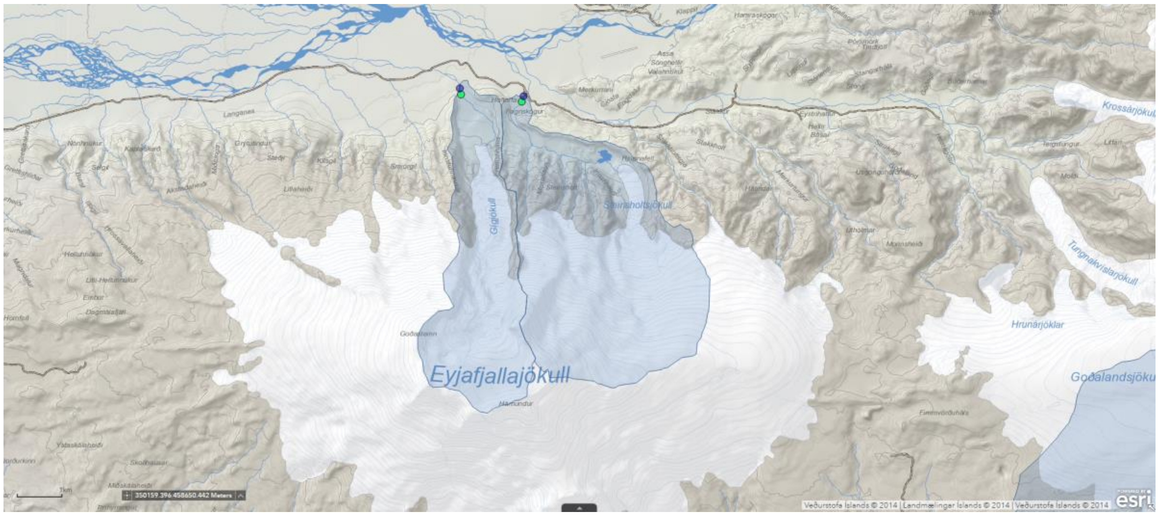


Figure 4.2: Watershed for Gígjökull (left) and Steinsholtsjökull (right) (Source: IMO).

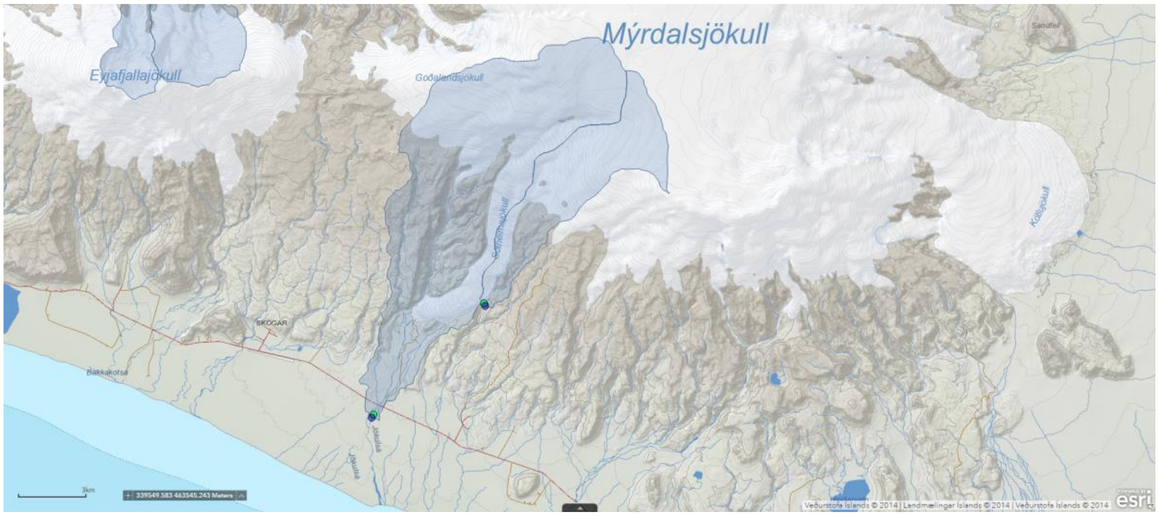


Figure 4.3: Watershed for Sólheimajökull (Source: IMO).

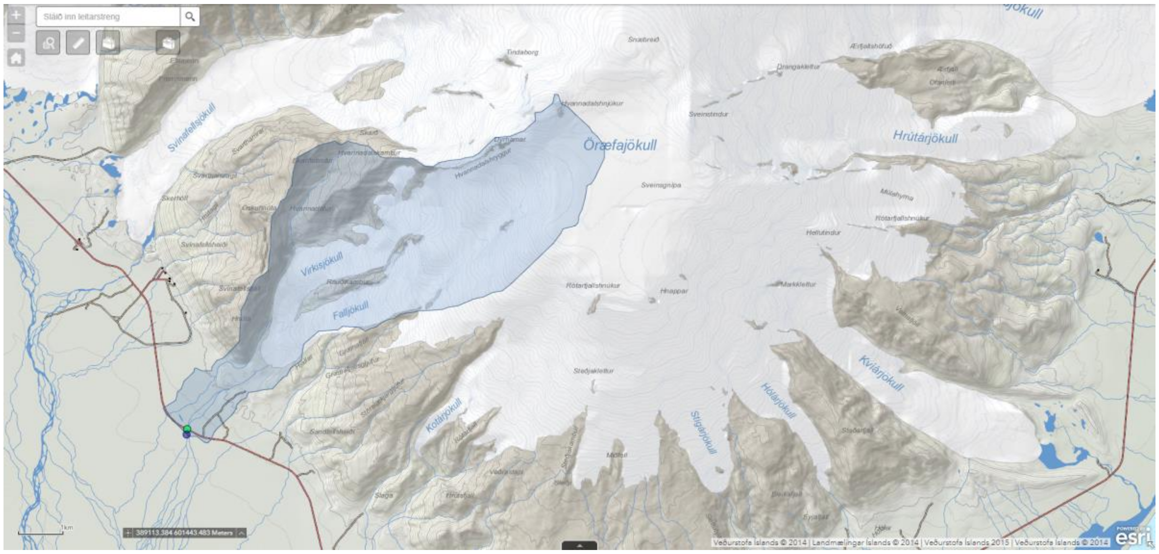


Figure 4.4: Watershed for the Falljökull study sites (Source: IMO).

Table 4.2. Watershed size by site (Source: created by Author).

Watershed	Size (km^2)
Gígjökull	10.69
Steinsholtsjökull	19.32
Sólheimajökull	106.04
Falljökull	28.20

4.2.3 Weather Station

Temperature and precipitation data for the appropriate field areas were obtained from two of IMO's weather stations, Básar á Goðalandi and Skaftafell, for the duration of the sampling week and three weeks prior. Additional watershed data were also provided by the IMO for the sites within the study. The weather data were used with field observations to supplement and support trends observed in geochemical data.

4.4 Determination of Alkalinity

Samples for alkalinity were filtered using a 0.45µm nitrocellulose filter and collected in an un-acidified 250 mL bottle. Titration of all samples occurred after the sampling portion of the field day concluded. Alkalinity is the chemical measurement of a solution's ability neutralize an acid and the sample's buffering capacity, or its ability to resist changes to pH when acids or bases are added. Alkalinity was measured by adding sulfuric acid to 50 mL of sample until a pH of 4.5 was reached. The equation used to calculate total alkalinity is represented as:

$$\frac{(A \cdot N \cdot 50000)}{V} \qquad \text{(Equation 2)}$$

Where *A* is the volume of titrant added, *N* is the normality of the titrant, and *V* is the volume of the water sample. The method used and described above is the Gran acid titration method. The theory behind the Gran acid titration method includes an estimation of an equivalence point between an acid and base through calculations including the sample's change in pH and titrant volume (Gran 1950). The values for pH and titrant volume can be plotted against each other in a Gran plot to demonstrate the titration curve (Figure 4.5). The curve can be used to estimate the end-point, which represents the equivalence point at which enough titrant was added to complete the acid-base reaction of the solution. The determination of the samples' alkalinity was required to calculate the individual DIC species and, thus, DIC itself.

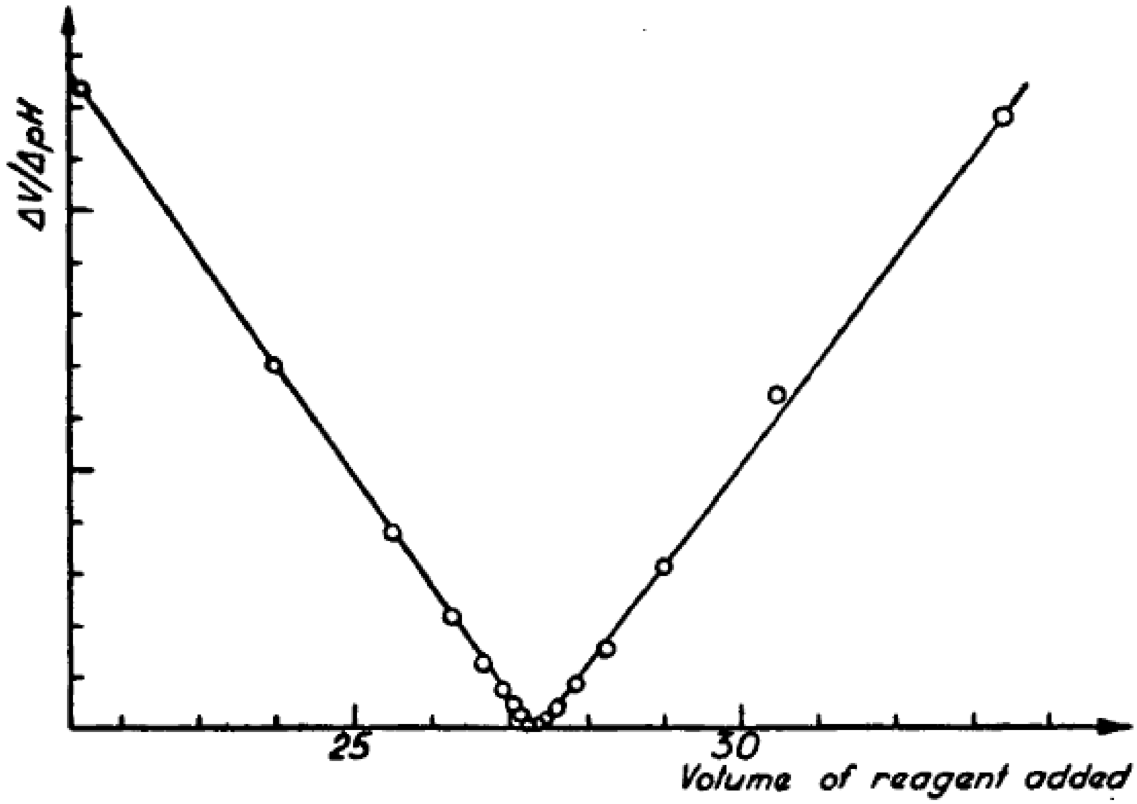


Figure 4.5. Gran plot (Source: Gran 1950).

4.5 Calculation of DIC using the THINCARB Model

DIC, excess partial pressure of CO_2 ($EpCO_2$), HCO_3 concentration, and $\%_{HCO3}$, $\%_{CO3}$, and $\%_{H2CO3}$ of the DIC were calculated using the Thermodynamic Modelling of Inorganic Carbon (THINCARB) method (Jarvie et al. 2017). The model requires user input of altitude, pH, alkalinity (μeq/L), temperature (°C), and Ca concentration (mg/L). THINCARB generates altitude corrected $EpCO_2$, charge balance, equilibrium constants coefficients, ion activities and strengths, HCO_3 concentration, calcite saturation, Total DIC concentration, and DIC speciation of the dataset. $EpCO_2$ is used within the Jarvie et al. (2017) model and this study, because it provides a normalized ratio of dissolved CO_2

in the water with that of the atmosphere and is considered an integral part of environmental-water quality studies (Neal et al. 1998).

The THINCARB model is an open source, free program built on principals laid down from a model developed from Neat et al. (1998). Unlike other models developed for DIC calculations, THINCARB is specifically built for calculations in non-marine environments. The authors of THINCARB translated the Neal et al. (1998) model into Excel and corrected minor formula errors, as well as added altitude compensation for $EpCO_2$ determination. The authors incorporated different macros to calculate for various estimates of $EpCO_2$ and charge balance of the dataset.

DIC and speciation are calculated from the principle that the molar concentration of an ion is equal to that ion’s activity, then divided by the appropriate activity coefficient. This principal accounts for both mono- and divalent activity coefficients and the model follows the most basic principle involved with DIC: that DIC is the sum of multiple sub-species of DIC, shown in Equation 3. THINCARB was coded to operate in Python as well, but was used in Excel for this study. Jarvie et al. (2017) developed THINCARB and then applied it to a 39-year dataset encompassing all major British rivers discharging into coastal zones to determine the model’s efficacy.

$$DIC = [CO_{2\,aq}] + [H_2CO_3] + [HCO_3^-] + [CO_3^{2-}] \qquad \text{(Equation 3)}$$

4.6 Calculation of DIC and CO_2 Flux

DIC flux and CO_2 flux was only calculated for the sites that had a measured discharge value: Gígjökull, Steinsholtsjökull, and Sólheimajökull; however, discharge data from a 2016 study was used for Falljökull. Glacier surface stream data were not included nor used in the calculation of DIC flux or CO_2 flux. The DIC flux was calculated by multiplying the stream's discharge (Q) by the concentration of DIC (mg/L) and the CO_2 flux by multiplying Q by the CO_2 concentration (mg/L). Discharge was converted from m^3/sec to L/year and DIC and CO_2 was converted from mg/L to kg/L. The CO_2^- concentration was determined by multiplying the percent contribution of CO_2 by the DIC, thus determining, in mg/L, the concentration of CO_2 within the sample. In both the cases of DIC flux and CO_2 flux determination, the final results are presented in g/yr format. To determine the portion of carbon of the CO_2 flux, the molar weight of carbon was divided by the molar weight of carbon dioxide, and then multiplied by the concentration of CO_2 in mg/L. Since the sampling occurred during baseflow conditions, the results from this study are indicative of conservative, baseflow carbon flux characteristics of the studied glacier rivers.

4.7 Data Processing

Data were stored and processed using Microsoft Excel and SigmaPlot 11.0. ArcGIS was used to create and compile study area maps for each of the sample sites. Ion concentrations, elemental ratios, and species partitioning were used to provide additional insight and information for carbon sourcing. Raw data were stored and processed primarily in Microsoft Excel, which was also used for the construction of the tables. SigmaPlot was used for data processing and figure creation. As discussed in section 4.5, the THINCARB model developed by Jarvie et al. (2017) was used to calculate DIC and $EpCO_2$ concentrations of the samples using the Excel option for the model.

DIC, $EpCO_2$, Ca concentration, and $\delta^{13}C_{DIC}$ were plotted together to show trends in CO_2 sources affecting the hydrogeochemistry of the streams. pH, water temperature, alkalinity, and DIC were plotted to show downstream trends in processes affected by influencing carbon flux. DIC concentrations in mmol/L were plotted against $\delta^{13}C_{DIC}$ to show the distribution of samples in terms of the most dominant weathering system. DIC (mmol/L) was plotted against Ca (mmol/L) to determine the dominant weathering system, carbonate or silicate, at each of the major sites: Gígjökull, Steinsholtsjökull, Sólheimajökull, and Falljökull. Various tables were constructed using the source sample, typically the glacier surface stream sample, pro-glacial lake sample, if present, mid-stream sample, and end member sample to display conservative trends in geochemistry. These tables provide a basic visualization of the overall geochemical relationships within the rivers. The figures were constructed to provide a more in-depth explanation of longitudinal trends. The flux data were also used to determine a conservative, low-end estimate of yearly flux from the studied glacier rivers.

CHAPTER FIVE: RESULTS AND DISCUSSION

5.1 Introduction

The hydrogeochemistry and carbon flux of four glacial-fed rivers (GIG, SHJ, SOL, and FAL) in Iceland were examined during the spring-summer season transition in 2017 to determine the function of the systems with respect to carbon flux, the volume of carbon within the system, species of DIC, and sources of carbon. A longitudinal sampling strategy was applied at each glacial river to capture changes in the hydrogeochemistry, including the possible transformation of carbon, downstream. An ancillary, ten-hour temporal study was done at one site, Sólheimajökull, to capture any diurnal influence over the river's hydrogeochemistry, despite the 20 hours of daylight present during the sampling period. Although the sampling distance in this longitudinal study was short, downstream changes in carbon were measured and observed.

Various elemental ratios were calculated and used to support the determination that the studied rivers are acting as sources of CO_2 with respect to the atmosphere. Geochemical data indicate that excess CO_2 is transformed to HCO_3 within the river and, subsequently, transported downstream to the ocean. HCO_3 was the most dominant form of DIC, averaging 83.5% the total of DIC for most of the rivers, which agrees with results from other similar studies in both Iceland (Stefánsson and Gíslason 2001) and elsewhere (Khadka et al. 2014). Although all the glaciers within this study are located within the region of Iceland that receives the most rainfall and have relative similarity in lithology, the differences between the glaciers and their associated rivers are more influential in the geochemical characteristics of the systems. These differences are the result of glacier dynamics, watershed size, groundwater inputs, and volcanic influence and are discussed further below.

5.2 Glacial Stream Geochemistry

Multiple geochemical processes occur, at different rates and quantities, within surface waters. Examples of such processes include carbonate dissolution, silicate dissolution, generation of carbonic acid, and generation of sulfuric acid from sulfide oxidation (Equations 4, 5, 6, 7, respectively). These reactions, and their products, are an indication of the dominance of different weathering processes active within a stream. Carbonate dissolution, indicated in Equation 6, utilizes CO_2 to drive the reaction forward and produce carbonic acid. This process increases the acidity of the water, which will result in the dissolution of rock. The next step of this process produces HCO_3, a form of DIC.

$$2H^+ + CaCO_3 \leftrightarrow Ca^{2+} + H_2CO_3 \qquad \text{Equation 4}$$

$$4H^+ + Ca_2SiO_3 \leftrightarrow 2Ca^{2+} + H_4SiO_4 \qquad \text{Equation 5}$$

$$CO_{2(diss)} + H_2O \leftrightarrow H_2CO_3 \leftrightarrow 2H^+ + CO_3^{2-} \qquad \text{Equation 6}$$

$$FeS_2 + 15O_2 + 14H_2O \leftrightarrow 16H^+ 8SO_4^{2-} + 4Fe(OH)_3 \qquad \text{Equation 7}$$

The solute fluxes driven by these reactions are expected to be elevated in glacial environments in comparison to global averages (Thomas and Raiswell 1984; Dessert et al. 2003; Torres et al. 2017).

5.2.1 Basic Geochemical Characteristics

The geochemistry results suggest the studied glacial rivers are somewhat homogenous, likely due to similarities in environment, geographic location, and lithology, but with measurable differences in certain parameters. Table 5.1 displays the primary parameters influencing the geochemistry of the streams at glacier surface streams, subglacial outflow streams, pro-glacial lakes, and the final sample from the river, representing a sampling end member. Across most sites, temperature increased downstream as distance from glacier increased. The highest SpC values were observed at sites with the highest concentrations of ions, of which Ca and Mg are included.

The pH varies longitudinally across the different sites. The range of pH in this study was between 6.93 and 8.91. In a previous laboratory simulation, pH in Icelandic glacial rivers was measured to be in the 9 to 10 range in environments not exposed to the atmosphere and stabilized right above seven after exposure to the atmosphere (Gíslason and Eugster 1987). A field study of Icelandic groundwaters describes higher pH values as indicative of water that had substantially reacted with basalt (Sveinbjörnsdóttir et al. 1995). As indicated by the pH values in Figures 5.1 and 5.2, water-air interaction must begin before the subglacial river exits from the glacier, which is supported by the recorded temperatures at the subglacial outflow streams. Since the pH is influenced by different chemical weathering reaction products and reactants, the processes that dominate chemical weathering and erosion, such as runoff, age of rock, slope angle, and rock composition, will similarly influence pH. The highest pH values were recorded at Gígjökull and Falljökull (Figures 5.1 and 5.2).

The data displayed in Table 5.1 and Figures 5.1, 5.2, and 5.3 are the hydrogeochemical constituents within the studied rivers and indicate they are not homogenous. Similar results were observed in a hydrochemical study on meltwater rivers in the McMurdo Dry Valleys in Antarctica that determined glacier melt chemistry varies between glaciers and is influenced by the presence or absence of both cryoconite holes and dust debris (Welch et al. 2010), which varies by glacier. That same study characterized river geochemistry based on TDS and suggests rivers with values less than 20 mg/L are dominated by intensely-weathered silicate bedrock; however, rivers characterized by bedrock with an abundance of carbonate minerals are observed to have a range of TDS values between 40 – 250 mg/L. The TDS results of this study range from 1.5 to 315 mg/L; however, the lowest TDS measurements were taken from glacier surface streams. Not including those outliers, the TDS range is 20.1 to 315 mg/L. These data fall in line with previous measurements of TDS in surface waters dominated by carbonate mineral-rich bedrock (Dessert et al. 2003).

The drainage river of Sólheimajökull, Jökulsá á Sólheimasandi, has two major documented tributaries feeding into the river downstream of the pro-glacial lake, which may explain the variability in some trends downstream. Sólheimajökull is, however, a more mature system than Gígjökull, as indicated by the presence of a pro-glacial lake, developed moraines, large watershed, and long river. Subsequently, the geochemistry of this system is more stable and clear in the trends of its associated geochemical data.

Falljökull is unique, because the primary river sampled for the study was draining two distinct glaciers, Falljökull and Virkisjökull. While the system lacks a traditional pro-glacial lake, like those of Sólheimajökull or Steinsholtsjökull, the river, as it exits from the glacier, becomes shallow and pools in areas of slightly lower elevation, before channelizing into the river proper.

Table 5.1: Geochemical results at four sample sites: GIG, SHJ, SOL, SOLA, and FAL (Source: created by Author).

Geochemical Characteristics of Icelandic Glacial-Fed Rivers

Location	Site	Distance from source (km)	Water Temp. (°C)	SpC. (ɥs/cm)	pH	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	HCO_3 (mg/L)	$EpCO_2$	DIC (mg/L)	$\delta^{13}C$ DIC (‰)	Discharge (m^3/s)
Gígjökulsá (GIG)	GIG1$_1$	0.0	2.9	16.0	7.8	0.8	0.2	1.2	2.4	1.6	-9.3	1.35*
	GIG2$_2$	0.0	3.4	275.0	7.5	14.5	8.4	19.3	5.3	20.2	-1.1	
	GIG6$_3$	4.1	5.7	308.0	8.3	16.9	10.0	21.7	2.5	22.2	1.2	
Steinholtsá (SHJ)	SHJ1$_1$	0.0	1.2	20.8	8.1	1.6	0.7	2.6	0.9	2.7	-10.7	2.31*
	SHJ2$_4$	1.1	4.0	49.2	8.1	2.7	1.3	5.8	1.6	6.1	-7.7	
	KRO$_3$	4.7	3.8	85.4	8.0	5.2	2.7	7.8	1.0	8.0	-5.1	
Jökulsá á Sólheimasandi (SOL)	SOL1$_1$	0.0	0.5	2.4	7.5	0.3	0.1	0.6	1.6	0.9	-11.1	18.5*
	SOL3$_4$	0.8	3.2	98.1	7.1	9.3	2.4	9.0	7.7	10.3	-7.9	
	SOL11$_3$	10.5	5.6	84.6	7.8	8.6	2.7	8.9	2.1	9.3	-4.3	
Jökulsá á Sólheimasandi (SOLA)	SOL1A$_1$	0.0	2.1	2.2	8.2	1.2	0.4	1.0	2.3	1.4	-12.2	18.5*
	SOL3A$_4$	0.8	3.7	87.2	7.1	7.3	1.7	9.7	27.9	14.5	-5.0	
	SOL11A$_3$	10.5	3.5	82.4	7.7	6.8	1.8	8.5	7.9	9.8	-5.4	
Virkisá (FAL)	FAL1$_1$	0.0	2.2	33.0	8.1	0.2	0.0	3.9	42.6	11.2	-20.8	5.3 - 7.9**
	FAL2$_2$	0.0	0.1	19.8	8.9	1.8	0.7	3.9	2.2	4.2	-16.5	
	FAL5$_3$	2.9	2.6	31.1	7.6	2.3	10.0	3.9	6.6	5.0	-12.5	

$_1$ - Glacier surface stream or melted ice

$_2$ - Subglacial outflow stream

$_3$ - Final sample from river

$_4$ - Pro-glacial lake

* Measured by IMO

** From MacDonald et al. 2016

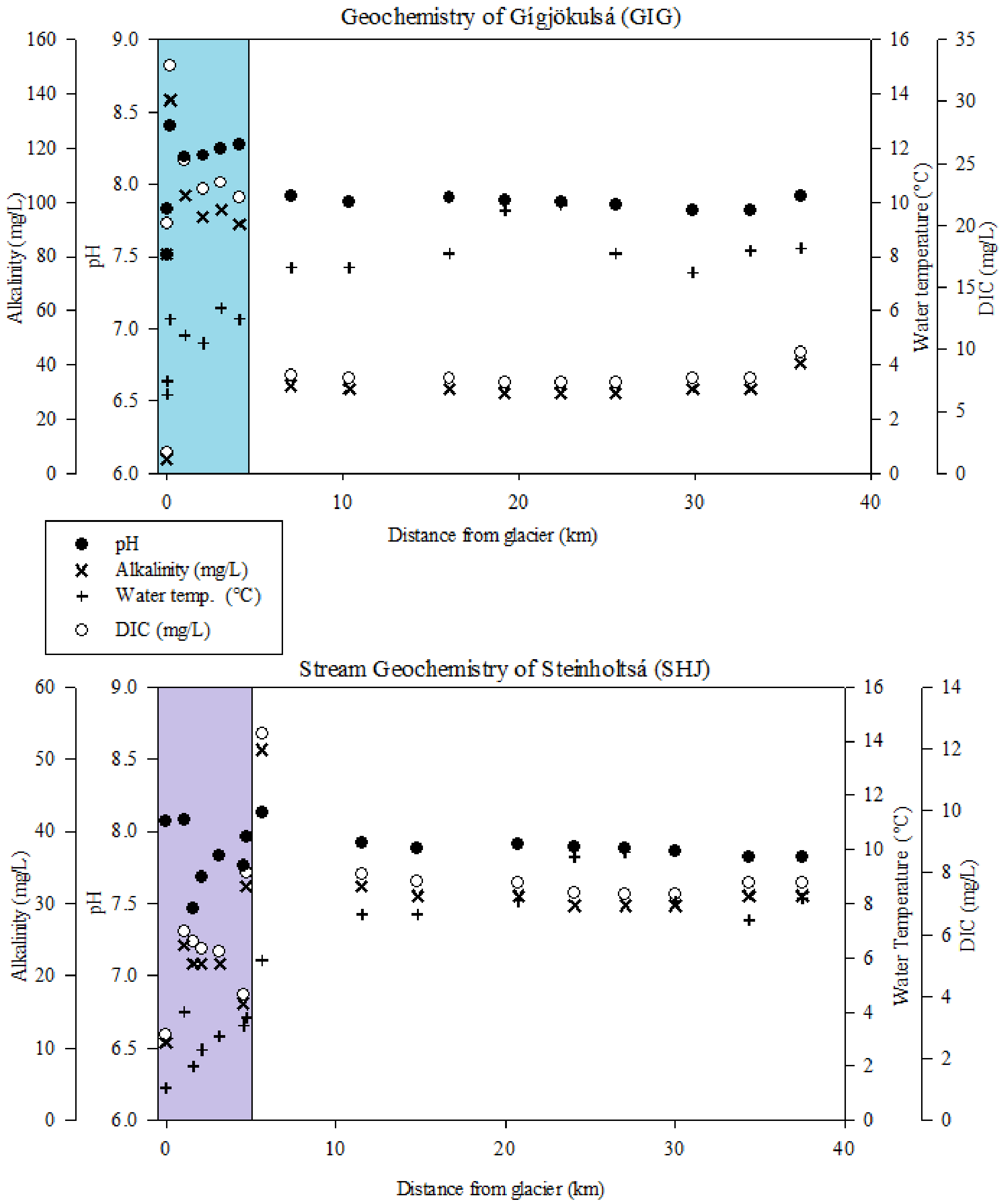


Figure 5.1: Basic geochemical parameters measured from the rivers of Gígjökull (blue background), and Steinsholtsjökull (purple background), with data from Markarfljót (white background) to display the downstream data from source (glaciers) to sink (ocean). The drainage from Steinsholtsjökull merges with Markarfljót upstream from Gígjökull and, therefore, had more samples from Markarfljót included (Source: created by Author).

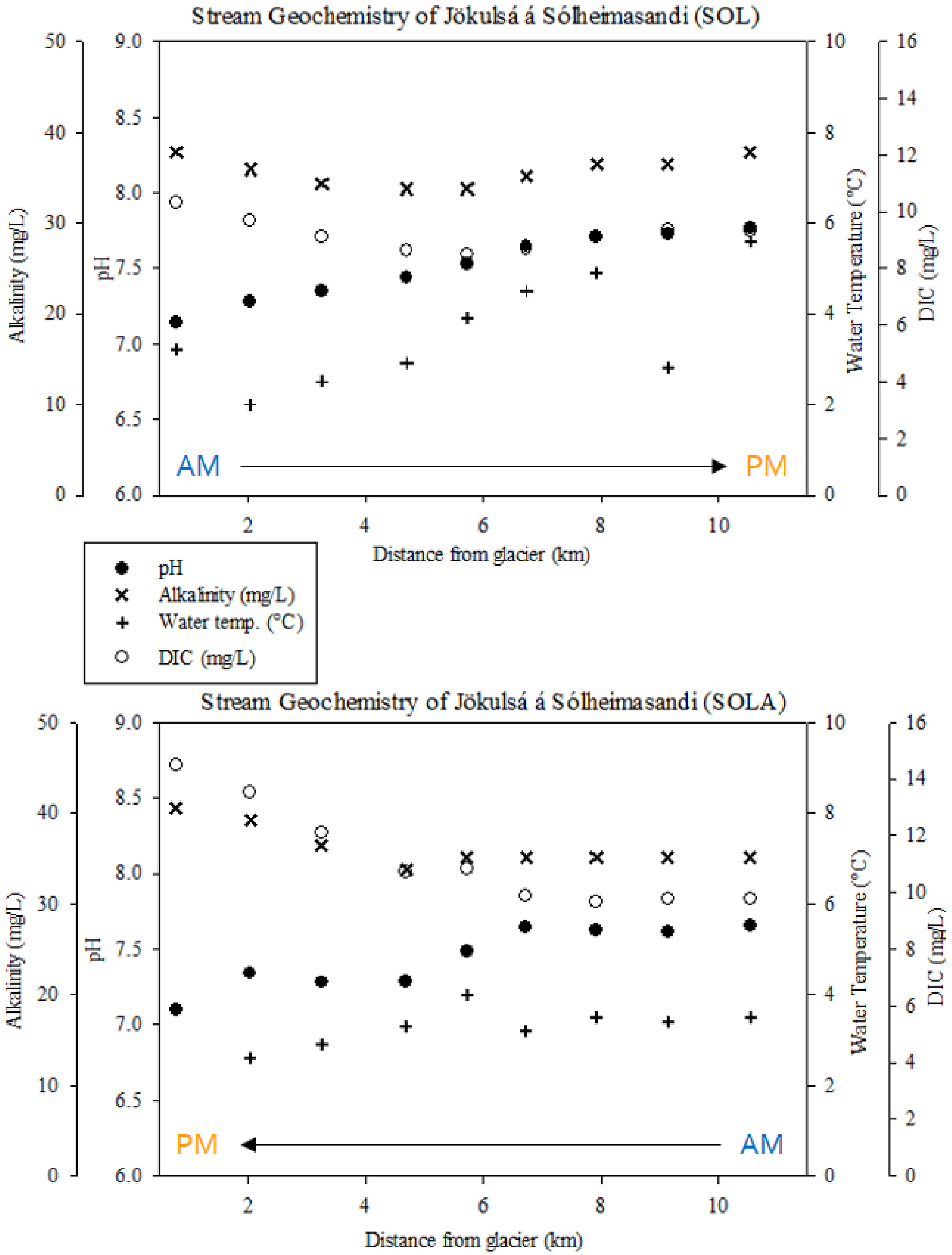


Figure 5.2: Basic geochemical parameters measured from the river of Sólheimajökull to display the downstream data from source (glaciers) to sink (ocean). SOL was sampled from glacier to ocean, beginning in the morning, and SOLA was sampled 48 hours later from ocean to glacier (Source: created by Author).

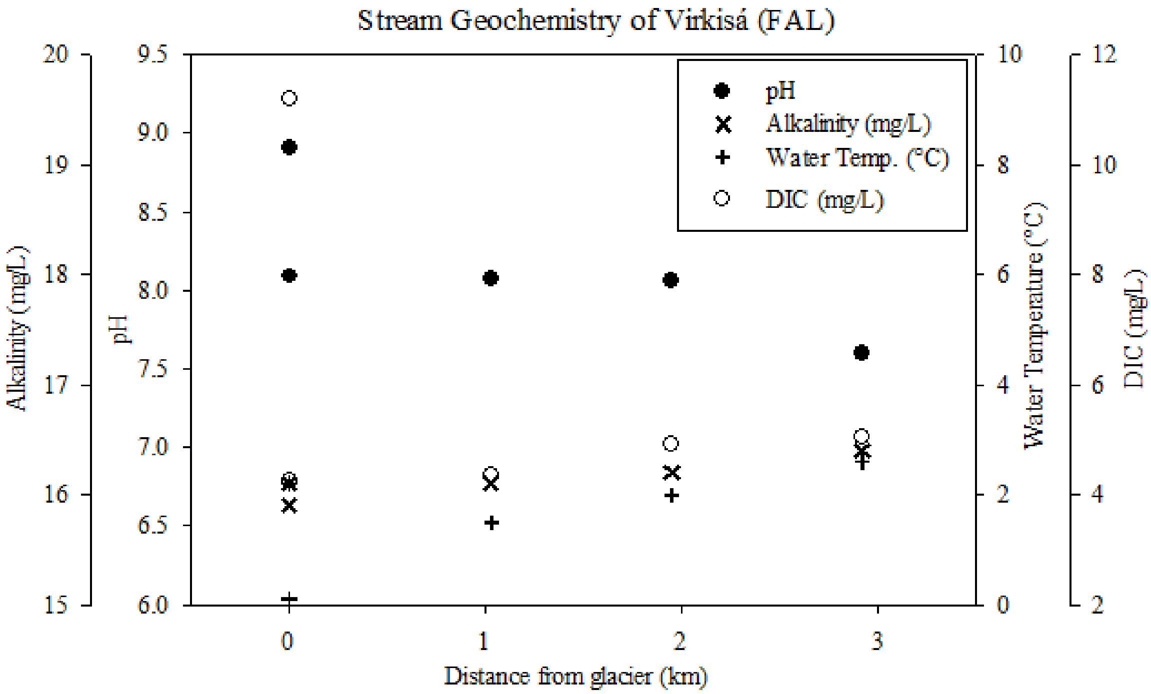


Figure 5.3: Basic geochemical parameters measured from Falljökull. Excluding the high concentration in the surface stream (distance = 0), the DIC concentration is lower at this location in comparison to the other study sites (Source: created by Author).

5.2.2 In-stream Weathering Processes

Figures 5.4, 5.5, and 5.6 provide additional insight into the in-stream geochemical processes affecting and influencing the carbon flux. Each site demonstrates some unique and similar longitudinal trends, which illustrates the complexity of the hydrogeochemical processes occurring in the streams and characterization attempts of these glacier rivers. These figures display changes in DIC, EpCO$_2$, Ca concentrations, and $\delta^{13}C_{DIC}$ downstream, from the source; however, the beginning samples are difficult to distinguish, due to the proximity of the surface stream sample to the next downstream sample, either a pro-glacial lake or river sample. Taking the environmental context into consideration, such that meltwater from the glacier is the ultimate source of most of the water in the system, whether for sub-glacial or supraglacial processes, the glacier surface stream sample is used in this discussion as the source.

Since $EpCO_2$ is a ratio between the excess CO_2 in the river and the CO_2 in the atmosphere, increasing values indicate an increase in CO_2 concentration in the water. The opposite is true of decreases in the $EpCO_2$ values: decreasing $EpCO_2$ values mean the CO_2 concentration in the river water is getting lower. The longitudinal trends, however, are more complex and insightful regarding the in-stream processes. Excluding Falljökull, the concentration of Ca ion remains relatively stable longitudinally, despite measurable changes in pH, water temperature, and $EpCO_2$, and HCO_3. When considering the relative stability of other ions as well, this trend suggests the streams are at, or near, equilibrium with respect to chemical weathering. The variability of the HCO_3 concentrations, despite the relative stability Ca, suggests atmospheric exchange of CO_2 has an influence over the inorganic carbon flux.

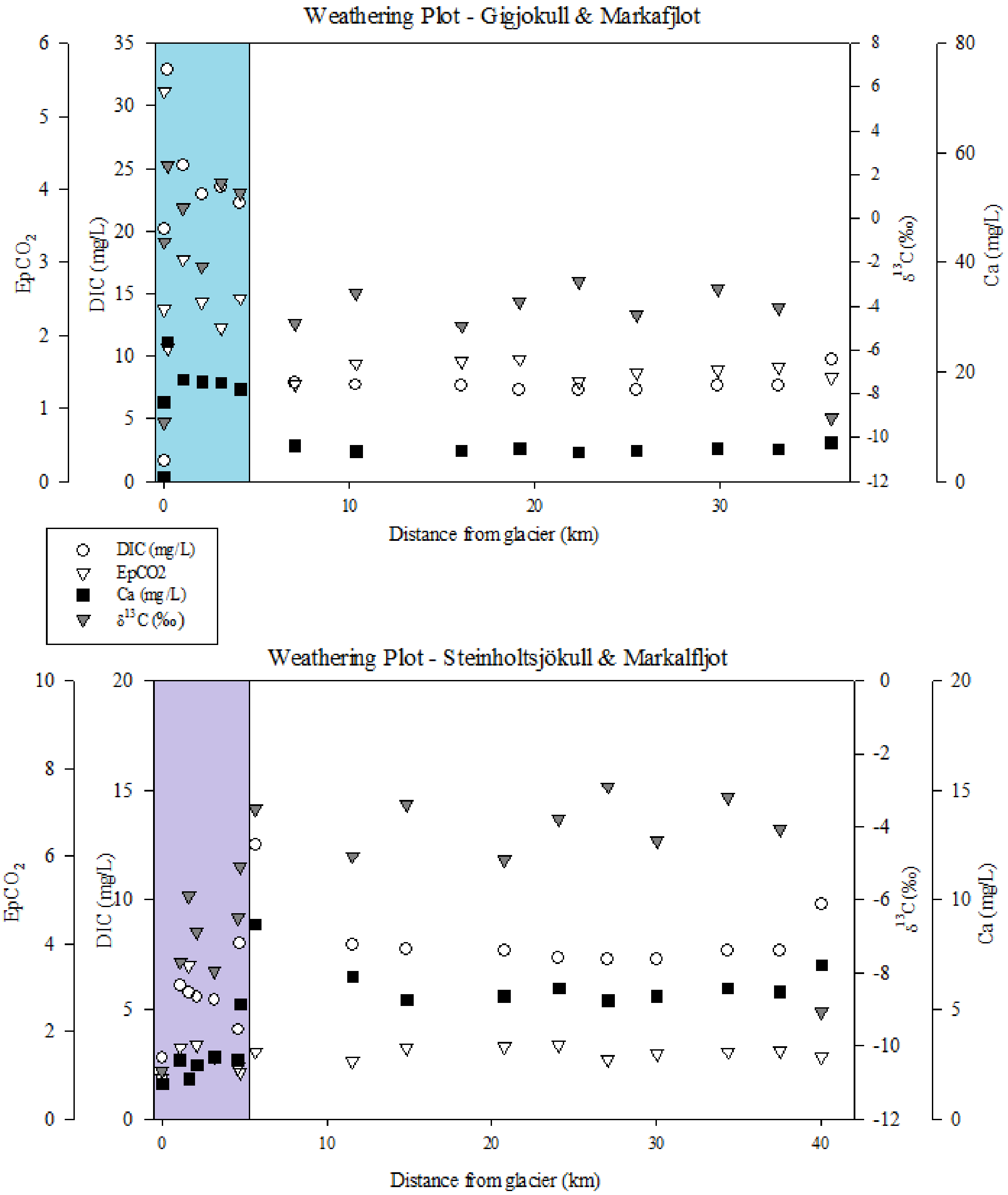


Figure 5.4: Weathering plots of Gígjökull and Steinsholtsjökull, with the appropriate Markarfljót data included. The X-axis across all plots is the distance downstream from source, while the parameters displayed on the Y-axis include EpCO$_2$ concentration, Ca, Mg, DIC concentration, and $\delta^{13}C_{DIC}$ values (Source: created by Author).

The longitudinal trends in DIC and $EpCO_2$ at Gígjökull and Steinsholtsjökull are complex. The larger river, Markarfljót, was sampled to serve as an end member source for analysis purposes for both Gígjökull and Steinsholtsjökull. GIG's elevated Ca, Mg, and HCO_3^- concentrations, despite having the smallest watershed, are indicative of the susceptibility of younger, glassy volcanic material to chemical weathering (Gíslason and Eugster 1987; Louvat et al. 2008). Concentrations of HCO_3 are highest with respect to the three primary species of DIC at all sites, with the combined dissolved components of CO_2 and H_2CO_3 being the second most concentrated. This determination agrees with the results of similar studies (Singh and Hasnain 1998; Khadka et al. 2014). There are multiple sources of HCO_3 in rivers, depending on the weathering regime, that come primarily from the exchange of atmospheric CO_2 with water (Amiotte-Suchet et al. 2003). The first source is after the CO_2 reacts with water to form carbonic acid. The carbonic acid then dissociates to form HCO_3 and an H^+ ion, which drives the pH of the solution lower, or it is buffered by increased alkalinity from mineral weathering. The presence of carbonic acid acts as a source of weathering and causes dissolution of carbonate mineral rich basalt and silicates. Although CO_3 concentrations were calculated, the results were negligible and contributed little to the DIC concentrations at these sites.

Gígjökull displays highly variable trends in all parameters represented in Figure 5.4, besides the Ca concentration. The river geochemistry stabilizes after the river from Gígjökull converges with Markarfljót. Figure 5.4 displays clearly the significantly higher concentration of ions present at Gígjökull, which dilutes and decreases in magnitude after mixing with Markarfljót. Steinsholtsjökull is different with respect to trends in DIC concentration, $EpCO_2$ values, and the magnitude of the ions. Gígjökull has much higher ion concentrations than the other sites and is the only glacier within the study to have most recently experienced an extrusive volcanic eruption recently. A portion of the lava flowed beneath the glacier and deposited highly volatile material, which is susceptible to weathering in comparison to the older volcanic deposits that characterize other sites. The highest ion concentrations of Gígjökull belong to the sample taken from an input tributary that drains a separate portion of the glacier and runs through a major crack in volcanic bedrock. From that point, the ion concentrations stabilize, but continue in a slight decreasing trend downstream.

Steinsholtsjökull differs from Gígjökull in ways that may explain these differences. Although both glaciers are outlets of Eyjafjallajökull, Steinsholtsjökull is a larger glacier in volume, has a pro-glacial lake, and multiple, major tributaries. Figure 5.5 shows the confluence of one such input, and is the sampled tributary included in the sample set. From field observations, the tributary was most likely draining glacier surface melt as the water was much clearer and colder than the river samples of Steinsholtsjökull. DIC decreases clearly downstream, despite a somewhat stagnant trend of $EpCO_2$ values. Similar to Gígjökull, however, once the drainage river of Steinsholtsjökull converges with Markarfljót, the geochemical constituents contributed by Steinsholtsjökull become indistinguishable from the main river.

Figure 5.5: Image of a sampled tributary stream draining primarily surface melt from Steinsholtsjökull mixing with the main drainage river of the glacier. Notice the difference in water clarity and turbidity (Source: created by Author).

The final sample from the Steinsholtsjökull dataset is after the river's confluence with another major river: the river Krossá, which is much larger in terms of velocity, volume, drainage, and watershed size. Following the Krossá sample are the Markarfljót samples. Markarfljót displays decreasing concentrations of DIC downstream, but increasing, albeit slightly, concentrations of Ca. These data indicate that once the glacier water mixes with the primary body of water within Markarfljót, the geochemistry of the individual glacier inputs become indistinguishable using traditional geochemical indicators. Markarfljót displayed a decreasing trend in DIC and $EpCO_2$ downstream. This suggests another dominant process occurring that is influencing the carbon flux downstream. The large catchment area provides larger magnitudes of ions which contribute to the alkalinity of the water. Among others, this is one reason the geochemistry of Markarfljót shows a relatively stable system; however, Markarfljót also deepens and increases in velocity in downstream, subsequently increasing the intensity of turbulence in the river and thus, CO_2 degassing. A link has been established in previous studies that show a correlative relationship between stream turbulence and an increase in CO_2 degassing (Wang et al. 2013), which translates directly to a fractionation effect on $\delta^{13}C$. As turbulence increases the occurrence of CO_2 degassing, the lighter isotope, ^{12}C preferentially degases leaving the heavier isotope, ^{13}C, behind; this process results in the increase of $\delta^{13}C$ values. Thus, the highly variable ranges of $\delta^{13}C_{DIC}$ values are indicative of in-stream fractionation effects and changes in surface stream or groundwater inputs (Telmer and Veizer 1999; Doctor et al. 2008; Khadka et al. 2014).

Despite the fact that Gígjökull and Steinsholtsjökull are from the same ice cap, Eyjafjallajökull, the differences between the two sites are clear. The difference in lithology between Gígjökull and Steinsholtsjökull, due to the eruption of Eyjafjallajökull in 2010, explains the vast difference in ion and DIC concentrations between the two sites. Although Gígjökull is the smallest glacier draining the smallest watershed within the study, that system has the highest flux of ions and DIC than any other site, due to the presence of young, recently deposited hyaloclastites, a rock highly susceptible to weathering; however, both are similar in that post-mixing with Markarfljót, the geochemical parameters stabilize, although the isotopic signature of the water continues to vary downstream. These trends alone provide inadequate in the determination of the rivers as a source or sink of carbon, therefore, other processing and analysis methods would be helpful.

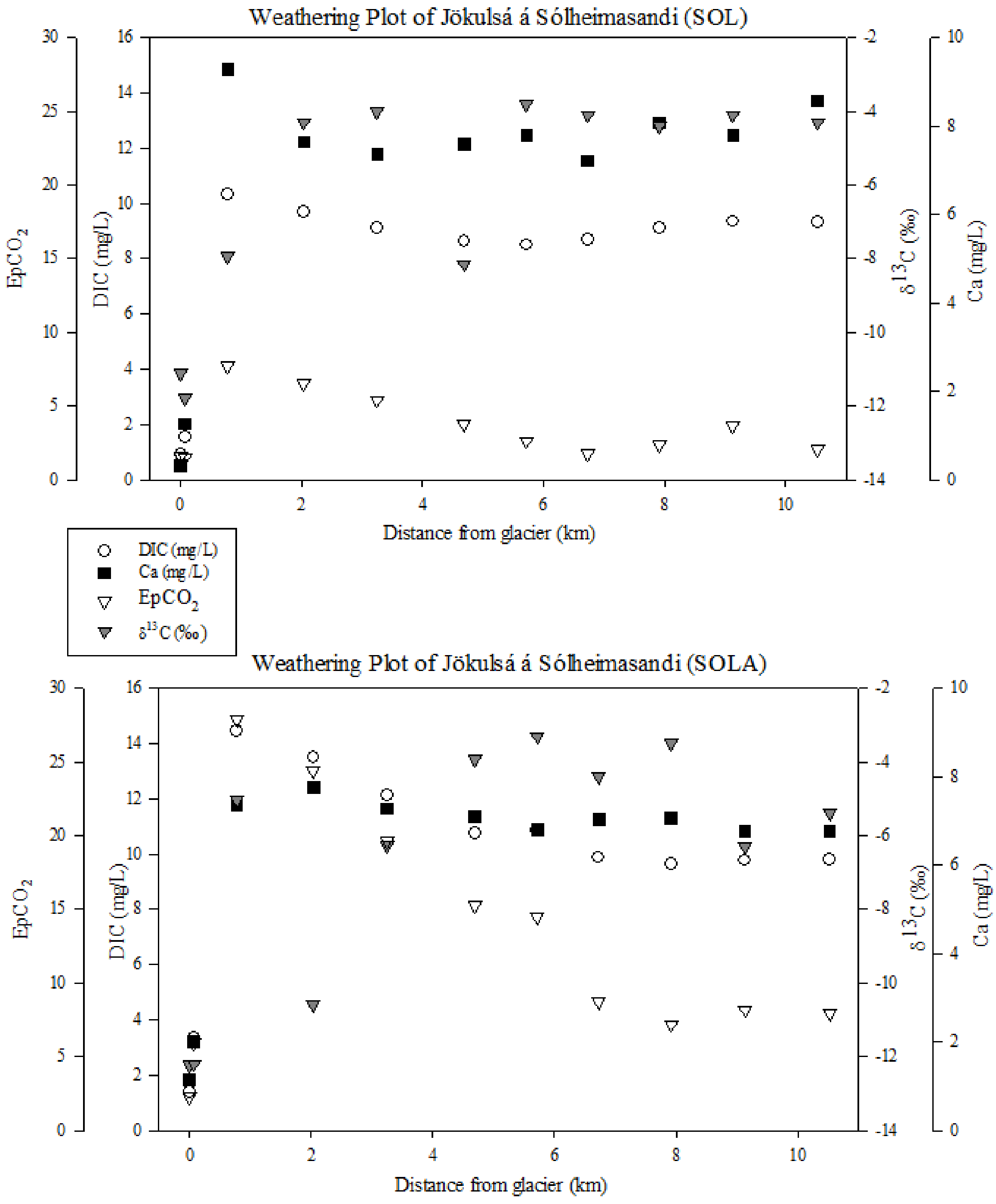


Figure 5.6: Weathering plots from both datasets at Sólheimajökull (SOL and SOLA). Carbon-related parameters were measurably different between the two datasets, sampled 48 hours apart and beginning at different ends of the river (Source: created by Author).

Longitudinal trends, in terms of the glacial surface stream sampling being the source, indicate a slight decrease in all parameters in both Sólheimajökull datasets. The Sólheimajökull datasets, SOL and SOLA, are slightly different in maximum DIC concentrations; however, both share the similar, but slight trend of decreasing downstream. The second set of samples, SOLA, displayed greater variation in DIC and $EpCO_2$ longitudinally, as well as more variation in $\delta^{13}C_{DIC}$. Both samples were collected on days of similar weather conditions: partly cloudy skies, warm temperatures, and no precipitation. SOL was sampled 48 hours before SOLA from glacier to ocean, starting in the morning. SOLA was sampled from ocean to glacier, starting in the morning. This was done to capture any diurnal influence on the riverine processes affecting DIC and carbon flux, including changes in temperature.

As seen in the SOL and SOLA weathering plots Figure 5.6, there were deviations in the SOLA sample set from the trends in the SOL sample set. On the day of the SOL sampling, air temperature values ranged from 11.8 °C in the morning to 13.0 °C in the evening, which was also the highest temperature recorded for that day. During the SOLA sampling, the temperature values were 11.6 °C in the morning and 15.7 °C upon conclusion of sampling. This means that temperature differences during sampling would have been greatest at the samples closest to the glacier and more alike during downstream sampling. This is reflected in the data, as the DIC concentrations increase in the SOLA dataset to a magnitude similar to the same downstream samples in the SOL dataset. The day the SOLA samples were gathered experienced a higher overall temperature range and maximum temperature than the sampling day for SOL. These results show the influence that surface air temperatures have on in-stream geochemical processes, and, ultimately, carbon flux. Glacial-fed rivers in Iceland display strong diurnal variations in discharge, due to daytime melting. The diurnal variation is seen during the melting period, but is absent in winter (Einarsson 2012). Diurnal influences in this study were mainly in response to seasonal temperature influence and associated changes in discharge. Water temperature measurably increased closer to the glacier in the SOLA sample set. The samples taken closest to the glacier would have been sampled later in the day, and, thus, had more time to respond to the increase in temperature throughout the day.

Similar to the previously discussed sites, Ca concentrations in both sets remained relatively stable with little variation downstream. In both datasets, the $EpCO_2$ and DIC values decreased from source to mouth. The weathering data wasn’t used alone to make the source or sink determination for each study river. However, the decreasing downstream trend in $EpCO_2$ suggests the rivers are losing CO_2. The CO_2 is likely transforming into a different form of carbon or degassing to the atmosphere. The latter of these instances is more likely, since the other geochemical parameters indicate the rivers to be at, or near, equilibrium with respect to carbonate weathering processes.

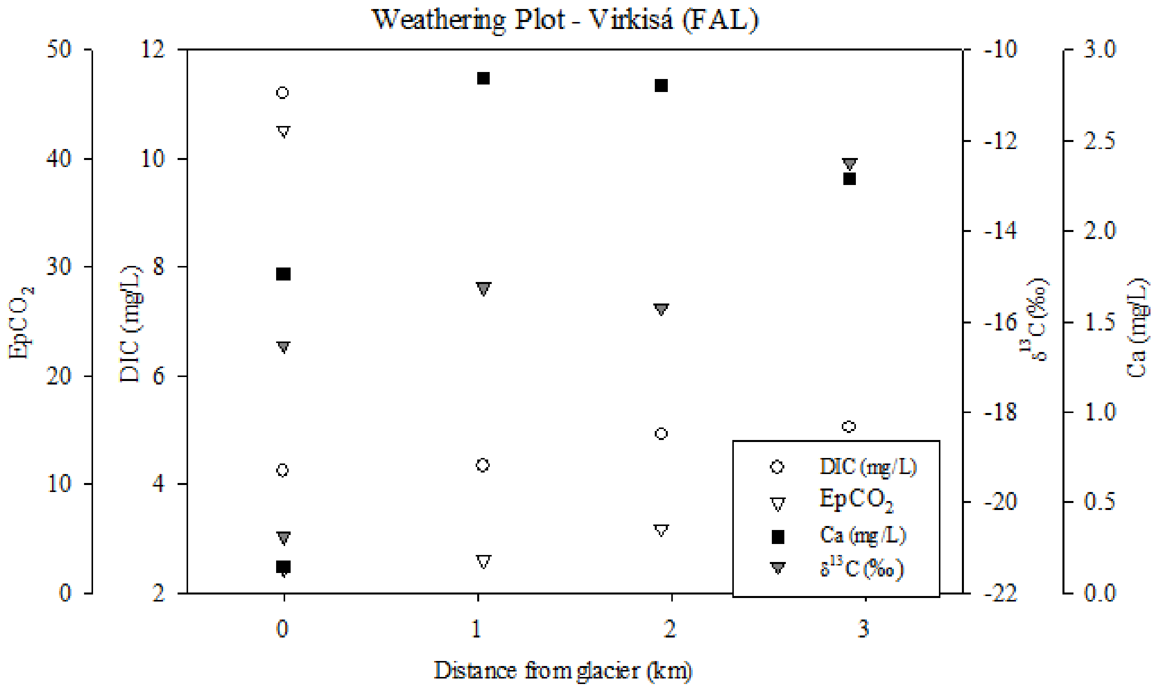


Figure 5.7: Weathering plots of Falljökull. The X-axis across all plots is the distance downstream from source, while the parameters displayed on the Y-axis include EpCO$_2$ concentration, Ca, Mg, and DIC concentration, and $\delta^{13}C_{DIC}$ values (Source: created by Author).

Although fewer samples were taken from the river at Falljökull, longitudinal trends are still visible, though less distinguishable. In Figure 5.7, an increase in the EpCO$_2$ values is visible. DIC concentration, however, remains relatively constant after the initial spike in DIC within the glacier surface stream. In addition, $\delta^{13}C_{DIC}$ values from Falljökull were more depleted than the other sample rivers.

5.3 DIC and $\delta^{13}C$ Interpretation

The $\delta^{13}C_{DIC}$ value of a sample, when used as an environmental tracer, is an indicator of the carbon sources that contribute to the carbon within the river's system. $\delta^{13}C_{DIC}$ values from this study ranged from -20.76‰ to 2.41‰ with an average of -7.74‰. As indicated Figure 5.8, the DIC and $\delta^{13}C_{DIC}$ results were split up into three separate groupings. Surface streams were more depleted, with respect to $^{13}C_{DIC}$, in comparison to pro-glacial lake and downstream river samples. Generally, the $\delta^{13}C_{DIC}$ values are more enriched within the pro-glacial lake samples than the surface stream samples. The relative depletion of the $\delta^{13}C_{DIC}$ results in the surface stream samples suggest the surface stream samples are either a mixture of multiple sources with heavier $\delta^{13}C_{DIC}$ values or fractionation effects are influencing the surface streams, most likely through degassing of CO_2 or from biochemical processes (Doctor et al. 2008). Atmospheric exchange of CO_2 and temperature influence fractionation in stable carbon isotopes and have been observed in glacial environments to effect $\delta^{13}C_{DIC}$; however, fractionation effects from water-debris interaction and organic processes cannot be ignored either, since these processes are known to occur on the surface, within, and beneath a glacier. These are likely possibilities, as the surface streams take variable paths as the water travels off the glacier with varying sediment and rock interactions, resulting in subsequent fractionation and potential alteration of the original source carbon (Tranter et al. 2002; Skidmore et al. 2004; Doctor et al. 2008).

Downstream river $\delta^{13}C_{DIC}$ values are similar to the $\delta^{13}C_{DIC}$ values from the pro-glacial lake samples, thus demonstrating the mixing effect the pro-glacial lake has on the carbon dynamics of the river. Figure 5.8 displays the DIC concentration and $\delta^{13}C_{DIC}$ results. With little exception, most sites trended similarly, with more negative isotopic values corresponding to lower DIC concentrations. Glacial surface streams, with the exception of Falljökull, are characterized by the lowest DIC concentrations and the most depleted values of $\delta^{13}C_{DIC}$. In both Gígjökull and Falljökull, the samples from the immediate outputs of sub-glacial waters were lower in DIC concentration and in the lower range of $\delta^{13}C_{DIC}$ values for their respective sites. Sites where pro-glacial lakes were present, Sólheimajökull and Steinsholtsjökull, were similar in $\delta^{13}C_{DIC}$ values and similar, but slightly lower in DIC concentration.

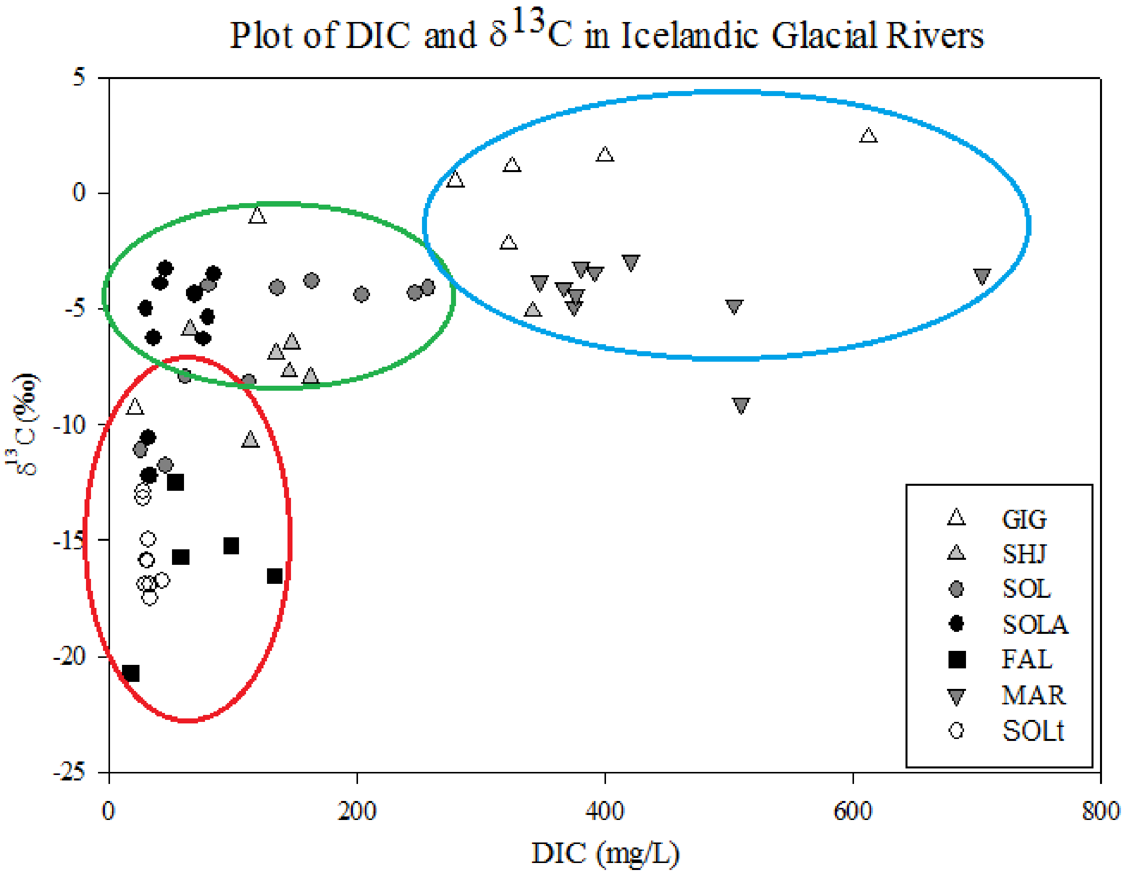


Figure 5.8: Plot of DIC (mg/L) and $\delta^{13}C_{DIC}$ (Source: created by Author). The data have been separated in three groups designated by a red circle (low DIC, most negative $\delta^{13}C$), green circle (low DIC, less negative $\delta^{13}C$), and blue circle (high DIC, less negative $\delta^{13}C$) (Source: created by Author).

Figure 5.8 displays the DIC and $\delta^{13}C$ data. The data was separated into three separate groupings as indicated by the red, green, and blue circles. The first group, circled in red, highlights the data grouping with DIC concentrations below 200 mg/L and the most negative $\delta^{13}C$ values, which range between -20.76‰ and -7.96‰.

The second grouping, circled in green, is characterized by low DIC concentrations and slightly less negative $\delta^{13}C$ values. This group is made up of the pro-glacier lake samples and downstream river samples, which shows the efficacy of mixing that occurs within the pro-glacier lakes downstream. This suggests that not only is the geochemistry of the pro-glacial lake samples heavily influenced by sub-glacial waters, but they are also a dominant influence on the geochemical characteristics of the rivers, post-mixing of all inputs to the systems. The river samples, as well as the pro-glacial lake samples, cluster between -3.5‰ and -9‰ being closer to, or slightly more enriched than, the atmospheric isotopic value of CO_2.

The third grouping, circled in blue, are samples with DIC values between approximately 300 mg/L and 650 mg/L and $\delta^{13}C$ values which range from -9.07‰ to 2.40‰. The first group is primarily made up of glacier surface stream samples, the temporal Sólheimajökull samples, and the Falljökull samples. Gígjökull and Markarfljót samples compose the third group.

This plot shows a distinct difference in glacier surface stream samples compared to downstream samples. The isotope values are more positive downstream from where the sub-glacial water first exits beneath glacier and is exposed to the atmosphere. Nearly all sites, excluding the second set of Sólheimajökull data (SOLA), have end members at lower DIC concentrations and more depleted isotope values and higher DIC concentrations with more enriched isotope values. These end members, and the plotted values on Figure 5.8, suggest fractionation occurring downstream during carbonate mineral dissolution and the consumption of carbon during that process, as well as the degassing of CO_2. This trend agrees with the results from a study of similar focus (Burns 2016), which suggests the more depleted isotopic signatures, when paired with $EpCO_2$ results, is a possible indication of early stage carbonate dissolution (Burns 2016).

Changes in $\delta^{13}C$ values are indicators of both fractionation effects and additional source inputs; therefore, the variability in the $\delta^{13}C$ values is also a possible indicator of surface stream inputs and geothermal inputs. In the weathering plots, Figures 5.4, 5.6, and 5.7, the $\delta^{13}C$ values display the most change downstream and no obvious trend. While no data from this study specifically address geothermal or hydrothermal fluids, previous studies have established that geothermal waters leach into surface streams (Burns 2016). These inputs have $\delta^{13}C$ values much closer to the value of mantle-sourced CO_2, which is typically more enriched and could be the influence at some sites, like Sólheimajökull, which have more enriched isotope values compared to the measured end members.

5.4 Elemental Ratios

5.4.1 Alkalinity to DIC Ratio

The ratio between alkalinity and DIC indicates the likelihood of weathering processes to be a source or sink of carbon in a given environment, depending on the balace between sulfide oxidation and silicate weathering (Torres et al. 2017). The average ratio of this study was 1.54, with a range of 0.62 to 2.31. According to Torres et al. (2017), the Alk:DIC ratio within the oceans is fixed at 1:1, while the Alk:DIC ratio of weathering processes varies. Based on their study, weathering processes that yield lower ratios are more likely to be a source of CO_2 to the atmosphere. Torres et al. (2017) established a link between Alk:DIC ratios to various forms of weathering processes, pairing the type of weathering, carbonate or silicate, with the acid present, carbonic or sulfuric. This resulted in three indicator ratios: an Alk:DIC ratio of less than one suggests the weathering is a source of CO_2, a ratio between one and two indicates the weathering is a source of CO_2 on longer timescales, and greater than two indicates that weathering is serving as a CO_2 sink. The majority of ratios from this study suggest the dominant weathering processes occuring within the sampled rivers are acting as a source of CO_2 on long timescales. The results from the temporal study, SOLt, are characterized by ratios less than one, suggesting that stream is acting as an immediate source of CO_2 to the atmosphere. SOLt data were the only temporal data in this study and were taken from the side of a stream catching runoff from surface and subsurface melt. This stream was located less than 10 m from the water's exit from the glacier. As such, it is likely this stream is acting as an immediate source of CO_2 to the atmosphere, due to the change partial pressure, and subsequent degassing of CO_2, that would occur as the pCO_2 of the water would quickly attempt to equilibrate with the pCO_2 of the atmosphere. No ratios from this study exceeded two; therefore, none of the study rivers were behaving as sinks of CO_2, according to this calculation method.

Figure 5.9 shows the longitudinal trend of the Alk:DIC ratios downstream, As shown, the downstream trend varies across most sites; however, slight increases at most sites are present. Gígjökull had the highest ratios, varying above and below 1.8. The higher ion concentration present at Gígjökull would contribute to the alkalinity of the river, thus driving that ratio higher. Gígjökull and Steinsholtsjökull both show little variation in the Alk:DIC ratio longitudinally. The Sólheimajökull sample showed the lowest ratios, but had some of the largest variation downstream, showing an obvious increase in ratios after the lagoon sample (sample 3). Alk:DIC ratios measured at Falljökull were different from the other sites, as the ratios decrease downstream.

The THINCARB results agree with those determinations and specifically indicate that all sites act as CO_2 sources to the atmosphere, despite a decreasing trend in $EpCO_2$

values present in the Markarfljót samples. The Alk:DIC results agree with a previous determination from a study that determined carbonate minerals, with respect to their role in weathering, are important to carbon cycling, especially to local environments and on shorter time scales in temperate glacial regions, such as Iceland (Khadka et al. 2014; Torres et al. 2017).

These results suggest the studied rivers, during the peak melt season between spring and summer, are acting as sources of CO_2 to the atmosphere. This determination showcases the complexity of these rivers and the multiple processes acting within them that are both using carbon and producing carbon. Chemical weathering is usually a sink for carbon, however, as seen in these results, is not a dominant enough process to utilize enough CO_2 to characterize the rivers as sinks solely based on these data. This may be due to the relative short distance of the studied rivers and, thus, not enough distance for the chemical weathering process to equilibrate with the CO_2 lost to degassing and river turbulence, or to consume the quantity of CO_2 remaining in the river. Icelandic glacial-fed rivers are heavily influenced by surface conditions, particularly when surface conditions lead to increased glacier surface melt, like during the season these samples were collected. The glacial fed rivers are turbulent, generally channelized, and transport large volumes of water and sediment. This combination of characteristics result in a riverine environment susceptible to atmospheric exchange.

As indicated by the results of this study, CO_2 drawdown through chemical weathering reactions and CO_2 degassing through atmospheric exchange are both occuring at differing rates and in amounts. Outside influences, such as daily and seasonal weather, may potentially alter the current influence of those processes over the other. Seasonal studies to identify the carbon flux of the studied rivers would elucidate any speculation on how the rivers act during other seasons, both during baseflow and flood events.

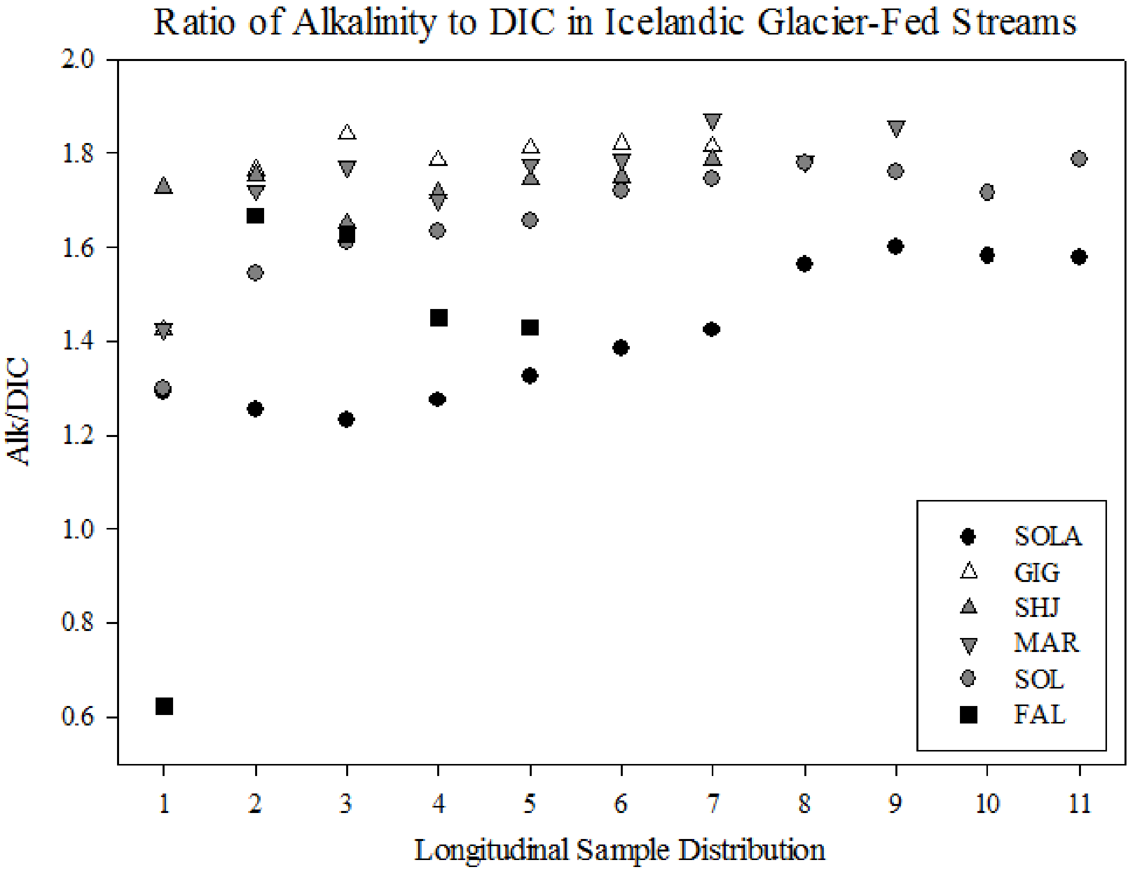
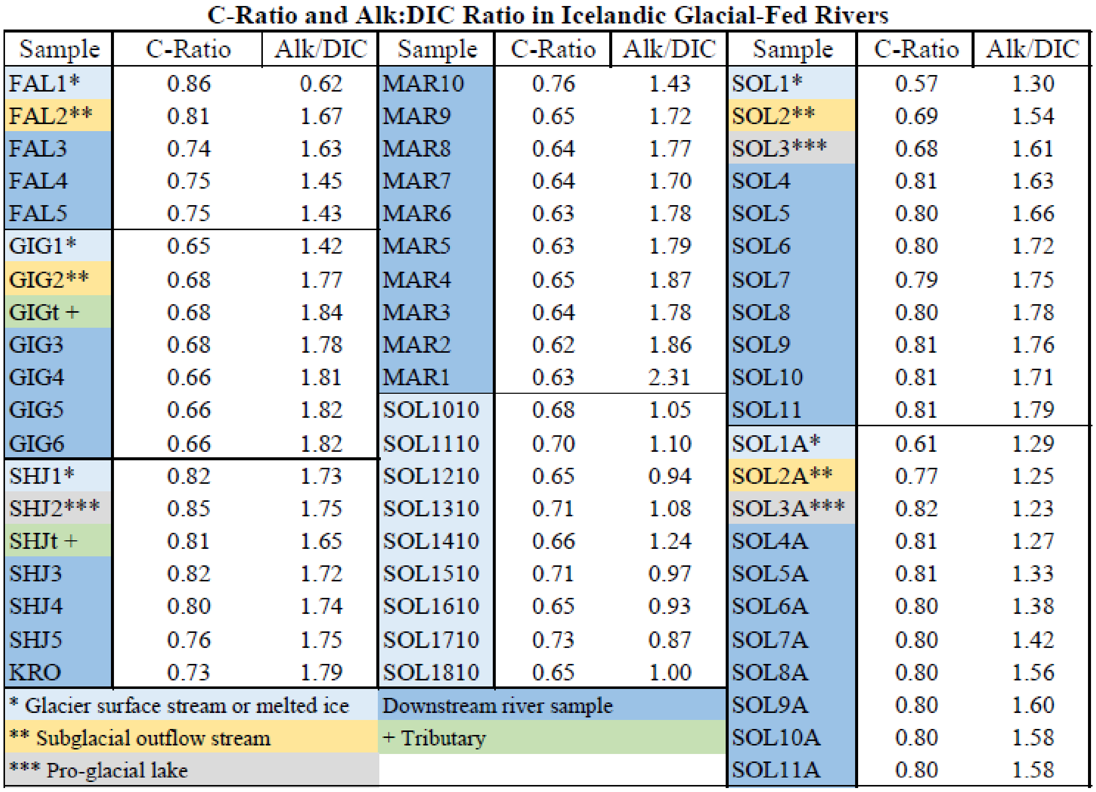


Figure 5.9: Longitudinal distribution of the Alk:DIC ratio for all sites. The numerals on the X-axis indicate the sample taken in the datasets, where “1” is equivalent to the first sample taken from a river, typically the glacier surface stream sample. Ratios are between the Alk:DIC thresholds of 1.0 and 2.0, thus showing the evolution of carbon on longer timescales (Source: created by Author).

5.4.2 C-Ratio

Table 5.2 shows the C-ratios and Alk:DIC ratios calculated for all the samples. Calculation of this ratio allowed for the determination of the dominant weathering process: solely carbonate weathering or a combination of carbonate weathering and sulfide oxidation. This ratio is calculated from dividing the HCO_3 by the combined value of the $HCO_3 + SO_4^2$ ions within the samples. This ratio, if closer to one, suggests carbonate dissolution as the singularly prominent weathering process occurring, but if the ratio is 0.5, the dominant processes are carbonate dissolution and oxidation of sulfide minerals (Williams et al. 2006).

Table 5.2: C-Ratio between HCO_3^- and $HCO_3^- + SO_4^{2-}$ in Icelandic Glacial-Fed Rivers and the Alkalinity to DIC ratio. A value of 1.0 for a C-ratio indicates the dominance of carbonate dissolution while a value of 0.5 suggests the dual influence of carbonate dissolution and sulfide oxidation on the carbon concentration of the waters (Source: created by Author).



C-Ratio and Alk:DIC Ratio in Icelandic Glacial-Fed Rivers

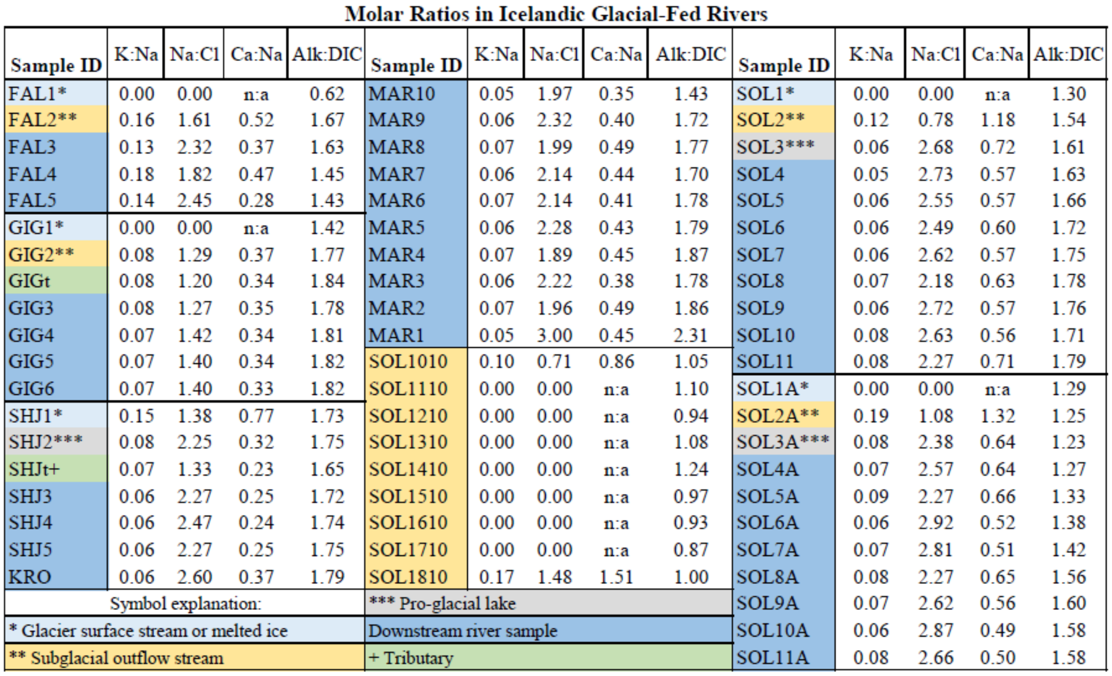
Sample	C-Ratio	Alk/DIC	Sample	C-Ratio	Alk/DIC	Sample	C-Ratio	Alk/DIC
FAL1*	0.86	0.62	MAR10	0.76	1.43	SOL1*	0.57	1.30
FAL2**	0.81	1.67	MAR9	0.65	1.72	SOL2**	0.69	1.54
FAL3	0.74	1.63	MAR8	0.64	1.77	SOL3***	0.68	1.61
FAL4	0.75	1.45	MAR7	0.64	1.70	SOL4	0.81	1.63
FAL5	0.75	1.43	MAR6	0.63	1.78	SOL5	0.80	1.66
GIG1*	0.65	1.42	MAR5	0.63	1.79	SOL6	0.80	1.72
GIG2**	0.68	1.77	MAR4	0.65	1.87	SOL7	0.79	1.75
GIGt +	0.68	1.84	MAR3	0.64	1.78	SOL8	0.80	1.78
GIG3	0.68	1.78	MAR2	0.62	1.86	SOL9	0.81	1.76
GIG4	0.66	1.81	MAR1	0.63	2.31	SOL10	0.81	1.71
GIG5	0.66	1.82	SOL1010	0.68	1.05	SOL11	0.81	1.79
GIG6	0.66	1.82	SOL1110	0.70	1.10	SOL1A*	0.61	1.29
SHJ1*	0.82	1.73	SOL1210	0.65	0.94	SOL2A**	0.77	1.25
SHJ2***	0.85	1.75	SOL1310	0.71	1.08	SOL3A***	0.82	1.23
SHJt +	0.81	1.65	SOL1410	0.66	1.24	SOL4A	0.81	1.27
SHJ3	0.82	1.72	SOL1510	0.71	0.97	SOL5A	0.81	1.33
SHJ4	0.80	1.74	SOL1610	0.65	0.93	SOL6A	0.80	1.38
SHJ5	0.76	1.75	SOL1710	0.73	0.87	SOL7A	0.80	1.42
KRO	0.73	1.79	SOL1810	0.65	1.00	SOL8A	0.80	1.56
* Glacier surface stream or melted ice			Downstream river sample			SOL9A	0.80	1.60
** Subglacial outflow stream			+ Tributary			SOL10A	0.80	1.58
*** Pro-glacial lake						SOL11A	0.80	1.58

As shown in Table 5.2, the C-ratios for the dataset varied; however, as indicated in the ratios, the dominant weathering processes occurring is a combination of carbonate weathering and sulfide oxidation. Several studies support these findings (Jacobson et al. 2015; Sveinbjörnsdóttir et al. 2005; Sveinbjörnsdóttir et al. 2010; Burns 2016).

5.4.3 Other Ionic Ratios

Table 5.3 displays other elemental ratios, which shed important insight into the geochemical processes occurring in-stream. In glacial environments, elevated SO_4:Na ratios indicate relatively more sulfide mineral oxidation than silicate weathering and elevated Ca:Na ratios indicate more carbonate than silicate weathering (Raiswell 1984; Tranter et al. 1993; Tranter 2003; Torres et al. 2017). The Ca:Na ratios of this study agree with the results from a previous study (Dessert et al. 2003) that determined the Ca:Na ratios in Icelandic basalt-draining rivers fall between 0.9 and 0.45. Elevated Ca and Mg concentrations, which drive the SO_4:Na and Ca:Na ratios higher, are explained by the weathering of Ca-bearing silicate rocks, such as basalt (Torres et al. 2017).

Table 5.3: Molar ratios across all major sample sites (Source: created by Author).



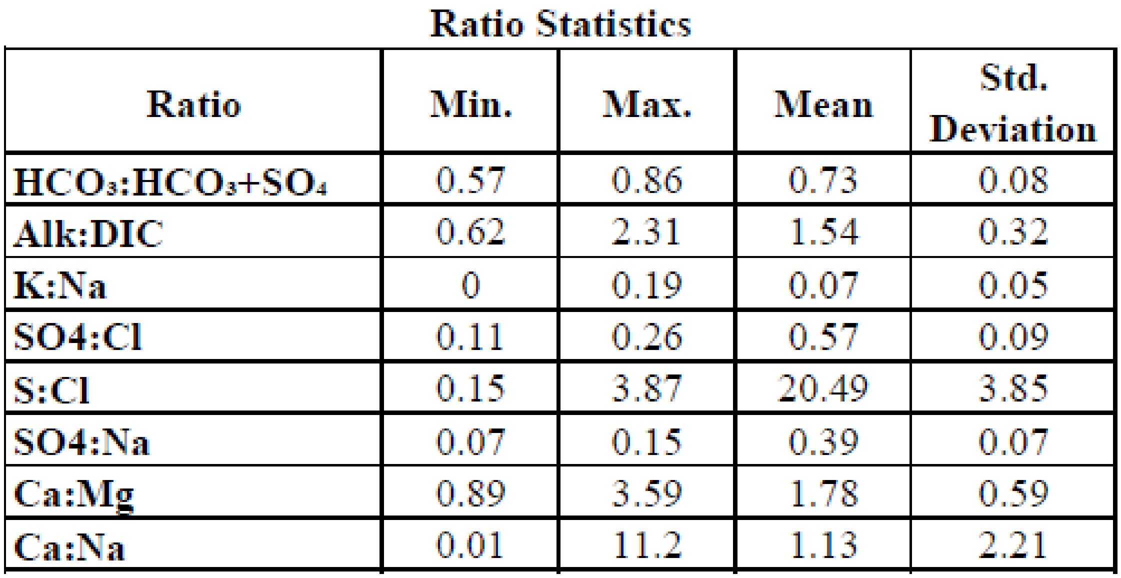
Molar Ratios in Icelandic Glacial-Fed Rivers

Sample ID	K:Na	Na:Cl	Ca:Na	Alk:DIC	Sample ID	K:Na	Na:Cl	Ca:Na	Alk:DIC	Sample ID	K:Na	Na:Cl	Ca:Na	Alk:DIC
FAL1*	0.00	0.00	n:a	0.62	MAR10	0.05	1.97	0.35	1.43	SOL1*	0.00	0.00	n:a	1.30
FAL2**	0.16	1.61	0.52	1.67	MAR9	0.06	2.32	0.40	1.72	SOL2**	0.12	0.78	1.18	1.54
FAL3	0.13	2.32	0.37	1.63	MAR8	0.07	1.99	0.49	1.77	SOL3***	0.06	2.68	0.72	1.61
FAL4	0.18	1.82	0.47	1.45	MAR7	0.06	2.14	0.44	1.70	SOL4	0.05	2.73	0.57	1.63
FAL5	0.14	2.45	0.28	1.43	MAR6	0.07	2.14	0.41	1.78	SOL5	0.06	2.55	0.57	1.66
GIG1*	0.00	0.00	n:a	1.42	MAR5	0.06	2.28	0.43	1.79	SOL6	0.06	2.49	0.60	1.72
GIG2**	0.08	1.29	0.37	1.77	MAR4	0.07	1.89	0.45	1.87	SOL7	0.06	2.62	0.57	1.75
GIGt	0.08	1.20	0.34	1.84	MAR3	0.06	2.22	0.38	1.78	SOL8	0.07	2.18	0.63	1.78
GIG3	0.08	1.27	0.35	1.78	MAR2	0.07	1.96	0.49	1.86	SOL9	0.06	2.72	0.57	1.76
GIG4	0.07	1.42	0.34	1.81	MAR1	0.05	3.00	0.45	2.31	SOL10	0.08	2.63	0.56	1.71
GIG5	0.07	1.40	0.34	1.82	SOL1010	0.10	0.71	0.86	1.05	SOL11	0.08	2.27	0.71	1.79
GIG6	0.07	1.40	0.33	1.82	SOL1110	0.00	0.00	n:a	1.10	SOL1A*	0.00	0.00	n:a	1.29
SHJ1*	0.15	1.38	0.77	1.73	SOL1210	0.00	0.00	n:a	0.94	SOL2A**	0.19	1.08	1.32	1.25
SHJ2***	0.08	2.25	0.32	1.75	SOL1310	0.00	0.00	n:a	1.08	SOL3A***	0.08	2.38	0.64	1.23
SHJt+	0.07	1.33	0.23	1.65	SOL1410	0.00	0.00	n:a	1.24	SOL4A	0.07	2.57	0.64	1.27
SHJ3	0.06	2.27	0.25	1.72	SOL1510	0.00	0.00	n:a	0.97	SOL5A	0.09	2.27	0.66	1.33
SHJ4	0.06	2.47	0.24	1.74	SOL1610	0.00	0.00	n:a	0.93	SOL6A	0.06	2.92	0.52	1.38
SHJ5	0.06	2.27	0.25	1.75	SOL1710	0.00	0.00	n:a	0.87	SOL7A	0.07	2.81	0.51	1.42
KRO	0.06	2.60	0.37	1.79	SOL1810	0.17	1.48	1.51	1.00	SOL8A	0.08	2.27	0.65	1.56
Symbol explanation:					*** Pro-glacial lake					SOL9A	0.07	2.62	0.56	1.60
* Glacier surface stream or melted ice					Downstream river sample					SOL10A	0.06	2.87	0.49	1.58
** Subglacial outflow stream					+ Tributary					SOL11A	0.08	2.66	0.50	1.58

The ratios between the dissolved K:Na ions represent the process of silicate weathering in the system and are shifted higher in glaciated terrains in comparison to the global average (Torres et al. 2017). The K:Na ratios from this study are low and support the previous determination that silicate weathering is not a dominant source of ions to the study rivers at the timescale represented in this study, but may be over longer timescales.

Previous studies have determined the S:Cl ratio from Icelandic basalts to be fixed at ~1.65 and ~5.0 for SO_4^{2}:Cl (Arnórsson and Andrésdóttir 1995); the SO_4^{2-}:Cl ratio of seawater is fixed at 0.14 (Kroopnic 1977). While leaching will occur when water is in contact with rock, the expected contribution of chloride to water from rock leaching is negligible in comparison with other sources of chloride (Arnórsson and Andrésdóttir 1995), and, thus, increases in the SO_4^{2} concentration, and the SO_4^{2}:Cl ratio, are more likely indicative of sulfide mineral oxidation, atmospheric deposition, or evaporite mineral dissolution (Torres et al. 2017). The ranges of SO_4^{2-}:Cl, S:Cl, and SO_4^{2}:Na are indicated in Table 5.4. Ranges of the SO_4^{2}:Cl ratio from these data do not indicate a strong silicate weathering influence on the SO_4^{2} concentration, though there is obvious deviation from the fixed seawater ratio of 0.14, suggesting there is some other influence on the SO_4^{2} concentration.

Table 5.4: Min., max., mean, and standard deviation of the various ratios used in the discussion (Source: created by Author).



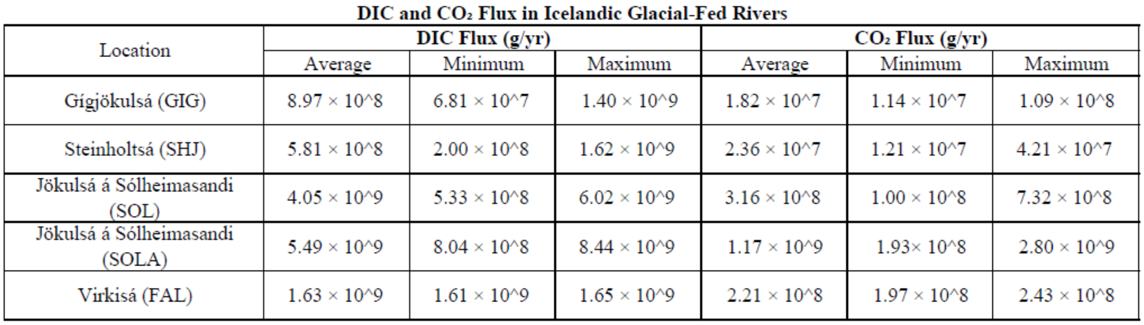
Ratio Statistics

Ratio	Min.	Max.	Mean	Std. Deviation
$HCO_3:HCO_3+SO_4$	0.57	0.86	0.73	0.08
Alk:DIC	0.62	2.31	1.54	0.32
K:Na	0	0.19	0.07	0.05
SO4:Cl	0.11	0.26	0.57	0.09
S:Cl	0.15	3.87	20.49	3.85
SO4:Na	0.07	0.15	0.39	0.07
Ca:Mg	0.89	3.59	1.78	0.59
Ca:Na	0.01	11.2	1.13	2.21

5.5 DIC and CO_2 Flux

The dissolved CO_2 concentrations in the studied rivers decrease downstream at all sites, which agrees with the decreasing trend in $EpCO_2$. Thus, all the studied rivers are determined to be acting as source of CO_2 to the atmosphere and contributing to a positive feedback of global warming. The DIC concentrations display more variation in concentration downstream, suggesting that CO_2 drawdown from the atmosphere is an influence on the DIC flux, but it is not the sole source of carbon contributing to the carbon dynamics within the system. An important factor to address in this discussion is that of the volcanic influence on the carbon flux. Volcanic environments are noted sources of CO_2 to the global carbon cycle. In comparison, glaciated terrains are typically cited as acting as CO_2 sinks. This interaction in Iceland is leads to a complicated and variable carbon flux system within the rivers.

Table 5.5: Min., max., and average of the flux calculations of DIC and pCO_2. Average was calculated by dividing the sum of the flux results from all the samples by the number of samples for each site. Not included in the table is the flux from the surface stream DIC and pCO_2^- concentrations (Source: created by Author).



DIC and CO_2 Flux in Icelandic Glacial-Fed Rivers

Location	DIC Flux (g/yr)			CO_2 Flux (g/yr)		
	Average	Minimum	Maximum	Average	Minimum	Maximum
Gígjökulsá (GIG)	8.97 × 10^8	6.81 × 10^7	1.40 × 10^9	1.82 × 10^7	1.14 × 10^7	1.09 × 10^8
Steinholtsá (SHJ)	5.81 × 10^8	2.00 × 10^8	1.62 × 10^9	2.36 × 10^7	1.21 × 10^7	4.21 × 10^7
Jökulsá á Sólheimasandi (SOL)	4.05 × 10^9	5.33 × 10^8	6.02 × 10^9	3.16 × 10^8	1.00 × 10^8	7.32 × 10^8
Jökulsá á Sólheimasandi (SOLA)	5.49 × 10^9	8.04 × 10^8	8.44 × 10^9	1.17 × 10^9	1.93× 10^8	2.80 × 10^9
Virkisá (FAL)	1.63 × 10^9	1.61 × 10^9	1.65 × 10^9	2.21 × 10^8	1.97 × 10^8	2.43 × 10^8

The largest range and variation in DIC flux was calculated from Sólheimajökull and Falljökull. Despite the presence of fresh, volatile volcanic material at Gígjökull, other influences, such as watershed size and runoff, are clearly more dominant influences on carbon flux. The DIC flux at Gígjökull and Falljökull varies little downstream, unlike both Sólheimajökull datasets, which have measurable variation downstream. In general, the CO_2 flux varies little across all the sample sites, in comparison to the DIC flux. The $EpCO_2$ values suggest that the dissolved CO_2 in the river is not at equilibrium with the atmosphere; in fact, the $EpCO_2$ values suggest that degassing would occur frequently within the studied rivers, which may relate to their origin, short reaches, and turbulence. The CO_2 flux from GIG varies little between the minimum and maximum calculated value. Both DIC and CO_2 fluxes increase downstream at Steinsholtsjökull and Sólheimajökull. Falljökull has the lowest DIC flux and least amount of variation in the CO_2 flux. Estimates in the 1990s suggested that the entire Arctic region was acting as a modest CO_2 sink, drawing between 0 and 0.8 Pg C/yr (McGuire et al. 2009); however, further refinements of that estimate more accurately suggest that value is closer to 0.8 Pg C/yr (Baker et al. 2006). Results from this study indicate the studied rivers are acting as sources of CO_2, with an estimated average flux of CO_2 of 4.44×10^8 g/yr.

The CO_2 flux data from SOL and SOLA vary somewhat; however, it is important to note the same discharge value, 18.5 m^3/s, was used to calculate DIC and CO_2 flux for both datasets. This discharge value was measured on June 6th, 2017, three days before SOL sampling occurred and five days before SOLA sampling occurred. The discharge value taken for this site, therefore, is considered a conservative, baseline for the site and differences in the CO_2 flux values may relate to changes in meltwater inputs and temperature variability.

There was no major difference in rainfall between during the sampling period; however, there was a measurable difference in surface temperature between the two sampling days. The temperature range on the day of sampling of the SOL samples was -

0.2 °C to 13 °C. On the day of the SOLA sampling, the temperature ranged from 1.2 °C to 16.5 °C. The average temperature on the SOLA sampling day was approximately 3 °C higher than the day on which SOL was sampled. Since surface temperature directly affects glacier surface melt, the increased temperatures on the 11th, when SOLA was sampled, would influence the river by increasing meltwater runoff, in-stream weathering processes, and volume of water transported. These data agree with previous determinations that increases in surface temperature have a major influence on downstream mechanics and is the likely explanation of the difference between the DIC and CO_2 flux between SOL and SOLA.

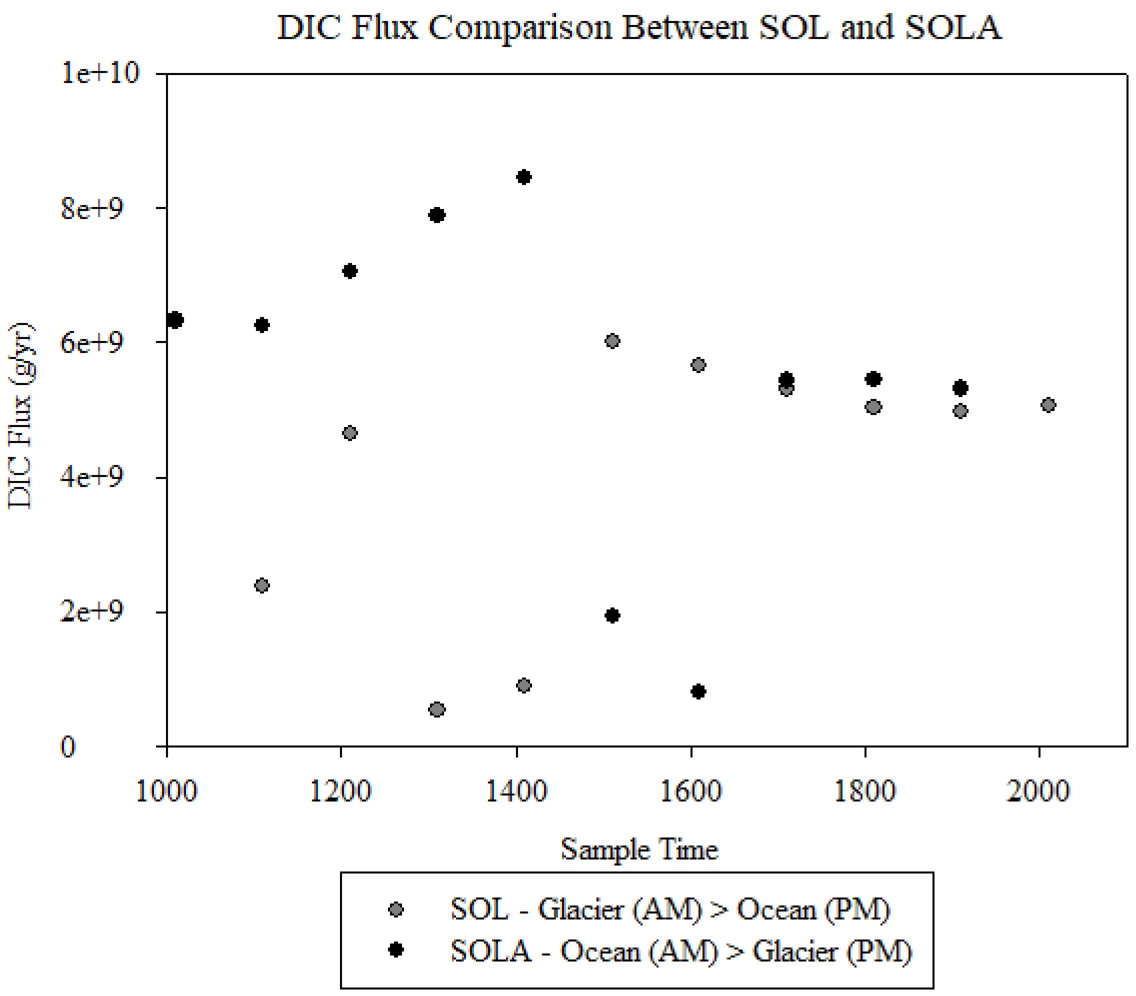


Figure 5.10: DIC comparison between the two Sólheimajökull datasets: SOL and SOLA. SOL was sampled starting in the morning, from glacier to ocean. SOLA was sampled 48 hours later beginning at the ocean in the morning and ending at the glacier in the evening (Source: created by Author).

There was measurable variation between the two Sólheimajökull datasets, SOL and SOLA, as seen in Figure 5.10. Figure 5.10 shows a different trend in the flux data between the two datasets. The upstream samples from SOL show great variability, but stabilize later in the day and downstream past the major tributary inputs. The first samples in the SOLA dataset display a clear increasing trend that decreases mid-afternoon and then, once again, stabilizes as the day progresses. These trends suggest that discharge cycles from atmospheric temperature changes are influencing the system and changes within the river. This agrees with field observations of lagging, surge-like pulses observed from streams near the terminus of the glacier.

The decrease in DIC and EpCO_2 downstream indicates the rivers are losing DIC and CO_2 downstream. The decrease in EpCO_2 downstream is an indication that the pCO2 of the water is decreasing downstream, with respect to the pCO_2 of the atmosphere, which stays constant. Most of the DIC in the rivers is in the form of HCO_3, a weathering product of carbonate minerals. CO_2 is used in that weathering process and can also decease in concentration by degassing to the atmosphere. The lack of variability in ions indicative of weathering, like Ca and Fe, indicate that weathering processes are likely not the only responsible mechanism for decreases in CO_2 concentration. The short distance of these rivers is a reason the dissolution of carbonate minerals is somewhat limited and competing with CO_2 evasion from the rivers. The short distance and discrete sampling also limits the ability to detect silicate weathering on the rivers, which often occurs over longer timescales. The short distance of these rivers translates directly to rapid transport of water from the glaciers to the oceans, so processes that are more dominant over longer timescales are not as obvious; however, this will likely change in Iceland as glaciers continue to melt worldwide. As sea level rises, glacial lagoons will continue to develop and progress closer towards the ocean. This will exacerbate the carbon flux dynamics already occurring in these rivers and proliferate the tendency of these rivers to act as sources of carbon.

CHAPTER SIX: CONCLUSIONS

Studying the source, transformation, and transport of carbon in glacier-fed riverine systems is integral to understanding dynamics of the carbon cycle within specific regions and environments. The objective to understand those dynamic relationships was, in part, from the results of a currently unpublished thesis (Tuladhar 2017), which focused on providing a robust hydrogeochemical characterization of many of the same study sites included in this study. That research suggests the geochemistry of the rivers is dynamic and changes downstream. Longitudinal investigations on these rivers were lacking, particularly in regard to carbon flux. The uniqueness of the Icelandic environment is pivotal in establishing an understanding of complicated interactions between the geology, hydrology, and atmosphere with respect to global warming and climate change.

Chemical weathering of carbonate minerals is the most dominant in-stream process occurring that influences the hydrogeochemistry of the rivers; sulfide oxidation and silicate weathering influences are indicated by the data, but in lower magnitudes. The potential introduction of subglacial meltwater, volcanic gas, and geothermal fluids to the rivers all could contribute excess CO_2 that ultimately oversaturates the water and counteracts the effect of rock weathering that would consume it entirely. Chemical weathering rates in Iceland are higher and more variable than the global average, due to the dominant presence of basalt, higher rates of mechanical weathering, and runoff. Despite this, studies have determined that the contribution of basalt weathering to atmospheric CO_2 consumption may be overestimated, and thus, less significant than previous estimations (Jacobson et al. 2015). The DIC concentrations and speciation of DIC within the rivers of interest in this study indicate the dominance of the chemical weathering of carbonate minerals from basalt within the system. Chemical weathering of Ca-Mg-rich silicates has been determined to be a primary method of removing CO_2 from the atmosphere (Dessert et al. 2003; Khadka et al. 2014; Jacobson et al. 2015); this trend is typically more obvious on longer, geologic timescales. Results from this study indicate the marginal influence chemical weathering of Ca-Mg has on carbon flux in relatively short-reach rivers and over short timescales in Icelandic glacial rivers. All the glaciers included in this study are non-surging, temperate glaciers and considered to be in a state of retreat. The ultimate disappearance of these glaciers will likely happen in the next 100 years and could contribute measurably to global carbon flux calculations and should, therefore, be included in them.

The DIC and $EpCO_2$ results indicate slight longitudinal trends in various carbon cycle associated parameters. DIC concentrations were characteristically different at the separate sample sites. At Sólheimajökull, during both sampling days, the DIC concentration slightly decreased downstream. The $EpCO_2$ values also decreased downstream. HCO_3 was determined to be the most dominant form of DIC present in the study rivers at the time of sampling and, thus, suggests that CO_2 is being removed from the atmosphere to drive carbonate mineral dissolution; however, it is also likely the CO_2 generated from weathering is contributing to the oversaturation of CO_2 in the river water and contributing to CO_2 degassing downstream, while other sources of unmeasured CO_2, such as geothermal fluids, may also be a contributor. The Alk:DIC ratios indicate that most of the samples from the study were acting as CO_2 sources to the atmosphere on the timescale of carbonate burial in the ocean. This fact is represented in the data, which display little changes and variation downstream, albeit with an overall decrease in CO_2.

Downstream trends did not display major variations; despite this, changes in geochemistry and carbon dynamics were still observed. The results from this study do not indicate which environmental influence is the most significant in the carbon flux of glaciated terrains. The data do, however, suggest that glacier systems, with regard to carbon flux, are somewhat homogenous when discussing carbon flux on longer time scales. Even through discreet, single-occurrence sampling the geochemistry of the rivers trend similarly. Longitudinal trends suggest that carbonate weathering and sulfide oxidation are a dominant source of DIC within these systems. The multiple inputs of CO_2 to the rivers suppress the sink effect of the carbonate weathering. Ultimately, these systems are presently working as positive feedback with respect to changes in global climate via global warming enhanced by glacial meltwater-derived river systems in Iceland acting as sources of CO_2 to the atmosphere.

DIC in the form of H_2CO_3, representing solvated CO_2 and carbonic acid, was present in 30 samples above 10%. The majority of those samples were glacier surface stream samples; however, pro-glacial lake samples and relatively upstream river samples were included. In terms of species portioning, % HCO_3 increased, longitudinally, at all sites, except for Falljökull and in the temporal Sólheimajökull dataset. DIC flux results from this study are lower than the global average of DIC contribution to riverine carbon flux; the highest DIC flux calculated from this study was 8.44×10^9 g/yr; the highest CO_2 flux was 2.80×10^9 g/yr. DIC and pCO_2 data from glacier surface samples were excluded from flux calculation, since DIC represents an insignificant fraction of the carbon present on the surface of a glacier (Stibal et al. 2008).

The results from this study suggest that multiple forms of chemical weathering are present and active contributors to the geochemistry of the waters: carbonate mineral dissolution, silicate weathering, and sulfide oxidation. HCO_3 was the dominant form of DIC present in the waters, which is primarily sourced from carbonate mineral weathering. The CO_2 used in those reactions was identified as a dissolved CO_2 source within the water and atmospheric exchange. These processes are responsible for most of the flux of CO_2 in the waters. The concentration of CO_2 decreases downstream, indicating that CO_2 is being utilized in carbonate mineral weathering and also degassing. This indicates the studied rivers undergo complex processes, utilizing, transporting, and evading carbon. The role of the studied rivers to act as carbon sources suggests these rivers, at least during the season sampled in this study, are acting in a positive feedback loop with respect to global warming. As concentrations of CO_2 in the atmosphere continue to increase, the global air temperature will similarly increase and, subsequently, exacerbate the continued retreat of glaciers in Iceland and elsewhere.

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