Author Comment to reviewer 1:

I. General comments

In this manuscript, the authors report a data set of carbonate system measurements in an Alaskan Inland Sea in spring and autumn from 2009 to 2012, with special attention to the impact of tidewater glacial melt on the marine carbonate system and CaCO3 corrosivity (i.e. saturation state). They observed the seasonal changes in in the upper layer ocean, e.g. in the surface plumes of glacial melt and in the mode water formed in proglacial fjords. For the waters affected by glacial melt, decoupling of pCO2, pH and is discussed together with the positive feedback of the atmospheric CO2 uptake which may further decrease . The authors also discuss the potential farther-reaching impacts of these processes due to physical transportation. Overall, the subject of this manuscript meets the general interest of Biogeosciences. The data are of good quality and the discussions are well organized. I thus support the publication of this work after a moderate modification. Please see below for my detailed comments.

II. Specific comments

1. The authors suggest that the addition of glacial melt would exacerbate CaCO3 corrosivity, but the cause of this decrease in seems not been discussed. The authors are encouraged to discuss the underlying controlling mechanisms of this phenomenon (what aspect of is affected? [Ca2+], [CO3–] or K'sp?).

It is the presence of corrosive and pCO2 undersaturated glacial melt that drives a positive feedback where air-sea exchange of CO2 further increases the corrosivity. The underlining mechanism is the increase in DIC due to atmospheric CO2 uptake. This interaction is discussed using first an example calculation and second using a graphic. We have edited some of the statements preceding the example calculation to clarity the controlling mechanism, and now this discussion as its own section in the revised version of the manuscript.

2. The use of "glacial melt" is a bit confusing and the sources of the freshwater in the study area need to be described more clearly. In the introduction, the authors emphasize the "direct glacial discharge at ocean/glacier interfaces which is free from additional alteration that would occur in proglacial streams draining mountain-terminating glaciers". This leaves the impression that only this kind of glacial melt will be discussed in this manuscript. However, in the 'study area' section, the authors state that "glacial runoff, either as a direct input to the sound or indirectly via progalcial streams, contributes an estimated 60% of the freshwater discharged to PWS". And in Fig. 9, they discuss the mixing process involving the glacial melt from the nearby land-terminating Bench Glacier (is this a proglacial stream?). The authors should better describe the sources of the freshwater especially in the fjords where the most corrective surface waters were observed. Although share a common salinity (S=0), the chemical properties of the direct glacial discharge may be significantly different to those of the progalcial streams, so do their influences on the seawater carbonate system. If there exists different sources of freshwater, it seems to me that the fjord under study is analogous to an estuary system with multiple tributaries. It is important to distinguish the different

impacts of various sources of freshwater. Current data set in this work may not be able to address this issue, but it is a problem needs to be studied in the future.

The reviewer makes a very good point, and unfortunately we do not have the data to resolve all freshwater sources in Prince William Sound. We comment on our inability to resolve all sources of freshwater in the revised version of the manuscript, however, our inclusion of the mixing lines versus melt lines in temperature-salinity space was an attempt to at least qualify that the more corrosive conditions observed in Icy Bay during September 2012 were due to a greater degree of glacier melt. Owing to the chemical conditions observed nearest the glaciers, and to the fact that glacial discharge is greater in September relative to May, we argue that the decrease in average omega in PWS in September is due to the addition of glacier melt and the feedback with air-sea gas exchange that ensues because of undersaturated pCO2.

3. Salinity is the indictor of the surface plumes of glacial melt, and the salinity data should be presented in the corresponding figures. If the plumes is strictly limited to surface, the depth of the plume should be given.

We have changed the figures to show salinity, and where appropriate mentioned the depth of the surface plumes.

4. Fig. 2 clearly shows the data points of the most corrective surface waters in the fjords which are characterized by low salinity, low TA, low pH, low and low TA/DIC ratio. Fig. 2A suggests that these points at the low salinity end are well below the linear TA-salinity relationship that most surface waters obey. Could this be a TA removal (indicated by the low TA/DIC ratio) or another segment of the TA-salinity mixing line? The TA-salinity regression could be extrapolated to get an intercept at S=0, which gives an indicate of the end member property of the freshwater. In Icy Bay, the intercept can be compared to the reported TA in the glacial melt from the nearby land-terminating Bench Glacier.

Following your suggestion, we have updated this figure and included a linear fit of the TA and salinity data. We have also added DIC to the new figure so that the reader can see TA is not the only variable that departed from an end-member mixing line (although we do not provide a fit for the DIC-TA data). TA is not solely being removed; both TA and DIC are depleted in the low salinity, glacially influenced water. We have not calculated mixing lines for the low salinity values because the few data points we have thus far do not portray a reasonable S=0 intercept, but we do note that low salinity data suggest a freshwater end-member much lower than the S=0 intercept for the mixing line calculated using data with salinity greater than 22.

5. As pCO2 and pH are more sensitive to the temperature change than , temperature might plays a role in the coupling/decoupling of pCO2, pH and . In Fig. 3, have the authors tried that if the use of temperature-normalized pCO2 (4.23% per degree, Takahashi et al. 1993) and temperature-normalized would improve the fitting?

We did not perform this calculation because we are including all PWS data from the

surface to depth in Figure 3 (now Figure 5), and therefore a temperature correction would be misleading for data below the surface that does not experience the same degree of warming. We present the data in this way so that the entire water column is represented, thereby showing the seasonal change in omega and pCO2 across the full dynamic range of observations. However we did include a discussion point of the role of temperature in the manuscript and highlight the difference in sensitivities between pCO2 and omega. We have also updated the figure to highlight the surface (< 9 m) values.

6. p14901, lines 16-21: Please discuss briefly what causes the undersaturated pCO2 in the glacial melt and why it differs from that of sea ice melt (oversaturated pCO2)?

We have clarified this statement following a comment from Reviewer 2.

III. Technical corrections
p14888, lines 15 “across the sound” : is it only in the western sound (as shown in Fig. 7)?

Thank you, we have rephrased this statement in the Abstract.

p14891, lines 9-15: Just for your reference, another possible reason for the mismatch between the direct observed pCO2 and calculated pCO2 could be the existent of organic alkalinity (Cai et al. 1998; Hernandez-Ayon et al. 2007; Muller and Bleie 2008; Kim and Lee 2009).

Thank you, we appreciate these references, and are aware of the organic component of alkalinity that can be an important component in some estuarine systems. However we leave these introductory statements as is because our intent is to report the conclusions of Sejr et al. 2011.

p14891, lines 15-18 “It follows : : :”: Please rewrite this sentence. According to the authors’ review on the work of Sejr et al., [2011], the “direct glacial discharge less buffered by high concentrations of reactive particles derived from river sediments” would result in lower TA and higher pCO2 in the glacial melt when compared to those in the proglacial rivers, but not necessary “severe CaCO3 corrosivity in addition to undersaturated pCO2”.

We have added an important reference following a comment by Reviewer 2, and have clarified this sentence in the manuscript.

p14896, lines 21: 0.06, should this be 0.60?

Corrected, thank you.

p14900, lines 9-11 “The cumulative pulse : : : the wholesale reduction in CaCO3 saturations in PWS”: do you mean in the surface layer in PWS? The seasonal changes in surface and sub-surface are different according to the discussion below.

Yes, thank you, we have clarified in the manuscript that our discussion here targeted our

p14900, lines 19-21: “the lesser increase in the dynamic range of DIC relative to TA” can result from either “a process is adding DIC with little effect on TA” or additional TA removal with little effect on DIC (also see the comment 4).

We argue that this pattern is not evidence of TA removal, as the TA and DIC data versus salinity (new figure) show both are reduced in the lowest salinity surface water with a slight increase in DIC relative to TA (reflected in the TA/DIC ratios), but rather the addition of DIC via air-sea gas exchange due to the undersaturated pCO2 in melt plumes. This is discussed in detail in the revised version of the manuscript Results and Discussion section.