An investigation into sources and distributions of iron, aluminum, and nutrients in Glacier Bay, Alaska

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Abstract

Glacier Bay is glacially fed fjord in southeastern Alaska where observations of nutrients, salinity and temperatures suggest continuous mixing due to rapid circulation that contributes to a large supply of iron into coastal waters. Iron concentrations of up to 56 µmol l^{-1} for iron in the surface waters of the bay were measured. Using iron to aluminum ratios, glacial ice and rivers were determined not to be a notable source and the continental shelf itself was implicated as the primary source for iron. The potential availability of this iron to biology in the open ocean is likely via eddy spin off and shelf advection. A horizontal gradient was extrapolated from previous studies and further suggested the continental shelf as a major source for iron in the open ocean.

Introduction

The Gulf of Alaska in the northeastern area of the subarctic North Pacific is known to be a High-Nutrient-Low-Chlorophyll (HNLC) zone, where iron is the major biologically limiting micronutrient for primary production (Martin and Gordon, 1988; Martin et al., 1989; Boyd et al., 2004). The continental margin is also a source for iron in the HNLC areas of the North Pacific (Lam and Bishop, 2008) and can seed eddies which transport iron into the open ocean (Johnson et al. 2005). Point sources for iron in the surface waters are an important aspect of the geochemical cycling which has great influence on both the concentrations and availability for biology. Understanding the concentrations and contributions of these different sources is necessary to understand how the coastal waters and HNLC zones are influenced by iron, and will also assist in future studies including iron fertilization experiments.

Iron plays a key role for the assimilation of nutrients by marine phytoplankton (Turner and Hunter 2001, Boyd et al. 2004, Johnson et al. 2005). There are three primary sources that exist for iron transport to phytoplankton: atmospheric deposition, upwelling and advection from the continental margin. It has been generally believed that of the three sources for iron in HNLC areas of the ocean, atmospheric deposition is the primary contributor (Bishop et al. 2002). However, sources for iron in Glacier Bay, Alaska were explored and suggest that the continental shelf plays large role in generating iron for possible transport to the open ocean. Additionally, much of this iron is available for transport to the open ocean via eddies that form in the winter months.

Glacier Bay, Alaska is a fjord system recently deglaciated within the past 300 years. It is located on the southeast corner of Alaska where high saline water of deep origin intrudes

over a sill into the bay, and freshwater runoff including that from melting glacial ice flows out of the bay on the surface (Mathews 1981). The estuarine circulation has possible unique and distinctive characteristics including great degrees of mixing where it has been suggested that nutrient availability is highly abundant and not biologically limiting even during the spring or summer months when large sustained phytoplankton blooms occur (Hooge and Hooge, 2002). Additionally, the sources of iron and its distribution in and outside of the bay is relatively unknown and was explored in this study to better understand the possibility of the continental margin as a possible source for iron in a High Nutrient Low Chlorophyll (HNLC) area as in the Gulf of Alaska.

The goals of this study were to: (1) determine the sources for and distributions of iron and aluminum in Glacier Bay (2) use the iron and aluminum data from the lithogenic crust as a tracer and determine how much iron is in detrital versus sorbed metal oxide phases and which point sources in the bay are the primary iron source contributors. (3) Tracking the distribution of iron through the bay will establish the amount of iron that is potentially exported into coastal waters, and by determining correlations with nutrients and salinity, the pathway of iron can be further explored.

The implications of this work extend into the influences of global warming and have a direct impact on the carbon cycle. Iron is the 4th most abundant element in the earth's crust, yet is limiting as a micronutrient for biology in a vast majority of the worlds' oceans (Turner 2001). Determining the point sources for biologically available iron is essential to understanding the biogeochemical cycles which control primary production and the consequent drawdown of atmospheric CO_2. A greater understanding of the sources and relationships of iron with salinity, POC, and nutrients will assist in understanding a wide range of areas from ocean acidification and general global management to influencing important decisions such as management of coastal environments, and glaciers.

Materials and Methods

Sampling

Surface waters were sampled at several locations in and outside the mouth of Glacier Bay at 14 locations between 18 March – 21 March 2008 (Fig. 1) using trace-metal clean techniques. At these stations, a Kevlar line mounted with thirty liter Go-Flo sampling bottles was utilized to sample water from the surface (2 meters) as well as below the mixed layer. The surface layer depth was determined by salinity and temperature gradients gathered from the cast of a rosette mounted with a Seabird model SBE 911 CTD where vertical distributions of salinity, temperature, as well as dissolved oxygen and chlorophyll were measured. A weight at the bottom of the Kevlar line was wrapped in plastic and the metal shackle was enveloped in a latex glove. Brass messengers were used to close the bottles and water samples were then racked on a separate stationary CTD rosette.

Go-Flo bottles were pressure filtered at approximately 12 psi passing sample water through a 0.8 µm Supor filter membrane. The filters were mounted in a polyethylene filter holder with Teflon screws and an inline filter was installed with glass wool between the pump and the Go-Flo bottles to prevent contamination. To finalize the preparation of the sample, pressure filtration was changed to a vacuum filtration system and a small amount of deionized water was passed through the filter to remove residual salt. The final volume filtered was measured and the filter was then folded in quarters and placed in a clean plastic bag using Teflon tweezers.

During filtration, samples from inline on the filtration apparatus were collected for total as well as dissolved iron and aluminum in 10% HCL acid baked at 60° C 60ml Nalgene high density polyethylene (HDPE) clear bottles. Bottles

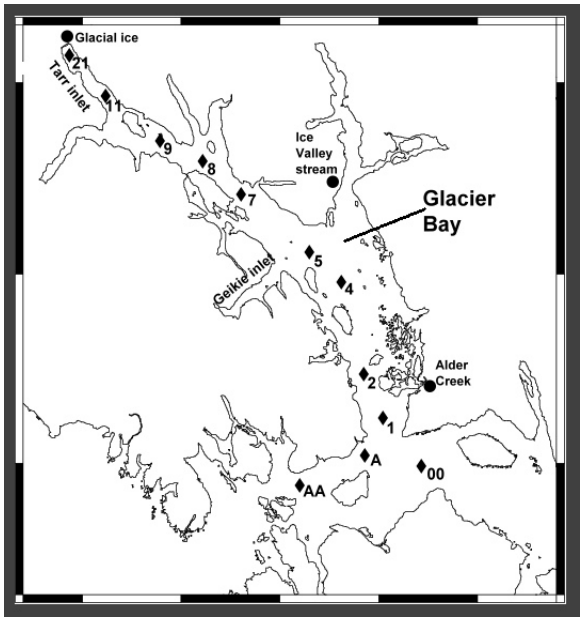


Figure 1: Map of study area in Glacier Bay on Ocean 443 cruise during 18 March – 21 March 2008. Circles indicate locations of river and ice sampling, diamonds denote Go-Flo casts.

were then acidified to a pH < 2 using trace metal clean HCL. Nutrient samples were taken concurrently and analyzed on the ship as well as a duplicate analysis at the UW Chemical Analysis Laboratory in the School of Oceanography. Additionally, 2 liters of seawater were removed from the Go-Flo bottles for Particulate Organic Carbon (POC) analysis on a standard GFF filtration rack using 0.7 micron GFF filters. Filters were then individually wrapped in aluminum foil and stored cold to be later analyzed and catalogued at the University.

Two river samples were collected using HCl acid cleaned 20 liter cubitainers well above tidal water line. River water samples were vacuum filtered directly out of the cubitainers and final volumes were recorded.

Ice samples were collected out of the seawater directly at the face of the glacier. Ice was then separated according to transparency: clear, white, and mixed. All glacial ice was first thoroughly rinsed in deionized water and then allowed to melt in acid cleaned cubitainers. To remove larger particulate matter, ice water was then passed through a 100 micron filter into rinsed Go-Flo bottles for pressure filtration as performed with seawater samples.

Contamination Management

To assess for potential contamination, filter blanks were left out in the open working environment for time periods of up to 12 hours and DDZ water was run through the filtration process as well to quantify and determine possible sources of contamination. All filter blanks were below the detection limits for both iron and aluminum. Detection limits for the samples were

Nutrient Analysis

Phosphate, nitrate and ammonia were analyzed on the cruise. Ammonia was analyzed by measuring fluorescence to treated samples outlined by Holmes et al. (1999). Nitrate was measured on a single channel AA II using methods by K. Krogslund (personal communication, 2008), and phosphate was measured spectrophotometrically in a reaction using ammonium molybdate and ascorbic acid according to methods summarized by Grasshoff et al. (1983)

Iron and Aluminum Analysis

Digestion of Filters

Filters were dried in an oven overnight at 50° C, weighed, and placed in a desiccator. Samples were digested in sets of 11 with a reagent blank for each run. All samples were digested according to the method of Murray and Leinen (1993), modified by Nameroff (1996), and further described by Yigiterhan (2005). Each sample was digested twice in trace metal cleaned high pressure Teflon vessels bathed in an acid cocktail solution of 4 ml 23 M HF, 1 ml 16 M HNO_3, and 1 ml 6 M HCl for the first digestion and 4 ml HNO_3 for the second digestion. Preparation of samples was conducted in a dust-free laminar

fume hood for both safety and to prevent contamination.

Two separate digestions were both conducted under a hood in a 900W commercial microwave oven for 2 minutes at 100% followed by 60 minutes on 10% power, reduced, and returned to the microwave for 2 minutes at 100% followed by 30 minutes on 10% power. The Teflon bombs were placed in a round plastic container and a sheet of plastic was sealed between the lid and the body to contain any escaping acid vapor. Additionally a plexiglass shield was pulled over the oven and the room was vacated in case of bomb explosion. After initial digestion, samples were cooled overnight, transferred into 50 ml Teflon beakers, and heated on a hot plate in a trace metal free laminar fume hood to near dryness. Filters were then transferred back into their respective bombs and 4 ml of 16 M HNO_3 was added to the beakers and poured into the bombs for the second digestion. Following the final digestion, samples were dried again to near dryness, residual filter material was washed with 5 ml 1% HNO_3 and discarded. An additional 5 ml of 1% HNO_3 was added to the sample solution and clear liquid was then transferred into clean acid baked 30 ml HDPE Nalgene bottles for analysis.

Digestion of sediment samples

Sediment samples were first oven dried in clean Teflon containers. A sample of approximately 0.075 grams was removed and bathed in a cocktail of 5ml 23 M HF and 1 ml HNO_3. Samples were then treated in the same way as filter samples. After digestion, samples were centrifuged to separate any undissolved particulate matter and analyzed.

Atomic Absorption Spectrophotometry

Atomic Absorption Spectrophotometry (AAS) is a very sensitive technique for analyzing trace metals. Samples are dried and then heated into a cloud of gasses where hollow cathode lamps of a specific wavelength are absorbed by the element of interest and concentrations of that element can then be determined. Most samples were concentrated enough to run on the open flame of a Perkin Elmer ELAN 5000 AAS, however some of those with lower concentration levels had to be run in an automatic Hitachi Z-9000 AAS graphite furnace.

Dissolved and Total trace metal samples

The collected dissolved and total iron samples collected in the 60 ml HDPE Nalgene bottles were used as an accuracy measurement for the filter concentrations. They were collected during filtration directly from the Go-Flo bottles and directly out of the filter. They were collected as a measure for accuracy of the particulate iron and aluminum measurements where the dissolved + particulate should equal the total. Samples were acidified to a pH <2 and catalogued.

Results

Iron and Aluminum

Both iron and aluminum were found in notably high concentrations in the surface waters of the bay ranging from 4 µmol l^{-1} to over 50 µmol l^{-1} for iron, and less than 1 µmol l^{-1} to 28 µmol l^{-1} for aluminum. The concentrations varied between stations, however the ratio of iron to aluminum between stations was notably more stable (Table 1). There was no linear gradient through the length of the bay for iron, however there were elevated concentrations of over 30 µmol l^{-1} primarily near the sill at the mouth of the bay. Concentrations of iron and aluminum were often higher at depth than in the surface waters, which agrees with past measurements made away from the continental shelf (Johnson et al. 2005, Lam et al. 2006). Concentrations in glacial ice were generally very high for both iron and aluminum with over 357 µmol l^{-1} for iron, and up to 650 µmol l^{-1} for aluminum.

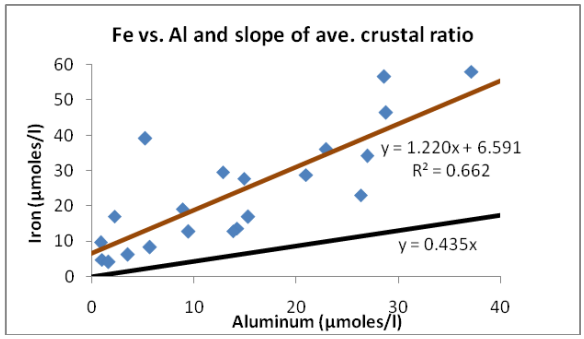


Figure 2: Iron and aluminum concentrations plotted from all stations in Glacier Bay. Dark line shows average global crust ratio for iron and aluminum.

The primary river samples of Ice Valley river which feeds into the East Arm of Glacier Bay reflected a slightly higher than average global crust value for iron. Although the Bartlett Cove Alder Creek samples had average iron values comparable to the other areas of the bay, they were both extremely low in aluminum with less than 0.8 µmol l^{-1}.

Ratios for iron to aluminum maintained a clear correlation throughout the bay (Fig 2) with a higher than average concentration of iron than the average global crust value. The Ice Valley River Fe:Al ratios were very close to the average global river values.

Nutrients

Nutrient data was relatively constant through the bay. Concentrations of Total Nitrogen ($NH_4 + NO_3 + NO_2$) to phosphorus ratios were consistently around 12.4 : 1 for both surface uptake and remineralization (Fig. 3). Ammonia concentrations were notably higher in older clear ice samples as well as the glacially fed river where levels as high as 3 µmol l^{-1} were observed. There was no correlation of iron with salinity or nutrients anywhere in the bay (Fig 4). It was also found that silica concentrations were relatively constant with the exceptions of Ice Valley River and outside the bay.

Discussion

Biogeochemical iron sources

Aluminum is a good tracer for iron in that it functions almost identically as iron and shares the same terrestrial sources, however it does not share the interactions with biology. There is an excess of iron relative to aluminum compared with the average global values in all samples (Fig. 2). It could be assumed that this excess iron is coming from either the ice input or the rivers, which both had high concentrations of iron and aluminum, however the resulting ratio of Fe:Al did not match that in the bay. Additionally, the low fresh water input for winter suggested another source: the sediments. Multiplying the total aluminum in an individual sample by the Fe:Al ratio of the river gives the total iron in the water sample in the bay, and subtracting that from the total concentrations of iron illustrates the percent iron not coming from the river source (equation 1).

$$[\mathrm{Al}]_{\mathrm{sw}} \left(\frac{\mathrm{Fe}}{\mathrm{Al}}\right)_{\mathrm{river}} = [\mathrm{Fe}]_{\mathrm{river}} \qquad (1)$$

$$[\mathrm{Fe}]_{\mathrm{sw}} - [\mathrm{Fe}]_{\mathrm{river}} = [\mathrm{Fe}]_{\mathrm{biogeochemical}} \qquad (2)$$

Using relationship of iron and aluminum in equation 1, biogeochemical iron is calculated in the seawater in equation 2.

Ice Valley River particulate material had an iron to aluminum ratio of 0.576 which is quite close to the global river values of 0.51 with higher values of iron being responsible for the difference (Table 1). Using the iron to aluminum ratio in the Ice Valley River as a standard value, the percent of iron coming from other non-lithogenic sources was calculated (Table 2), and ranged from 40-90%. Water in the bay below the surface mixed layer generally showed higher values for iron than the surface water (Table 3) and further suggests the sediments as a primary source for the excess iron in Glacier Bay.

Location	Iron:Aluminum	Reference
	Global	
River/Creek	0.511	Taylor & McLennan (1995)
Crust	0.435	Taylor & McLennan (1995)
Shale	0.548	Wedepohl (1995)
	Glacier Bay	
Ice valley river	0.576	This study
Glacier Ice	0.694	This study

Table 1: Average global crust and river values for iron and aluminum

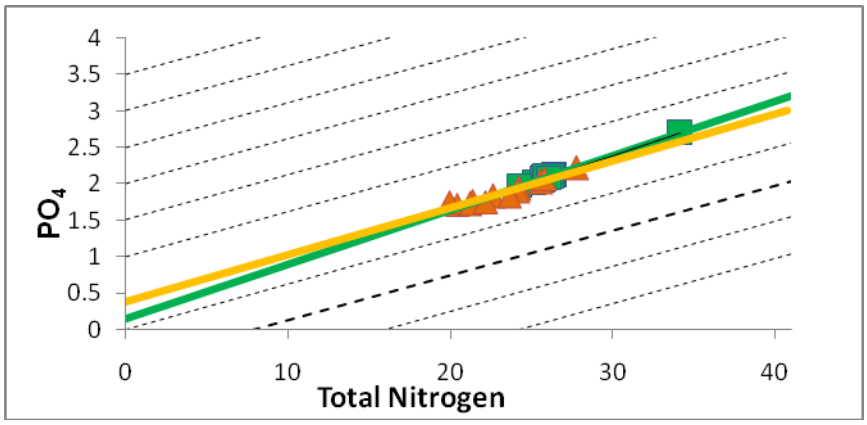


Figure 3: Total Nitrogen (NH_4 + NO_3 + NO_2) plotted with phosphate concentrations with slope of 12.4 compared with the standard Redfield slope of 16. Triangles show surface water samples while squares denote samples below the mixed layer. Dotted lines illustrate Redfield slope.

At no point during sampling period were any nutrient limitations observed (Fig. 3), even where phytoplankton blooms existed (Boas, 2008). Additionally, there was no correlation between iron and nutrients confirming that the iron in the bay is not coming in alongside nutrients with the upwelling of ocean waters. Iron to salinity also displayed no direct correlation which strongly suggests that neither the ice nor the river input is a major contributor to the iron found in the particulate matter during the winter. Therefore, during the winter months when fresh water terrestrial input is notably low (Royer, 1982) the continental crust is implicated as the primary source of iron which is further amplified by this low volume fresh water flux into the bay.

The high percentages from non-lithogenic sources strongly implicate remineralization from the sediments or biological processes. Noting that the values for iron are consistently higher below the mixed layer, and diminish as distance increased from the shallow sill, further implicates a flux of iron from the sediments (Fig. 3A). Additionally, there was no density stratification at or near the 40m sill and average currents are known to reach nearly 3m sec^{-1} (Hooge and Hooge, 2002). These strong currents around the sill extend to the benthic region (Kilbourne, in progress 2008) and further suggest that there is resuspension of iron from the sediments. As current velocities decrease away from the sill,

Station	Total Fe (µmol l^{-1})	Total Al (µmol l^{-1})	% Fe from non-crustal sources
Ice Valley River	79.35	137.8	0%
AA	17.1	2.2	93
00	13.7	14.2	40
A	34.1	26.9	55
1	46.5	28.8	64
2	56.7	28.6	71
4	16.9	15.3	48
5	12.9	13.8	38
7	4.8	1.0	88
8	9.7	0.8	95
9	4.1	1.6	77
11	28.9	20.9	58
21	23.1	26.3	34

Table 2: Particulate iron and aluminum and fraction from non-crustal sources at the surface. Station locations shown in Figure 1.

the amount of resuspended and remobilized material is also seen to decrease. At the head of bay where the glaciers are depositing high concentrations of iron and NH_4, and large anomalous phytoplankton blooms have been recorded (Hooge and Hooge, 2002), diffusion and remobilization of iron from the surface rich sediments is likely. A point by point analysis of biogeochemical iron as well as a best fit regression analysis demonstrate that biogeochemical iron values below the mixed layer are consistently higher than the surface and further reinforce the indication that the excess iron is coming from the sediments and likely is due to resuspension, remobilization, and possibly diffusion at the head of the bay.

Glacial ice and river sources should not be discounted though; they will most likely contribute more notably to the high value of particulate iron during the summer months when freshwater influx is at a peak.

Iron transport

During the winter and spring months, eddies in two primary areas known as the Sitka/Yakult and the Haida eddy regions (Fig. 4) form and move off the coastline carrying nutrients, including iron, into the HNLC North Pacific (Johnson et al. 2005; Henson and Thomas, 2008). It is apparent that very little iron is supplied to the open ocean via climatological average surface currents, but is episodically supplied via eddy spin off and deep water advection followed by upwelling (Johnson et al. 2005; Lam and Bishop 2008).

The Haida-Eddies tracked by Keith Johnson and colleagues (2005) spin off from the Canadian coast at approximately 55° N and generally carry up to 3000 km^3 of seawater. They were estimated to contain up to 4.8 x 10^7 mol iron integrated to 600m depth, a concentration of 16 µmol l^{-1}; this is over 2500 tonnes of iron being transported into one of the three major HNLC areas of the ocean. Glacier Bay iron values for the particulate matter measured an

Station	Total Fe (µmol l^{-1})	Total Al (µmol l^{-1})	% Fe from non-crustal sources
Ice Valley River	79.4	137.8	0
AA	29.6	12.8	75
00	6.2	3.5	67
A	58.0	37.1	63
1	36.1	22.9	63
4	12.8	9.4	57
7	27.8	14.9	73
9	8.3	5.6	61
11	39.1	5.2	92

Table 3: Particulate iron and aluminum and fraction from non-crustal sources below the mixed layer. Station locations shown in Figure 1.

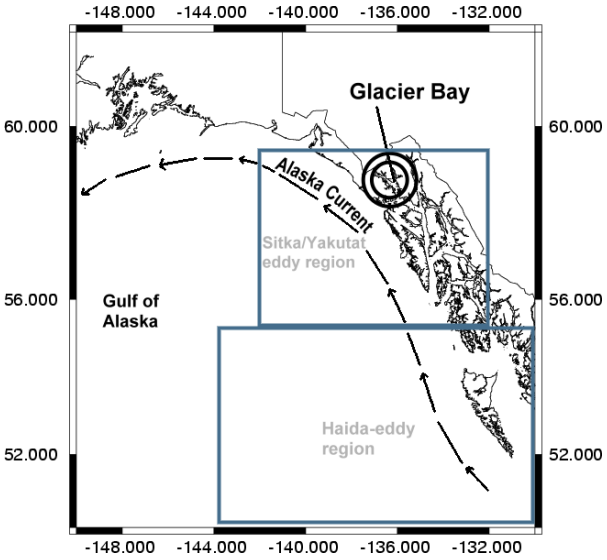
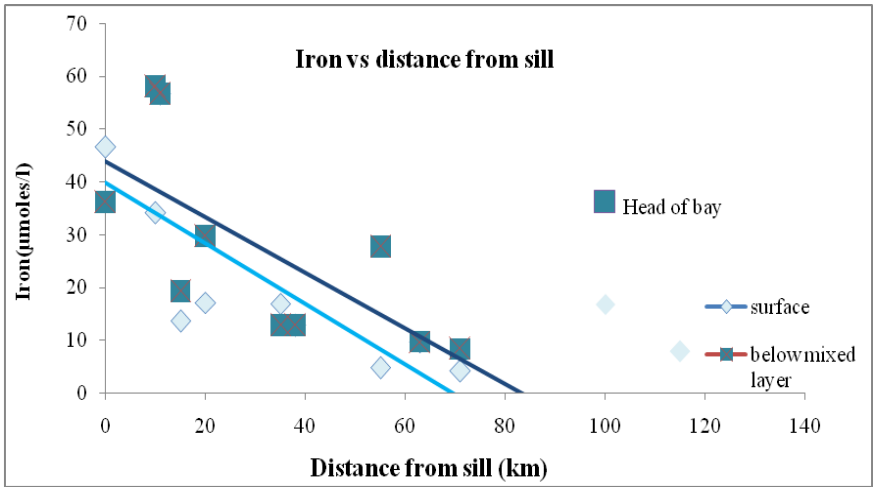


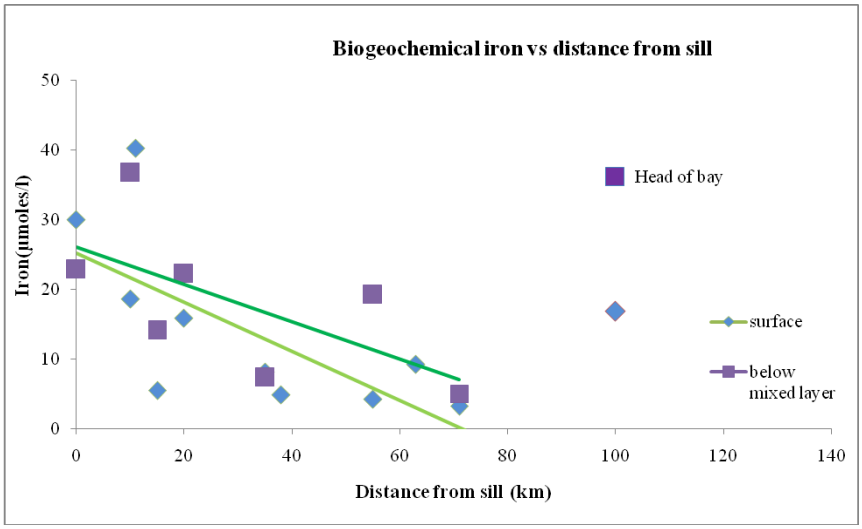
Figure 5: Map showing study area of Glacier Bay and two areas of eddy formation in the Gulf of Alaska: Sitka/Yakutat and Haida eddy regions.

average of 26 µmol l^{-1}, with other values above this average located outside the bay. This iron has the potential to reach the coast and during winter months when winds are from the northwest (Henson and Thomas 2008), the iron can further its journey along the Alaska Current (Fig. 4) and be transported via eddy spin off into HNLC open waters.

Glacier Bay surface and water source samples contained an average of 26 µmol $l^{-1}$of iron, and these high concentrations in the particulate matter will sink rapidly, but some of this iron is likely to be remineralized and remain in solution by binding to organic ligands (Rue and Bruland, 1995). Iron that is available to biology is generally soluble iron (II), with some bacteria documented with the ability to reduce the primary constituent of seawater iron (III) for nutrient assimilation using siderophore based reactions (Turner and Hunter, 2001). Bio-available iron in seawater makes up between 5 and 18% of the total iron concentrations (Duinker et al., 1974), with more current studies strongly favoring the lower end estimations (Turner and Hunter, 2001; Fitzwater et al., 2003; Johnson et al., 2005). Labile iron, (iron that can be reduced and made biologically available) was not distinguished in this study, but a greater particulate iron pool must supply a greater source of reducible iron in coastal waters. Given such high values of particulate iron, proximity to land, and low dust deposition over the HNLC North Pacific, atmospheric deposition in unlikely to be major source in coastal waters. Some labile iron will be utilized by biology and then recycled while some other quantity could continue via advection through the water column into the open ocean.



(a)



(b)

Figure 4: (A) Total particulate iron concentrations in relation to distance away from the sill showing a reduction in concentration at greater distances. The location at the head of Glacier Bay was plotted separately (B) Biogeochemical iron with sources outside the river values also showing a general decrease in iron with distance away from the sill.

Ocean Station Papa has been documented as an HNLC zone (Martin et al., 1989), and using data collected by Johnson et al. (2005), from coastal Washington waters to Station Papa a gradient at 800m was calculated. Assuming labile iron is around 7% of the total iron concentration (Fitzwater et al., 2003), and moving from 58-50° latitude a gradient of 0.39nm km^{-1} was established consuming nearly 96% of the beginning concentration. Treating the sill in Glacier Bay as a source of iron (Fig.7) and assuming it represented the advection of iron at 800m, there would be 1.01µmol of this particulate iron at Station Papa or classifying 7% of the particulate as labile iron, 71 nmol would be available for biology compared to Johnson et al.'s (2005) measured 0.7 nmol. Obviously, this is not a realistic gradient in view of the many

biogeochemical processes including scavenging and biological utilization, but it does show a potential upper bound for iron and that the continental shelf has the potential to influence on the open ocean through deep water advection.

The high concentrations of iron in and around Glacier Bay are most likely similar along the continental shelf surrounding the Gulf of Alaska and are a possible iron sources within the Alaskan Gyre. It has been noted that very little iron gets transported away from the coastal environment into the Gulf of Alaska via surface current advection (Johnson et al., 2005), however other forms of transport are likely as both Sitka/Yukatat and Haida eddies that spin off of the continental margin do carry valuable nutrients including iron into the HNLC Gulf of Alaska and deep water advection off the continental shelf also may have a large role in the transport of iron into the HNLC areas of the ocean.

Conclusions

Glacier Bay demonstrates unique and unexpected results where:

- No correlation with iron and salinity show that rivers and ice are not the primary source for iron, and no correlation with nutrients also illustrates that iron is not coming in with nutrients from outside the bay
- Iron to aluminum ratios suggest that iron is not coming from crustal sources and imply biological processes and remineralization are responsible for the excess iron in the bay
- Physical processes and well mixed station samples suggest shear in bottom water remineralizes iron out of the sediments

Glacier Bay is potentially just one of many glacially fed fjords on the continental margin where

- concentrations of iron are high
- the potential for advection into the HNLC areas of the open ocean is likely through eddy transport and deep water advection
- The possibility of storm wind transport also could be further explored. Storm winds are known to transport large volumes of material great distances (Anderson and Walker, 2006) so that perhaps short-distance airborne eddies of iron rich coastal spray may also be a factor in transporting iron into HNLC areas of the ocean.

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