Below, we address comments from RC1. We cluster each comment and separate them as "1" Reviewer comments, "2,3" are responses and revisions. A pdf version (bg-2018-226-RC1-supplement\_Krause\_etal\_response) of this response has been uploaded in the supplement.

- 1) The manuscript of Krause *et al.* presents interesting results concerning biogenic silica production and export levels as well as estimates of kinetic constants from an opportunistic sampling near the Svalbard Archipelago in the Arctic Ocean by late spring. The data presented are the first direct (silicon–32 method) measurements of biogenic silica production in the Arctic Ocean, which in themselves deserve publication. From these data, the authors then attempt to establish the potential control of diatom production by the availability of silicic acid as well as the contribution of diatoms to total primary production. However, I think the authors are pushing their limited data set much too far and that the manuscript should be shortened by getting more concise.
- 2,3) We thank the reviewer for their constructive criticism, these have strengthened the manuscript. Below, we address the concerns. The revisions are aimed to both address these issues and make the manuscript more concise.
- 1) Firstly, I find it difficult to understand why the authors focus on the so-called Egge & Aksnes  $2 \mu M$  H4SiO4 threshold value, as it is clear that the data from this publication have been wrongly interpreted in several past publications (which is recognized by the authors besides).
- 2,3) We agree with the reviewer and the focus on this apparent threshold value is due to its wide use in the field. As of August 2018, the Egge & Aksnes (1992) manuscript has been cited over 700 times, and many explicitly regard this 2  $\mu$ M threshold for diatom microplankton dominance among many systems. This type of prose has been reduced; however, given this is the threshold embraced by the community it must be acknowledged.
- 1) I suggest just using their kinetic values to discuss the potential limitation of diatom uptake by H4SiO4 availability, and then shortly discuss hypotheses for growth limitation, which is another point not directly assessed in this study. ON the other hand, authors might consider that the actual limitation starts under 2 times KS.
- 2,3) The original version discusses kinetic limitation between lines 434 to 455, followed by a section discussing the possibility that silicic acid may be low enough to limit diatom growth (line 459-462). If we interpret the reviewer correctly, then "under 2 times Ks" (i.e. half of the half-saturation constant) is the silicic acid concentration when uptake (i.e. Vb) is 25% of maximum uptake (i.e. Vmax) which is the same metric we use to diagnose potential growth limitation (line 461). We have made this clearer (e.g. denote the equivalence of "under 2 times Ks" and Vb/Vmax <0.25) in the revision.
- 1) The comparison between nitrate and silicic acid concentrations is not very clear. First, they mention a 2.5 slope (Figure 5) between nitrate and silicic acid, which means that nitrate is taken up 2.5 times faster than silicic acid, and then authors take a 1:1 ratio to discuss the potential for silicic acid limitation. I can imagine that they are trying to decipher the relative contribution of siliceous vs. non-siliceous (e.g. *Phaeocystis*) components of the phytoplankton community but this should be clearly indicated. On top of that, the use of a 1:1 Si:N ratio is questionable (large species—specific variations, see below for Si:C).
- 2,3) The approach silicic acid versus nitrate drawdown has been used elsewhere (Monterey Bay, Brzezinski et al. 1997; Barents Sea, Rey et al. 1987; Southern Ocean diatom cultures, Takeda 1998). We agree with the reviewer that this nutrient data includes drawdown of

nitrate from other organisms, likely *Phaeocystis*, hence the slope exceeds 1 (i.e. nitrate drawn down faster than silicic acid). We also agree that there is variability within the drawdown ratios. However, there is currently a lack of relevant culture data to apply for the conditions observed here (we know of recent manuscripts, currently in preparation, to address this gap; but those cannot be cited formally here). Most polar culture work focuses on Fe-limitation (e.g. Takeda 1988, Nature) and Southern Ocean clones; however, under Fe-replete conditions Takeda (1998) reported Si:N between 0.73 (*Chaetoceros dichaeta*) and 1.2 (*Nitzschia* sp.), the average among these clones is 0.97, not unlike the 1:1 ratio from Brzezinski 1985. In the revision, we make this point with more clarity and provide the Takeda (1998) study as a literature basis for the 1:1 ratio (beyond the canonical Brzezinski stoichiometry).

- 1) The contribution of diatoms to primary production is also another weak point of the ms. The calculation is based on a transformation of rSi to rC by using the average Brzezinski' ratio of 0.13. This is a very simplistic way of addressing this important question as this ratio is known to be subject to large species—specific variations (e.g. Brzezinski gives a Si:C biomass range of  $0.15 \pm 0.04$  for large diatoms, which could result in a ~2 times range for rC estimates). This is somehow risky business and should, at least, be acknowledged and discussed.
- 2,3) As with the previous point, we agree that there is variability within the stoichiometry. While admittedly simplistic, we also have coarse resolution (n = 6 coupled measurements), which (in our opinion) do not merit as sophisticated approach given it would not be robust; this data coupling was serendipitous (hence the low n value) but do provide a first-order estimate. The estimates cluster into three modes when using Si:C of 0.13, i.e. diatoms insignificant (line 482), diatoms ~50% of primary production (line 483-484), diatoms ~all primary production (lines 484-485). We point out the simplicity of this ratio for the Erik Erikssenstretet station (line 486-487) which could be due to stoichiometric changes associated with growth rate shifts (likely modifying Si:C) during a bloom phase. To this section, we added a minor addendum stating that if we are off by a factor of two in Si:C (i.e. up to 0.26), then the resulting clusters would change (i.e. 2 stations where diatoms were insignificant, 2 stations where diatoms did ~25% of production, 1 station where diatom did ~50% of production, 1 station where diatoms did ~100% of production). However, this does not change the two main interpretations: 1) diatoms quantitative contribution to primary production is "bloom and bust" and 2) even when diatom biomass is relatively low, their contribution to production can still quantitatively important (e.g. 25 - 50%).
- 1) Finally, the authors present data for direct diatom cell export but the underlying issue is not clearly stated: Do they want to compare direct diatom sedimentation by mass sinking to other export vectors such as repackaging? If yes this should be clearly stressed.
- 2,3) The reviewer's interpretation of our intent is correct, we suggest that the reason why cellular export is similar to previous studies, despite biogenic silica export being higher than other studies, is consistent with repackaging which we termed "food web effect." We have revised this for clarification.
- 1) line 28 : "diatom cellular export" the wording is misleading (could be export from a diatom cell). I'd rather use "export of diatom cells".
- 2,3) Changed as suggested.
- 1) line 65 : " A more recent analysis demonstrated a decline in pre-bloom [Si(OH)4] concentrations by  $1-2 \mu M$  across the north Atlantic subpolar and polar regions over the last 25 years (Hátún *et al.*, 2017); this is consistent with the general Arctic region being a net

- exporter of silicic acid (Torres-Valdés *et al.*, 2013)." I don't see the consistence between the decrease of H4SiO4 concentrations and the net exportation of this nutrient; please rephrase.
- 2,3) This is rephrased, e.g. removing "this is consistent with the general Arctic region being a net exporter of silicic acid (Torres-Valdés *et al.*, 2013)"
- 1) line 68: "This is in stark contrast to the 10– $60~\mu M$  [Si(OH)4] observed in the surface waters of the Southern Ocean and the marginal ice zone around Antarctica (Nelson and Gordon, 1982; Brzezinski et al., 2001), where [Si(OH)4] is unlikely to limit the rate of diatom production or biomass yield." I disagree; There are ample references to state that actually the reverse is true, due to sometimes unusual high KS (e.g. Nelson & Tréguer MEPS 1992, Nelson et al. DSR II 2001, Mosseri et al. DSR II 2008).
- 2,3) The reviewer is correct as kinetic limitation (i.e. ambient silicic acid limits the rate of diatom silica production) is clearly observed in the Southern Ocean. Our intent (which was not clearly conveyed) was to contrast the effects of the high silicic acid in the Southern Ocean vs. the Arctic. This is rephrased "... where [Si(OH)4] is unlikely to limit diatom growth unless iron is replete, and stimulates exceptional blooms which consume Si, or assemblages are highly inefficient for Si uptake."
- 1) line 76: " ... and a 2  $\mu$ M threshold [Si(OH)4] defines where diatoms are outcompeted by flagellates (Egge and Aksnes, 1992)." I strongly disagree with that sentence. The work of Egge and Aksnes did not evidence any real threshold (no kinetic values measured) and just merely indicated areas of realized niches for diatom vs. flagellates with regards to Si vs. P availability. Please do not cite this reference in such a way that was even not addressed by the authors of this paper. + as indicated above.
- 2,3) We agree with the reviewer, especially in the lack of physiological data used in the original study to assess a threshold silicic acid; please see reply to general comment. We have revised by using more specific language "e.g. based on correlating microplankton abundance and nutrient concentration, opposed to physiological analyses, a 2  $\mu$ M threshold [Si(OH)4] has been used extensively to define where diatoms are outcompeted by flagellates (Egge and Aksnes, 1992)."
- 1) line 137: "... suggesting that N was likely more important than P for primary production." As authors refer to absolute concentrations, the correct phrasing should be: "... suggesting that N was likely more important than P for potentially limiting primary production." 2,3) Changed as suggested.
- 1) line 138: "These phosphate data are not discussed." Even though a range would be welcome.
- 2,3) Revised (e.g. "These phosphate data (0.1 0.6  $\mu M$  in the upper 50 m) are not discussed.")
- 1) line 153: "... fixed with an aldehyde mixture of hexamethylenetetramine-buffered formaldehyde and glutaraldehyde at 0.1 and 1% final concentration, respectively, as suggested by Tsuji and Yanagita (1981) ..." although this should be OK this is not the usual fixative for diatoms (acidic Lugol preferred), partly due to its toxicity for the microscopical examiner.
- 2,3) Agreed. Given the interdisciplinary nature of the cruise and the lack of excess operation time, this fixative was used by the Norwegian Polar Institute group to do multiple analyses from one sample (which cell counts were only one); these other analyses are beyond the

scope of this communication.

- 1) line 164: " ... neutral density screened bags ... " please mention the photometric levels used.
- 2,3) Added "... neutral density screened bags simulating 50%, 20% and 1% of irradiance just below the surface."
- 1) line 207: "Export rates were calculated using the standing stock measurements, length of deployment, and trap opening area." Please give the model/type of sediment trap.
- 2,3) Added "KC Denmark design (outer diameter 72 mm, length 450 mm)."
- 1) line 260: "... except for the Hinlopen ice algae, where the melt water ..." Is that naturally– melted ice or meltwater produced by ice melting in the lab? Please clarify.
- 2,3) Thank you for the question, this has been clarified, e.g. "... where water, which was melted at ambient air temperature on the vessel, had ..."
- 1) Line 331: "Brown *et al.*, 2003 " Comment: For some strange reason L. Brown's incubations lasted for only 6 hours, which renders her production results questionable.
- 2,3) We agree with the reviewer; however, given the limited data from the Northeast Atlantic, we prefer to be thorough and include the Brown et al. publication here.
- 1) line 339: "... Varela *et al.* (2013) recently reported that [Si(OH)4] in surface waters (>5 µM) are unlikely to be significantly limiting to diatoms in any sector of the Bering, Chukchi or Beaufort Sea regions." Although for Subarctic waters Brown *et al.* (2003, mentioned just above) kinetic experiments show a strong limitation (non-saturating kinetics) up to 30 µM.
- 2,3) We agree with the reviewer; however, the goal of this sentence is to isolate the Canadian-& United-States Arctic waters with the European Arctic waters. We add the Brown et al. (2003) kinetic data in a section below (e.g. with Allen et al. 2005, Kristiansen et al. 2001 discussion near lines 442, 443).
- 1) line 387: "Suboptimal silicon availability affects the rate of diatom bSiO2 production and can limit their growth. A widely cited [Si(OH)4] threshold, below which diatoms will be outcompeted by other phytoplankton, is  $\sim$ 2.0  $\mu$ M; this metric was derived from a comparison of diatom abundance (relative to total microplankton) versus [Si(OH)4] during mesocosm experiments in a Norwegian fjord system (Egge and Aksnes, 1992)." should be removed: No need to discuss this threshold as it is mentioned that it is strongly criticized (and see my comment above).
- 2,3) Thank you for the perspective; this section will be removed (please see response to earlier comments on this topic).
- 1) line 410: "This indeed indicates that phytoplankton can deplete nitrogen to levels below detection while they appear unable to deplete Si(OH)4 pools below 0.5  $\mu$ M, which would indicate 0.5  $\mu$ M is the ultimate Si(OH)4 concentration required to support diatom growth." I disagree with this interpretation. The 0.5  $\mu$ M Si level just reflects the residual H4SiO4
- I disagree with this interpretation. The 0.5 μM Si level just reflects the residual H4SiO4 stock after complete removal of nitrate.
- line 422: "... if diatoms are limited by an absolute [Si(OH)4] (e.g. 2  $\mu$ M), ..." This is speculative: By what evidence is this proposition supported?
- 2,3) Given the reviewer's comments for the paragraphs starting in line 405 (i.e. line 410 comment) and line 41 (line 422 comment), these paragraphs have been revised to

demonstrate that assessment of nitrate vs. silicic acid consumption relationships can be used to infer either N or Si limitation of diatom biomass and growth; this sets up the importance of infers growth dynamics using more direct physiological uptake data (e.g. Si uptake kinetics).

- 1) line 436: "... the relationship between  $V_b$  and [Si(OH)4] also supports that Si regulates diatom productivity to some degree." The large dispersion of data points on Figure 5 results in a very weak relationship, so that there is certainly something else explaining the low realized  $V_b$  at the 4.5  $\mu$ M H4SiO4 level.
- 2,3) The reviewer is correct, there is variability. But please note these data are from all diatom assemblages sampled among all stations and depths; thus, the linearity of the relationship would not be expected to be high, in fact, we were surprised to see nearly 40% of the variability explained given so many assemblages. However, we feel the Fig. 5 data, which integrates all samples, is the most conservative way to estimate the aggregate Ks value for euphotic diatoms among all stations at the time of this cruise.
- 1) line 443: " ... Allen *et al.* (2005) observed a linear response in Vb between ambient and 5  $\mu$ M [Si(OH)4], which suggests uptake did not show any degree of saturation at this concentration." Also in Brown *et al.* (2003); as mentioned above.
- 2,3) The Brown et al. (2003) reference has been added.
- 1) line 517: " At van Mijenfjorden, the rate of export in the upper 40 m represented 39% of the ∫bSiO2 standing stock (23.3 mmol Si m-2) in the same vertical layer." − I don't understand as from Table 1 it seems that the standing stock is 10.8 and the export 9.03?
- 2,3) Thank you for the observation, this is clarified. We now reference Fig. 2C as the source of the van Mijenfjorden integral data. Table 1 integrates biogenic silica stock and production to 20 m (i.e. deepest depth among all profiles) whereas the shallows depth for sediment traps among all deployments was from 40 m. For van Mijenfjorden, we have biogenic silica measurements down to 50 m (Fig. 2C), which allows for the comparison as described in line 517.
- 1) line 524: "The rate of bSiO<sub>2</sub> export was also at least a factor of four higher than  $\rho$  in the upper 20 m." I was not able to find where did this come from.
- 2,3) Thank you for the observation, this is a general trend among stations in Table 1 (i.e. biogenic silica export was much higher than integrated biogenic silica production). This has been revised "The rate of bSiO2 export among all export- and production stations was also at least a factor of four higher than  $\int \rho$  in the upper 20 m (Table 1)."