Responses to reviewers and changed manuscript

Please, find below our responses to reviewer 1-3. We marked the text and suggestions by the reviewer in a light-brown and our response in a black font color. The changes in the manuscript are tracked using track-changes in Microsoft word. We are thankful for all reviewers' comments and revised the manuscript according to all suggestions. We believe that the revised manuscript is considerably improved.

Response to reviewer 1

We want to thank reviewer 1 for the very positive constructive feedback. All specific comments are changed in the manuscript as suggested by the reviewer. A more detailed response regarding the representativeness of *Chaetoceros socialis* for Arctic coastal systems is given below.

Reviewers comment: *Chaetoceros socialis* may not be representative of the the most important diatom species across all of the Arctic coastal areas. How representative do you expect that it is?

We acknowledge that many different species contribute to the bloom formations in the Arctic coastal areas, including pennate sea ice algae and several pelagic centric diatoms (see below) *Chaeotoceros socialis* may not be the most dominant species in all coastal Arctic spring blooms, however it has been reported as dominating blooms in several areas (see below). We thus consider *C. socialis* to be overall a representative model organism.

Chaetoceros socialis is a widely occurring marine diatom species that has been observed from Arctic seas into warmer oceans like the Gulf of California (Hasle and Syvertsen 1997) that differ physiologically and morphologically (Degerlund et al. 2012, Huseby et al. 2012). Current research indicates several cryptic species to be within the *C. socialis* complex (Gaonkar et al. 2017, De Luca et al. 2019). It is frequently used in culture based experiments to evaluate for example the role of ocean acification (Li et al. 2017), and DMS (Baumann et al. 1994) and lipid production (Artamonova et al. 2017).

In Arctic waters, it has been observed as bloom forming species across the Arctic with for example bloom occurring in the North Water Polynya between July and September (Booth et al. 2002), the Barents Sea (Rey and Skjoldal 1987, Rat'kova and Wassmann 2002) and other Arctic coastal sites, often dominating phytoplankton biomass following the blooming of *Thalassiosira* spp. (von Quillfeldt 2005).

Besides *C.socialis*, coastal Arctic spring blooms are typically dominated by other chain forming diatoms, such as *Thalassiosira* spp., *Fragillaripsis* spp., *Chaetoceros* spp., *Navicula* spp., or *Skeletonema* spp.. All of these pennate or centric diatoms share similar requirements for inorganic nutrients and all of these groups are typically limited by silicate and/or nitrogen limitation in coastal Arctic systems. In addition, all of these groups have similar physiological opportunities to respond to nutrient limitations, can excrete EPS and interact with bacteria. Hence, we are confident that *C. socialis* is suitable as model organism, representative for coastal Arctic spring blooms unless silicate is limiting from the start in which case, flagellates, such as *Phaeocystis* may dominate (As discussed in line 302 in our manuscript).

We added a few more details and references in the manuscript to support these statements in the following way, with changes highlighted in green:

Line 53-58: Phytoplankton blooms may be dominated by a single or a few algal species, often with a similar physiology during certain phases of the bloom (e.g. Eilertsen et al., 1989; Degerlund and Eilertsen, 2010; Iversen and Seuthe, 2011). Chain-forming diatoms, sharing physiological needs and responses to nutrient limitations (e.g. Eilertsen et al., 1989; von Quillfeldt, 2005), typically dominate these blooms. In some Arctic and sub-Arctic areas the Arctic phytoplankton chosen for this model, *Chaetoceros socialis*, is a dominant species during spring blooms (Rey and Skjoldal, 1987; Eilertsen et al., 1989; Booth et al., 2002; Ratkova and Wassmann, 2002; von Quillfeldt, 2005; Degerlund and Eilertsen, 2010).

Line 297-299: While *C. socialis* may not be the dominant species in all coastal Arctic phytoplankton blooms, we argue that it is representative for chain-forming diatoms typically dominating these systems due to their shared needs and responses to nutrient limitations (e.g. Eilertsen et al., 1989; von Quillfeldt, 2005).

References

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von Quillfeldt 2005. Common Diatom Species in Arctic Spring Blooms: Their Distribution and Abundance.

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Response to reviewer 2

Summary

This manuscript presents an interesting combined laboratory and modelling study of the nutrient dynamics of a diatom species common in the Arctic. The laboratory component uses two experimental set-ups: 1. axenic cultures of the diatom species; 2. cultures of the diatom species that include associated bacteria. Short incubations (_2 weeks) of these cultures take them from exponential phase through to stationary phase, with the cultures sampled throughout to measure cell counts, nutrient concentrations, etc. After an initial period of diatom cell number growth (week 1) in both cultures, this stops as NOx and dSi concentrations approach limiting concentrations. However, NH4 is consistently higher in the non-axenic cultures, and the bacterial cell counts in these cultures increase exponentially during the latter period of the incubations (week 2). The authors interpret the presence of bacteria as being conducive to supplying the diatoms with regenerated nutrients. The modelling component uses a base model, G98, and an extended model based on this that includes a number of additional processes with relevance to the laboratory setting and the hypothesised role of bacterial remineralisation in supporting phytoplankton growth. The models are tuned to fit the laboratory data, with a manual phase to retain consistent parameter values between the models. The authors conclude with a discussion on the application of their results to the real Arctic and its expected future state.

I have listed a number of significant general comments below, followed by more specific and often minor comments. Overall, my assessment is that the manuscript requires major revision to clarify and amend the work described.

We want to sincerely thank the reviewer for the very thorough review and believe the suggestions helped to improve the manuscript considerably. We included all suggestions into a revised version as described below. We also changed the fixed 80% reduction term in our model to a parameter that was subject to the fitting approach and sensitivity analyses. We fitted the model again with a more automated fitting approach and reached better fits for both the G98 and extended EXT model.

General comments

Upfront, my modelling background means that I cannot comment directly on the details of the laboratory work in the study. However, I note that the experiments conducted exhibit anomalies that are not addressed in the manuscript. In Figure 1c, phosphate in bacterial cultures exhibits a strong spike upwards at day 8 that persists and shows high variability. In Figure 3d, chlorophyll in bacterial cultures shows a marked but temporary spike downward at day 8. While the latter is likely a replication or measurement issue, the former is harder to understand, and the manuscript does not discuss its scale or variability. It would be useful to know what the authors believed happened here, particularly in the case of phosphate where bottle concentrations approximately double against a backdrop of slowly declining phytoplankton and rising bacteria concentrations. The model may even be able to help on this point.

Since our study, does not focus on phosphate, we did not describe its dynamics in detail. However, we acknowledge that a short description and explanation of the anomalies is helpful for the reader to understand the overall experiment and nutrient dynamics and added some details.

The strong spike of phosphate after day 8 corresponds with the end of the exponential phase for algal growth and a spike of ammonium. At the same time bacteria abundances start increasing considerably. Thus, we explain the phosphate peak by increased bacterial regeneration (source of phosphate) and decreased algal uptake (sink of phosphate) at the same day. Due to the small bacteria biomass compared to algae, we assume limited phosphate incorporation in the bacteria biomass pool. Besides, the diatom culture may excrete additional DOM under stress, such as silicate limitation, contributing to labile DOM available for regeneration and thereby increasing the phosphate peak, which is however not part of the current extended model. We calculated the N:P ratio of the NH4 and PO4 peak at day 8, and realized that the ratio is approximately 1:1, which is different from the Redfield ratio. We see this as evidence that increased regeneration of NH4 and PO4 is not the only explanation for the PO4 peak and suggest the storage of (organic) polyphosphate in diatoms and release under stress as another potential source.

Changes in the text:

3.1) “With the onset of the stationary phase in the BAC+ experiment, PO4 and NH4 concentrations doubled within 2 to 4 days and stayed high with variations in phosphate concentrations, while they stayed low in BAC-. With depletion of NO3 in BAC+, NH4 concentrations remained high, while PO4 concentrations dropped.”

4.1) “With the start of the stationary phase, NH4 and PO4 concentrations doubled, presumably due to decreased assimilation by the silicate starved diatoms and increased regeneration by bacteria, supplied with increasing labile DOM (doubled remineralisation rate in EXT) excreted by the stressed algae. After NO3 depletion at day 15, also PO4 concentrations drop, indicating a coupling of N:P metabolism “…” Excretion of organic phosphate by diatoms is also common in cultures with surplus orthophosphate (Admiraal and Werner, 1983), which can be another explanation of the phosphate peak after silicate becomes limiting.”

The spike in Chl is based on one single measurement, since the upper and lower range represent max and min values. Since chlorophyll measurements are sensible towards light, and pH, we argue that this negative spike is a measurement artifact of a single sample of the experiment.

Changes:

Figure 4, 5, B1) “…Chlorophyll a concentration in experimental cultures with a potential outlier at day 8, presumably due to photodegradation, causing a negative spike.”

The manuscript’s model description appears incomplete, with equations for terms such as those for dSi omitted. More generally, the manuscript would be improved by simply making clear which models are being run – while the text refers to model G98 and “the extended model”, the plots shown refer instead

to “model + excr” and “model - excr”. What might be helpful is to have some sort of diagram of the two main models being used (G98 and Extended) to help illustrate the main connections between state variables, and make clear the differences between the two models.

We added the missing equation and double-checked for any other incomplete model descriptions.

Changes in Table:

7a)	Silicate uptake *(Monod kinetics after Spilling et al., 2010)*	$\frac{dSi_d}{dt} = V_S^C = \left(V_{max} Si_d \frac{Si_d - S_{min}}{K_{si} S_{min}}\right) C$
7b)		$\frac{dSi_p}{dt} = \frac{-dSi_d}{dt} 14$

We also clarified, which models are run and defined abbreviations (G98 and EXT models/ BAC- and BAC+ treatments) for the different models that we kept throughout the manuscript and figures. We also added a schematic diagram of the two main models, which we agree helps clarifying the differences considerably.

Changes:

2.2) “Details regarding model equations are provided in the Appendix (Table A1) and a schematic representation of the models is given in Figure 1. We used a dynamic cell quota model by Geider et al. (1998) to describe the BAC- experiment (G98). We then extended the G98 model to represent the role of silicate limitation, bacterial regeneration of ammonium, and different kinetics for ammonium and nitrate uptake (EXT) and fitted it to the BAC+ experiment while retaining the parameter values estimated for G98.”…” For testing the importance of DON excretion we also ran the EXT model without DON excretion (EXT–excr).“

Fig. 1)
“

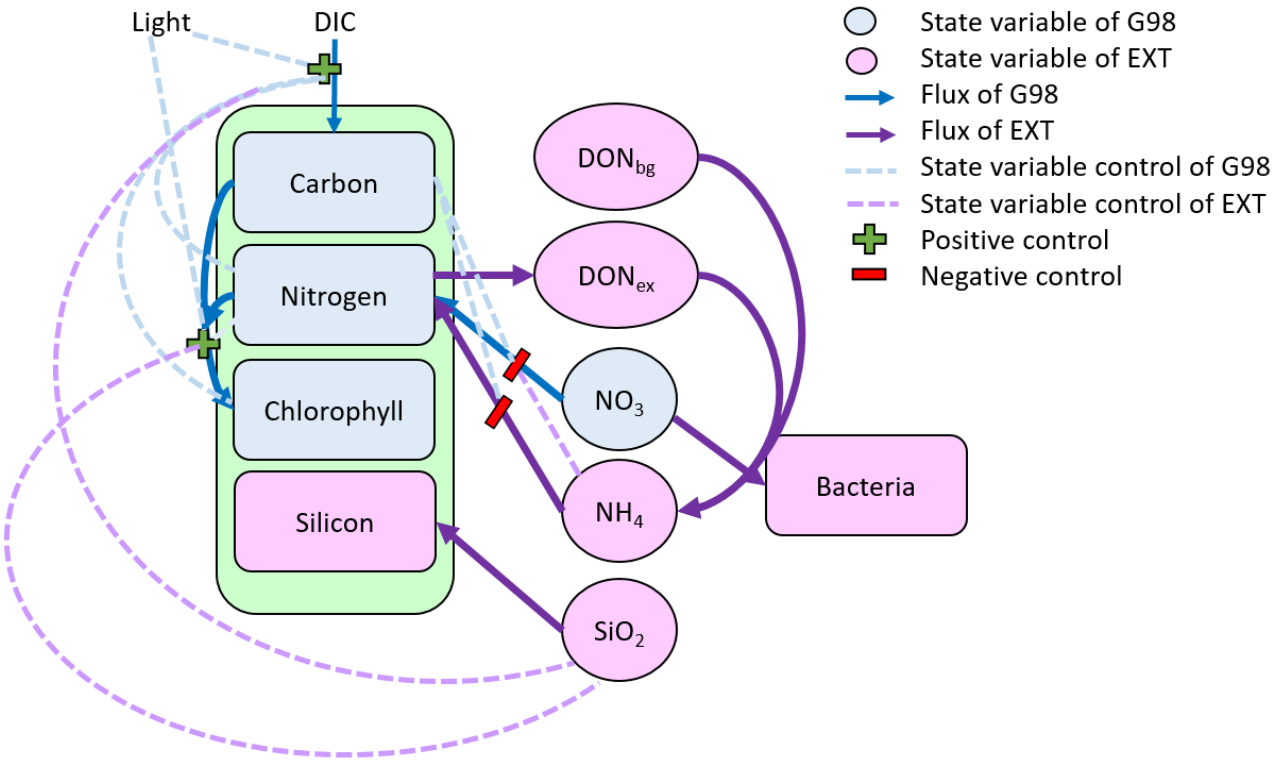

Figure 1. Schematic representation of the state variables and connections and controls in the G98 model (blue) and EXT model (purple). The EXT model has the same formulations as G98 with the additions shown in purple.”

The description of the model tuning needs to be clearer. It’s unclear why some parameters were picked for tuning while others weren’t (e.g. remineralisation parameters were not tuned), or what the rationale for picking the training data streams was (e.g. model ammonium was “loosely constrained” to observations). The text mentions several R packages used, but these are presented without any information about what they do, how they work, or what assumptions they make. For instance, is parameter space sampled by latin hypercube, genetic algorithm, or via local misfit gradient? There’s also an unclear distinction made around a “manual” component of this tuning exercise.

We added a detailed description of the model tuning, R packages, and parameter selection.

Changes:

2.3) “2.3 Model fitting

The model was written as a function of differential equations in R. All model equations are provided in the Appendix (Table A6) and the R code is available in the supplement. The differential equations were solved using the ode function of the deSolve package (Soetaert et al., 2010) with the 2nd-3rd order Runge-Kutta method with automated stepsize control. deSolve is one of the most widely used packages for solving differential equations in R.

Parameter of the G98 model were fitted to the BAC- experiment data and the EXT model was fitted to the BAC+ experiment data. The G98 parameter values were fitted first and retained without changes for the EXT model fitting. The maximum Chl:N ratio (θNmax), minimum and maximum N:C ratios (Qmin, Qmax), and irradiance (I) are given by the experimental data and needed no further fitting (Table A2). The start values and constraints for the remaining six variables (ζ, RC, αChl, n, Kno3, PCref, Table A3) were based on model fits of G98 to other diatom cultures in previous studies (Geider 1998, Ross and Geider 2009). The parameters were first fitted manually via graphical comparisons with the experimental data (POC, PON, Chl, DIN, Fig. 5 and 5), and via minimizing the model cost calculated as the root of the sum of squares normalized by dividing the squares with the variance (RMSE Eq. C2, Stow et al., 2009). The initial manual tuning approach allowed control of the model dynamics, considering potential problems with known limitations of the G98 model (e.g lag phase not modelled; Pahlow, 2005). The manual tuning also allowed obtaining good start parameters for the automated tuning approach and sensitivity/ collinearity analyses, which are sensitive to the start parameters.

After the manual tuning, an automated tuning approach was used to optimize the fits. The automated tuning was done using the FME package (Soetaert et al., 2010b), a package commonly used for fitting dynamic and inverse models based on differential equations (i.e. deSolve) to measured data. The automated analyses were based on minimizing the model cost calculated as the sum of squares of the residuals (SSR, Fitted vs measured data). The experimental data were normalized so that all normalized data were in a similar absolute range of values. This involved increasing Chl and PON values by an order of magnitude while decreasing DIN (NH4 + NO3) data by one order of magnitude. The data were not weighted, assuming equal data quality and importance. Prior to the automated fitting, parameters were tested for local sensitivity (SensFun) and collinearity, or parameter identifiability (collin; e.g. Wu et al., 2014). sensFun tests for changes in output variables at each time point based on local perturbations of the model parameter. The sensitivity is calculated as L1 and L2 norms (Soetaert et al. 2009; Soetaert et al., 2010b). The sensFun output is further used as input for the collinearity, or parameter identifiability analyses. Parameters were considered collinear and not identifiable in combination with a collinearity index higher than 20 (Brun et al., 2001). In this case, only the more sensitive parameter was used for further tuning. Eventually, RC, Kno3, n, and αChl were subject to the automated tuning approach using the modfit function, based on minimizing the SSR within the given constraints. Parameters were first fitted using a Pseudorandom search algorithm (Price, 1977) to ensure a global optimum. The resulting

parameters were then fine-tuned using the Nelder-Mead algorithm (Soetaert et al., 2010b) for finding a local optimum. A model run with the new parameters was then compared to the initial model via graphical comparisons of the model fit to the experimental data, and via the RMSE value.
The parameter values obtained for the G98 fit to the BAC- experiment were retained without changes or further fitting in the EXT model. The additional parameters of the EXT model were then fitted to the BAC+ experimental data (POC, Chl, PON, DIN). The model was only fitted to total DIN, due to the potential uncertainties related to ammonium immobilization in the biofilm. In fact, a test run, fitting the EXT model to NO3 and NH4 separately lead to a substantially worse overall fit (RMSE=8.79). Otherwise, the data were not weighted. Since the aim of the study was to model the effects of silicate and bacteria on algae growth and not to develop an accurate model for bacteria biomass and silicate concentrations, the parameters μbact, bactmax, Ksi, and Vmax were only fitted to the corresponding data (Bacteria, Silicate) prior to fitting the other parameters of the EXT model. Bacterial growth parameters (μbact, bactmax) were fitted to the bacterial growth curve. Silicate related parameters (Ksi, Vmax) were constrained by the study of Werner (1978) and fitted to the measured silicate concentrations. The remaining parameters were subject to the tuning approach described for G98. Ammonium related parameters (Knh4, nh4thres) were constrained by measured ammonium concentrations, and constants available for other diatom taxa described by Eppley et al. (1969). Remineralization parameters for excreted (rem) and background (remd) DOM were constrained by the data with the limitation of rem > remd, assuming that the excreted DOM is more labile. The parameters related to the effect of silicate limitation on photosynthesis and chlorophyll production (smin, SiPS) were constrained by the study of Werner (1978) and fitted as described for G98. None of the added parameters were collinear/ unidentifiable or given by the measured data and thus retained for the automated tuning approach. Eventually, the 15 parameters (Table A3) were fitted against 160 data points (Table A1)."

On a point related to tuning, I noted that the model has a key parameter for restricting phytoplankton growth (by 80%) in the absence of silicate, but that this parameter is not included in the tuning, which seems something of an omission (and, on a more presentation level, is hard-wired into the equations as a number rather than a parameter).

We agree that this parameter should be included in the tuning process since there may be variations from the study where this parameter is based on depending on the species and environment. It would also be interesting to include it in the tuning exercise to test if the 80% reduction can be confirmed after rigorous model tuning. We changed the model formulation and number of parameters in the tables accordingly and did the model fitting and sensitivity analyses again. The best fit is still an approximately 80% reduction.

Changes:

2.2) "Werner (1978) found that silicate limitation can lead to a 80% reduction in photosynthesis and a stop of chlorophyll synthesis in diatoms within a few hours. Hence, we added a parameter for the

reduction of photosynthesis under silicate limitation (SiPS) and formulated a stop of chlorophyll synthesis under silicate limitations."

3.2) "The most sensitive added parameters in EXT were the remineralisation rate of refractory DON (remd, L1=0.24), the half saturation constant for ammonium (Knh4, L1=0.1) and the inhibition of photosynthesis under Si limitation (SiPS, L1=0.07), which was comparable to other sensitive parameters of the G98 model (Qmax, RC, αChl, ζ, n, I, ΘNmax, Table A1)."
4.2) "we modelled the response of diatom growth to silicate limitation by reducing photosynthesis through a parameterized fraction (SiPS) and a stop of chlorophyll synthesis below a certain threshold, based on experimental studies (Werner, 1978; Gilpin et al., 2004) and in accordance to other ecosystem scale approaches. Automated fitting showed the same 80 % reduction of photosynthesis as described by Werner (1978)."

Table A7, 1b)

$$\frac{dC}{dt} = Si_{PS}(P^C - \zeta V_N^C - R^C - xf)C = \mu C$$

Finally, the authors identify three central hypotheses in their study:

1. Bacterial regeneration of ammonium will extend a phytoplankton growth;
2. Silicate or nitrogen limitations have different physiological responses;
3. A simple experiment can adequately represent Arctic spring bloom dynamics.

On the first, the model has a very poor performance replicating the time history of ammonium concentrations. On the second, this study would be more convincing if the concentrations of Si and N had been experimentally manipulated to enhance / diminish limitation of each. On the third, the model's inconsistent fit with observations, and its omission of significant real world factors (e.g. zooplankton) make it difficult to evaluate whether this is true. And because the model is only being run for the short incubation period (i.e. rather than beyond the incubation period, or in some mode investigating more realistic or extrapolated settings), it's not clear how it behaves when "unleashed".

Overall, I very much like the combined laboratory and modelling approach, but judge that the modelling component in particular needs to be made much clearer, and evaluated more critically.

We agree that the hypotheses are not perfectly addressed with the data and model, due to the resons mentioned and reformulated the hypotheses in the following way:

We hypothesize that: I) Bacterial regeneration extends a phytoplankton growth period and gross carbon fixation; II) Diatoms continue photosynthesis under silicate limitation at a reduced rate if DIN is available; III) Cultivation experiments are powerful for understanding the major spring bloom dynamics.

1. Bacterial regeneration extends a phytoplankton growth period and gross carbon fixation
2. Diatoms continue photosynthesis under silicate limitation at a reduced rate if DIN is available
3. Cultivation experiments are powerful for understanding the major spring bloom dynamics

Each hypothesis can be tested by the cultivation experiment and can be discussed and evaluated in more detail with the modelling approach.

Concerning hypothesis 1 we suggest that the poor fit to ammonium is mainly related to the measurements rather than the model. Ammonium is most likely immobilized in the biofilm via adsorption to the EPS and accumulation in pockets unavailable to diatoms (See response to Referee #3). These immobile NH4 pools are still part of the measured data. With the model assuming all NH4 being available for algae growth, this is a problem. Hence, we did not put a strong weighting on ammonium for the fitting routines but fitted the parameter to DIN instead. We did try to fit the model with heavy weighting on ammonium, but could still not reproduce the high ammonium concentrations in the stationary phase, while having a substantially worse fit for the other measured variables (RMSE=8.8).

Response to Referee #3:

"This could, in particular, explain the high values of measured NH4 compared to the model results as shown in Figure 5c. In addition, the higher values of measured NH4 could be explained in terms of a potential pH dependence of NH4+ adsorption to the EPS in terms of the pKa values of NH4+ and carboxylic groups, which belongs to the acidic polysaccharides as a fraction of EPS:

- Carboxylic groups have a pKa < 5, i.e. far away from seawater pH ~ 8, which means that they are always in the deprotonized negatively charged form R-COO- in seawater.
- NH4+ has a pKa ~9 closer to seawater pH.
- Thus, the NH4+/NH3 ratio will be higher in more acidic microenvironments (pH ~7.5-8).
- Thus, a lower pH due to bacterial respiration would increase the concentration of NH4+ in comparison to the bulk medium, which results in a higher immobile NH4 pool due to adsorption to the EPS.
- This could explain the higher discrepancy between modelled and measured NH4+ values in the experiments with bacteria (as seen in Figure 5c)."

We also included a model run going beyond the experimental time frame in the supplementary material. Overall, the model reaches stable state after approx. 20 days when all nutrients are used up. Bacterial regeneration can keep some levels of N and C assimilation going beyond the loss for excretion and maintenance respiration, but they do not build substantially more biomass, which would be expected in the environment, where, however, sinking and grazing would lead to an additional export leading to a net

loss. In order to keep the keep the manuscript streamlined, would prefer not adding these plots to the main manuscript, but to the Supplement instead. In the main manuscript, we suggest adding a short statement of the models stability if run longer (stable state after all nutrients are used up).

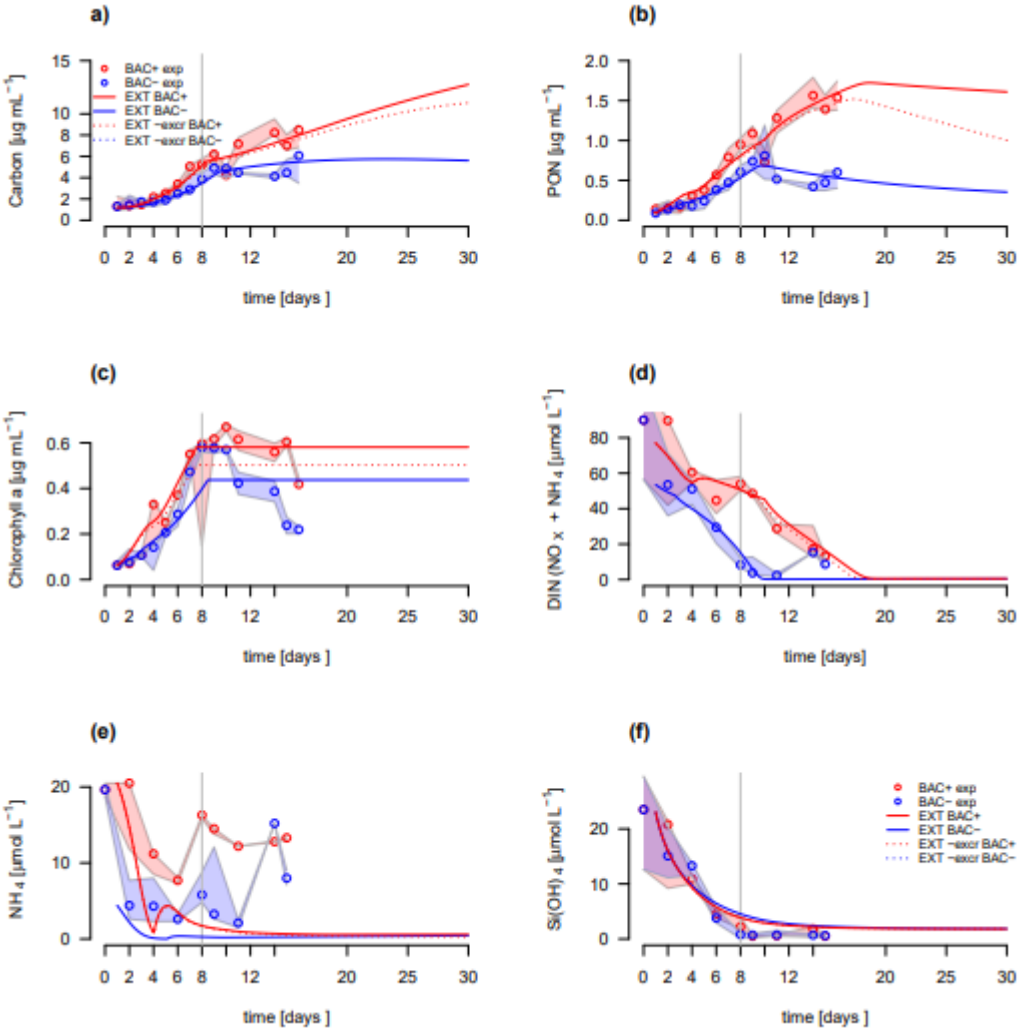

Figure S1: Model fit of the EXT model to the BAC- (blue) and BAC+ (red) experiment. Circles show median values and the colored polygons show the minimum and maximum of the measured data (n=3). Solid lines show the model outputs extended to 30 days of a) POC, b) PON, c) Chl (including outlier at day 8 in BAC+), d) NOX, e) NH_4, and f) Silicate. The dotted line show the output of EXT without excretion.

Specific comments

Pg. 1, ln. 20: "neglect" or "simplify"?; the distinction is important

We changed it to the term “simplify” since a general regeneration component is common in most models. When using the term “neglect” we were mostly focusing on a regeneration component dependent on bacteria biomass which is typically neglected in favor of a general purely substrate dependent remineralization formulation.

Pg. 1, ln. 23: surplus "and"

We removed the surplus “and”

Pg. 1, ln. 25: regarding the importance of organic matter excretion, was this based on observational evidence?

Yes, the excretion is based on the observation of algae aggregation in the stationary phase. However, since this is not the strong part of the model, we removed this statement from the abstract. We also added a more detailed discussion of the biofilm formation and implications for the model as described below and in more detail in the response to reviewer 3.

Pg. 1, ln. 26: “model complexity is comparable to other ecosystem models” – this is misleading as the model here is really an incubator model and not an ecosystem model; it’s missing most of the components that such models include (e.g. detritus, zooplankton)

When comparing model complexity (or number of parameters), we only compare the phytoplankton growth compartment within the ecosystem scale models, which are comparable to our extended model. We omit the complexity in ecosystem scale models not part of our model (e.g. Zooplankton, detritus). We clarified this by following change:

“Overall, model complexity (number of parameters) is comparable to the phytoplankton growth and nutrient biogeochemistry formulations in common ecosystem models used ...”

Pg. 2: maybe be a little clearer on the distinction between autotrophic and heterotrophic bacteria throughout; cyanobacteria, for instance, are unlikely to play the role that’s described as "bacterial" here

We distinguished heterotrophic and autotrophic bacteria clearer by adding the term heterotrophic to the bacteria, we are discussing for remineralization. We also agree that the term phototroph for cyanobacteria and heterotroph for the cyanobacteria associated bacteria is especially important to avoid confusion. However, we suggest adding the term heterotrophic only to the first occurrence of bacterial regeneration to keep the text readable.

Pg. 2: also, you should probably say something about the role of zooplankton; they graze phytoplankton and excrete some of the nitrogen they acquire; how quantitatively

important is this here?; (I've added a cite to a paper that hints that they might not be all that important)

We added a section about zooplankton with a statement that their importance is low for regenerated production, compared to bacteria regeneration citing additional literature including the suggested paper doi:10.1016/j.dsr.2012.10.003

Change:

"Zooplankton grazing is typically of low importance for terminating blooms (e.g. Saiz et al., 2013), while inorganic nutrients are considered driving bloom termination (Krause et al. 2019, Mills et al. 2018)."…"Zooplankton may also release some ammonium after feeding on phytoplankton, but we suggest that this process is likely far less important than bacterial regeneration (e.g. Saiz et al., 2013). Previously measured ammonium excretion of Arctic mesozooplankton is typically low compared to bacterial remineralization (Conover and Gustavson, 1999), with the exception for one study in summer in a more open ocean setting (Alcaraz et al., 2010). "

Pg. 2, ln. 36: "marine phytoplankton *are*"?

We corrected the term accordingly

Pg. 2, ln. 41: predictions of what?

Corrected to: "predicitons of primary production with climate change"

Pg. 2, ln. 47: you might want to cite something like doi:10.1016/j.dsr.2012.10.003 as evidence of the reduced role of mesozooplankton in controlling / terminating blooms

As described above, we added following statement:

"Zooplankton grazing is typically of low importance for terminating blooms (e.g. Saiz et al., 2013), while inorganic nutrients are considered driving bloom termination (Krause et al. 2019, Mills et al. 2018)."

Pg. 2, ln. 48: remineralisation of what?; a bit of clarity would be helpful; dead diatoms, TEP, faecal material, etc.?

Clarified in the following way: "remineralisation of organic matter"

Pg. 2, ln. 57: heterotrophic bacterioplankton?

Yes, we clarified it by using the term "heterotrophic bacterioplankton" (See also response above).

Pg. 2, ln. 60: regarding "neglected", do you mean omitted or simplified?; most models include remineralisation of detrital material, and this implicitly bacterial

We are mainly referring to culture based experiments such as Ross and Geider 2009, and Flynn 2001, where remineralisation is omitted. We changed the term to "simplified or omitted".

Pg. 2, ln. 63: cultivation experiments normally provide parameter values for things like

maximum rates of processes, half-saturations, etc., so it's not clear this is problematic; if model tuning is using cultivation experiments at equilibrium then this might be more of an issue

Problematic is that obtaining axenic algae cultures is challenging, not possible for most species, and does usually not last long (See our response to bacteria growing in the axenic treatment below). In previous cultivation experiments, no efforts for obtaining axenic cultures were mentioned, which hints to bacteria contaminated cultures. In these cultures, regeneration of e.g. ammonium and phosphate takes part. If half saturation constants, maximum uptake rates e.g. are based on non-axenic cultures with the assumption of the absence of regeneration, the values are likely too high, since the experiment will have a nutrient source not accounted for, which leads to underestimations of nutrient uptake, or in the worst case overestimation of growth efficiency if ammonium is not measured at all.

Changes:

"These latter models have been often developed and tuned based on cultivation experiments in which microbial remineralization reactions were assumed to be absent (e.g. Geider et al., 1998; Flynn, 2001) despite the fact that most algae cultures, likely including Geider et al., (1998) and Flynn (2001) are not axenic. Parameters estimated by fitting axenic models on non-axenic experiment may be misleading, mostly by an inflated efficiency of DIN uptake."

Pg. 3, ln. 72: this process was well-known long before this citation (Flynn, 1997); dig a bit deeper

We chose the citation by Flynn (1997) due to the modelling component in the paper, but agree that we should cite earlier literature. Thus we included the review by Morris (1974) and the review by Dortch (1990).

Dortch, Q.: The interaction between ammonium and nitrate uptake in phytoplankton, Marine ecology progress series, 61(1), 183-201, 1990.

Morris, I.: Nitrogen assimilation and protein synthesis, Algal physiology and biochemistry, 10, 1974.

Pg. 3, ln. 72: "iron has a strong control on silicate uptake" - I'm not sure that this is quite right; Si:C ratios are affected by Fe availability, but this is through continuing Si uptake but reduced C/N uptake and no cell division; my understanding is that Si *uptake* (within a certain range of Si:C) is not immediately affected by Fe; also the recent source given for this statement, Hohn et al., 2009, is a modelling PhD thesis

We agree that the control of Fe on Si is controversial and not well documente din earlier literature and that a modelling PhD thesis is not the best support for this hypothesis. Thus, we changed the statement in the following way:

Change:

"…C and N uptake is reduced under Fe limitation, while Si uptake continues, leading to increasing Si:C/N ratios (Werner, 1977; Firme et al., 2003),…"

Pg. 3, ln. 75: "ultimately too complex" - they add computational expense to large-scale ecosystem models; it's not clear that they are "too complex" (or even what is meant by this)

We changed it to "ultimately too computationally expensive when implemented in a global biogeochemical model"

Pg. 3, ln. 79: is phosphate limiting in the Southern Ocean?; in parts of its northern extent, yes, but in the south its concentrations are high, no?
Pg. 3, ln. 79: "coastal"?; is there a distinction to be drawn with deep Arctic locations?

We clarified the sentence by: 1) removing the statement about phosphate, which is indeed only limiting in the northern parts of the Southern Ocean, 2) by removing "coastal", since Fe is not limiting and DIN is the primary limiting nutrient in most Arctic marine systems and by 3) adding supportive citations.

Change:

"In contrast to Antarctica, DIN is the primary limiting nutrient and iron is not limiting in most Arctic systems (Tremblay and Gagnon, 2009; Moore et al., 2013)"

Pg. 3, ln. 81: yes and no; if simple lab experiments exclude factors such as zooplankton excretion which might help fuel phytoplankton growth in parallel with bacterial remineralisation, then it is questionable that they are demonstrating something that's important in the real ocean

We agree that zooplankton N excretion may have an additional role, but as mentioned above, argue that bacterial N regeneration is quantitatively more important. However, we relativized the sentence in the following way:

Change

"While simple lab experiments cannot represent all nutrient dynamics found in the environment (e.g. N excretion by zooplankton), they can focus on the quantitatively most important dynamics, to facilitate the development of simple, but accurate multinutrient models scalable to larger ecosystem models."

Pg. 3, ln. 86: how "associated" is this?; is it something that lives in direct physical contact or shares the same waters?

The bacteria cultures were obtained from the diatom culture directly plated onto LB agar plates. This means they grew together with the diatom outcompeting other bacteria in an environment heavily influence by the algae, which was the only carbon source to the system. Microscopy showed bacteria attached to the diatoms (mostly in the stationary phase), but mostly free-living. However, since the bacteria are heterotroph and there was no other carbon soruce than the DOM coming from the diatom culture, we see them as associated.

Change:

"…inoculation with bacteria cultures, isolated beforehand from the non-axenic culture."

Pg. 3, ln. 87: again, what specifically is the issue with complexity?; is it model cost, or is there some other aspect of complexity that disfavours inclusion in large-scale models?

Yes, it is model cost. We changed it to: "... aiming to keep the number of parameters, and computational costs low to allow its use in larger ecosystem models."

Besides the computational costs a large number of parameters, as used in detailed physiological models, is also more difficult to tune or verify with experimental/environmental data, which leads to the issue of overfitting.

Pg. 3, ln. 93: good hypotheses!; however, you do not clearly return to them (e.g. "Regarding the hypotheses framed for this study . . .")

As mentioned above, we changed the hypotheses in the following way:

We hypothesize that: I) Bacterial regeneration extends a phytoplankton growth period and gross carbon fixation; II) Diatoms continue photosynthesis under silicate limitation at a reduced rate if DIN is available; III) Cultivation experiments are powerful for understanding the major spring bloom dynamics.

Pg. 4: this all sounds good, but my expertise in laboratory work is very limited

Thank you

Pg. 4, ln. 111: just for simplicity in the labelling, you might want to come up with nice short names for these experiments; e.g. BACT- (for the axenic) and BACT+ (for the non-axenic), or similar; this will make it easier to refer to the experiments in clear, non-wordy ways later on

We appreciate the suggestion and used the abbreviations BAC- and BAC+ throughout the corrected manuscript and figures.

Pg. 5, ln. 143: the origin of the f-ratio should be cited so that less familiar readers can understand what it is

We added following citation:

Eppley, R. W.: Autotrophic production of particulate matter, Analysis of marine ecosystems/AR Longhurst, 1981.

Pg. 5, ln. 144-146: this is a little confusing; perhaps spell it out with equations?
We added following equation:

"Equation C1. F-ratio estimation in the cultivation experiments with the average PON concentrations at day 13 to 15 ($PON^{d13\text{-}15}$) for the BAC- and BAC+ treatments."

$$f-ratio = \frac{PON_{BAC-\ \ d13-15}}{PON_{BAC-\ \ d13-15} + PON_{BAC+\ \ d13-15}}$$

Pg. 5, section 2.2: I don't think it ever hurts to have a schematic of a model's dynamics to supplement equations and (especially) verbal description
We added a schematic of the model dynamics of G98 and the extended model and briefly described the main dynamics focusing on the controls of photosynthesis, nitrogen assimilation and chlorophyll synthesis by C:N and Ch:N ratios, DIN concentrations, and light.

The schematic figure is shown above (Fig. 1). The following details, were added to the text:

Change:
"The Geider et al. (1998) model (G98) describes the response of phytoplankton to different nitrogen and light conditions and is based on both intracellular quotas and extracellular dissolved inorganic nitrogen (DIN) concentrations, allowing decoupled C and N growth (Fig. 1). Within this model, light is a control of photosynthesis and chlorophyll synthesis. C:N ratios and DIN concentrations control nitrogen assimilation, which is coupled to chlorophyll synthesis and photosynthesis. Chl:N ratios are controlling photosynthesis and chlorophyll synthesis."…"The EXT model keeps all formulations of the G98 and adds dynamics and interactions of silicate, nitrate and ammonium uptake, carbon and nitrogen excretion and bacterial remineralisation (Fig. 1)."…

Pg. 5, section 2.2: similarly, this section would be a lot clearer if you spelled out which models you were using, and ensured that the later plots use the same nomenclature; I initially misread the work ; I reckon it's: 1. G98; 2. Extended; 3. G98 – excretion; 4. Extended – excretion
We clarified the model runs used in the manuscript and used consistent nomenclature: 1. G98, 2. EXT (by default with excretion) 3. $EXT_{\text{-excr}}$. G98 does not have an excretion compartment.

Change:

"We used a dynamic cell quota model by Geider et al. (1998) to describe the BAC- experiment (G98). We then extended the G98 model to represent the role of silicate limitation, bacterial regeneration of ammonium, and different kinetics for ammonium and nitrate uptake (EXT) and fitted it to the BAC+

680 experiment while retaining the parameter values estimated for G98."…" For testing the importance of DON excretion we also ran the EXT model without DON excretion (EXT–excr)."

Pg. 5, section 2.2: stating up front an outline about the modelling strategy might help
685 (i.e. two models, tuned to the lab work, DOM addition, etc.)
We added a summary of which models were used for which experiment in the beginning in the now estensive chapter describing the fitting routines.

Change:
690

"Parameter of the G98 model were fitted to the BAC- experiment data and the EXT model was fitted to the BAC+ experiment data. The G98 parameter values were fitted first and retained without changes for the EXT model fitting."

695 Pg. 5, ln. 149: some model equations by the looks of things; the model description appears incomplete
We added the missing equation and double-cheked for any other incomplete model descriptions.

Changes in Table:

7a)	Silicate uptake *(Monod kinetics after Spilling et al., 2010)*	$\frac{dSi_d}{dt} = V_S^C = \left(V_{max} Si_d \frac{Si_d - S_{min}}{K_{si} S_{min}}\right) C$
7b)		$\frac{dSi_p}{dt} = \frac{-dSi_d}{dt} 14$

700

Pg. 6, ln. 164: equation for dSi seems missing in appendix
We added the missing equation in Table 7

$$\frac{dSi_d}{dt} = V_S^C = \left(V_{max} Si_d \frac{Si_d - S_{min}}{K_{si} S_{min}}\right) C$$

705

Pg. 6, ln. 166: "80% reduction" - is this where the 0.2 in the equations comes from (i.e. 1 - 0.8 = 0.2)?
Yes, we clarified it in the following way: "Werner (1978) found that silicate limitation can lead to a 80%
710 reduction in photosynthesis and a stop of chlorophyll synthesis in diatoms within a few hours. Hence, we

added a parameter for the reduction of photosynthesis under silicate limitation and formulated a stop of chlorophyll synthesis under silicate limitations."

However, we changed the fixed 80% value to a tunable parameter and rerun the fitting routine and sensitivity analysis as described above.

Pg. 6, ln. 167: some syntheses would suggest that N dynamics *are* coupled to Si dynamics: e.g. Martin-Jezequel, V., M. Hildebrand, and M. A. Brzezinski, Silicon metabolism in diatoms: Implications for growth, J. Phycol., 36, 821 – 840, 2000.

We argue that N dynamics are not directly coupled to Silicate limitation, but indirectly via reduced photosynthesis and inhibited chlorophyll production. The reference by Martin-Jezequel shows no direct coupling of N and Si, but overall different controls for Si and N/P, where Si is tighly linked to the cell cycle, fueled by heterotrophic respiration, while N/P are controlled by photosynthesis. Overall, Martin-Jezequel et al. supports our assumption of decoupled Si and N metabolism and is included in the manuscript as additional support:

Change:

"N and Si metabolism have different controls and intracellular dynamics, with N uptake fuelled by photosynthesis (as PCref in G98) and Si mainly fuelled by heterotrophic respiration (Martin-Jezequel et al., 2000). In general, we assume that nitrogen metabolism is not directly affected by silicate limitation (Hildebrand 2002, Claquin et al., 2002), but we expect cellular ratios to be affected by reduced photosynthesis and chlorophyll synthesis under Si limitation (Hildebrand, 2002; Gilpin, 2004)."

However, we acknowledge that there is 1 study by Gilpin et al., 2004, discussing a coupling of N:Si. Hence, we added it in the discussion:

Change:

"Studies on the coupling of silicate limitation on C, N, and Chl show inconclusive patterns, including a complete decoupling (Claquin et al., 2002), a relation of N to Si (Gilpin et al., 2004) and reduction of photosynthesis (Werner, 1978; Gilpin et al., 2004) while no new chlorophyll is produced (Werner, 1978; Gilpin et al., 2004)."…" Our cultivation study shows"…" ii) that coupling of Si:N:C:Chl is present. We do not expect a direct Si:N coupling, due to different controls of Si and N metabolism (Martin-Jézéquel et al., 2000.), but suggest indirect coupling via reduced photosynthesis."

Pg. 6, ln. 171: make it clear here that your model has labile and refractory DON

We agree that this needs to be clarified.

Change:

"It was assumed that this process is faster for freshly excreted DON compared to DON already present in the medium. Thus, we implemented a labile (DONl) and refractory (DONr) DON pool with different remineralization rates (rem, remd)."

Pg. 6, ln. 173: it seems unlikely that the bacteria would simply "give up" on remineralisation if the C:N ratio is too high; perhaps expand on why Tezuka suggests this is happening

We do not suggest a complete stop of remineralisation, but a net release of nitrogen of 0, since bacteria need more DIN on their own, rather than having the luxury of releasing it to the environment. We mention now two papers as support. Both papers base their fining on net changes in DIN. We tried to clarify it by following change:

Change:

"After Tezuka (1989), net bacterial regeneration of ammonium occurs at DOM C/N mass ratio below 10 and is proportional to bacterial abundances. Higher thresholds up to 29 have been found (e.g. Kirchmann, 2000), but we selected a lower number to stay conservative."

Pg. 6, ln. 175: this is unclear; when you say "substrate" what do you mean?; typically substrate is used to indicate a resource consumed by an organism; here you're talking about phytoplankton, so DIN and DIC would appear to be meant - but DIC will likely be much higher than 10x DIN

We refer to DOM as substrate for bacteria and clarified it: "DOM C/N ratios…. "

Pg. 6, ln. 177: does this mean that bacteria won't remineralise material with a C:N > 10?; that seems a little unlikely

As for line 173 we changed "bacterial remineralization" to "net bacterial ammonium regeneration".

Pg. 6, ln. 178: as the paper makes a fuss earlier about other models glossing over bacterial remineralisation, this simplified form is surprising

The main improvement of the model is to include a remineralisation rate controlled by: i) bacteria biomass, ii) substrate (DOM) C:N ratios, and iii) substrate origin (autochthonous, allochthonous). Other models typically have a fixed remineralisation rate either only dependent on the DOM/POM, or not controlled by any environmental variable. Thus, we still see our extension as a considerable improvement and consider a simple logistic growth estimate sufficient.

We could of course model bacteria growth via Michaelis-Menten kinetics based on 2 DOM pools, but this would not have any effect on the parameterization or modelling of algae physiology, which is the main goal of the paper, while increasing the number of parameters and computational costs, which we tried to keep low. Since, the aim of the model is not to model bacteria growth, but algae growth and intracellular C:N:Chl ratios we do not see that a more accurate and more complex model of bacteria growth would improve the manuscript.

Pg. 6, ln. 185: Table A6 - it looks to me like some equations are missing

We added the missing equations as mentioned above.

Pg. 6, ln. 186: which order of RK?; e.g. 3 or 4 (or higher)

We used the 2nd-3rd order Runge-Kutta method with automated stepsize control and added this information in the manuscript.

"The differential equations were solved using the ode function of the deSolve package (Soetaert et al., 2010) with the 2nd-3rd order Runge-Kutta method with automated stepsize control."

Pg. 6, ln. 185-191: this description of model tuning is far too brief; I'm not sure what's going on here; readers unfamiliar with R (I am one) will not understand what these different packages are doing or what their underlying assumptions are; this aspect of the modelling is too important to be glossed over so quickly; in general, to avoid the appearance of having just used the first package that occurred to you, expanding on the detail of the tuning (tools, approach, goal) would greatly benefit this description (hence and on Pg. 7)

We added a more detailed and extensive description of what the R packages are doing. deSolve is the most widely used solver for differential equations in R, and FME is a package for model fitting and sensitivity analysis developed as add on to deSolve. The tuning approaches via

1st manual fitting (based on RMSE error and graphical comparisons),

2nd automated fitting of selected parameters (avoiding collinearity/ linear dependence of sensitivity of 2 parameters = unidentifiable parameters), and choosing the more sensitive parameter in case of conflicts) via the Pseudorandom algorithm (searching for a global optimum),

3rd fine tuning for a local optimum using the Nelder Mead algorithm. 4th check if the new parameters give a better fit regarding graphical comparisons and RMSE.

See the added chapter above in this response.

Pg. 6, ln. 192: the text should be clear on which observed variables were used to fit the model, why these were favoured, and whether any weighting was made to account for those judged better observed or more important; I would naively expect nutrient concentrations to be of prime importance but it's unclear what criteria the authors are using here (see my later remark about ammonium)

We also added more details about the observed variables used for tuning. We used POC, PON, Chl, and DIN (NOx + NH4) with standardized values (POC, 10xPON, 10xChl, DIN/10) and no further weighting. Due to rather poor quality of the NH4 data, we did not fit the model to NOx and NH4 separately. Details about the parameters tuned and the constraints are also given in Table A3. Parameters were partly given by measured data, or tuned after constraining with measured or published constraints. In case of strong collinearity, only the most sensitive of the collinear parameters was tuned.

See the added chapter above in this response.

Pg. 7, ln. 197: this seems rather unsatisfactory; I would expect parallel runs with the same parameter values to be performed for axenic and not-axenic simulations, with an automated process (e.g. a genetic algorithm) to evaluate cost (i.e. misfit) before somehow generating new parameter values and iterating; having a manual component seems odd

We did do parallel runs with the same parameter values. The G98 model was fitted to BACT- data, but the resulting parameters were used for a G98 model run of both BACT- and BACT+ and kept without changes or further fitting as part of the EXT model. For the EXT model only the extended parameters relevant for describing our key observed variables (POC, PON, Chl, DIN) were fitted with the same rigorous fitting approach used for G98. The resulting parameters were used for the model of both BACT+ and BACT-.

We corrected the text in the manuscript to clarify what we did

We argue for an initial manual tuning approach in order to account have control of the model dynamics and to obtain good start parameters for the automated tuning approach and sensitivity/ collinearity analyses.

See the added chapter above in this response.

Pg. 7, ln. 198: what are these "known limitations"?; also, it's noticeable in the plots that the model solutions inflect strongly around the lag/stationary phase time point - is the model somehow different either side of this division?

The known limitations is that parameter tuning of the G98 in earlier attempts did not allow modelling the lag phase (Pahlow, 2005); later, howerver, Smith and Yamanaka (2007) showed that the Geider model can be brought to reproduce an initial lag phase.

The strong change around the beginning of the stationary phase is based on the threshold based approach to responses of Photosynthesis and Chl synthesis after Silicate limitation. Once silicate falls below a threshold, the physiology changes, which can be seen as a sudden change. Threshold based approaches are common in other dynamic models (e.g. threshold for cell division in Ross & Geider, 2009, threshold deciding which limiting nutrient decides the growth in Vichi et al., 2007).

Pahlow, M. (2005). Linking chlorophyll-nutrient dynamics to the Redfield N: C ratio with a model of optimal phytoplankton growth. Marine Ecology Progress Series, 287, 33-43.

Smith, S. L., & Yamanaka, Y. (2007). Quantitative comparison of photoacclimation models for marine phytoplankton. Ecological Modelling, 201(3–4), 547–552. https://doi.org/10.1016/j.ecolmodel.2006.09.016

See also the added chapter above in this response.

Pg. 7, ln. 202: “Colinearlity” - do you mean that you’re looking for linkages between parameters here?

Collinearity is a measure for the parameter identifiability in complex simulation models (Brun et al., 2001) and allow identifying which parameter(s) (sets) can be uniquely constrained from the data.
If the perturbation of two different parameters can lead to the same change in the output variables, they are collinear, which makes them unidentifiable. Parameters were considered collinear and not identifiable in combination with a collinearity index higher than 20 as described in (Brun et al., 2001). In this case, only the more sensitive parameter was fitted.

Brun, R., Reichert, P. and Kunsch, H. R., 2001. Practical Identifiability Analysis of Large Environmental Simulation Models. Water Resour. Res. 37(4): 1015–1030.

See the added chapter above in this response.

Pg. 7, ln. 205: ammonium was "constrained loosely" - perhaps given later results this was a mistake?

Due to potential uncertainties associated with the ammonium data (e.g. immobilization in the biofilm by adsorption and micro-pockets, leakage of intracellular NH4 during filtration, freeze-thaw cycle), and high variability in published parameters (e.g. Eppley et al., 1969), we used wider constraints for ammonium related parameters. We do not agree that narrower constraints would lead to a better model fit to ammonium, since the new values would be within the same parameter space/ constraints. However, for consistency and usability of the model in other settings we narrowed down the constraints of published half saturation constants by Eppley et al., 1969. The reason for the poor fit is partly the lower weighting of the ammonium output during model fitting (We only fitted to DIN (NH4+NO3) and not separately to NH4 and NO3, but also the uncertainty of the ammonium values which likely include immobilized ammonium from algae cells, and the biofilm. We did a test run where we fitted the EXT model to POC, PON, Chl, NH4 and NO3, but the overall fit was substantially worse (RMSE = 9, instead of 2 with the DIN fit) with parameter values reaching the limits of the constraints.

Eppley, R. W., Rogers, J. N., & McCarthy, J. J. (1969). HALF-SATURATION CONSTANTS FOR UPTAKE OF NITRATE AND AMMONIUM BY MARINE PHYTOPLANKTON 1. Limnology and oceanography, 14(6), 912-920.

See the added chapter above in this response.

Pg. 7: ecosystem models have notoriously non-linear misfits in their parameter space; when this is highly multidimensional (as here) it can be difficult for optimisation to find the global minimum misfit; how has this been achieved here?

We agree that his is a potential problem and therefore we approached the problem from different angles. First, we started with extensive manual tuning, as this gives a lot of insight for the modeler on how an optimal fit can be achieved and which parameters influence the results the most.
Secondly, we applied an automated parameter fitting procedure, which started with a collinearity analysis to make sure we are working with a parameter set that can actually be identified from the data. This reduces the risk of getting stuck in a local minimum. Subsequently, we ran a pseudorandom optimization routine to ensure a better coverage of the (identifiable) parameter space to increase the chance of approaching the global minimum randomly. The automated optimization routine ended with a directed descent algorithm, i.e. the Nelder Mead algorithm, that ensures quick convergence to the minimum.

See the added chapter above in this response.

Pg. 7, ln. 220: "stationary phase" - how exactly defined here?; particularly in the context of Figure B3c, which shows chlorophyll concentrations peaking _2 days later in the bacterial incubations

We defined the stationary phase by the sudden increase in phosphate and ammonium, silicate and DIN (for axenic cultures) values falling below minimum values in the model, and the Quantum yield dropping below 0.63. Since the explanation of all of these evidence is spread over the page, we changed the term "stationary phase" to "day 8", which is less objective.

Pg. 8, ln. 234: can you explain if these values are meaningful, or is it just the relative values between phases that's important?

The Quantum yield is a ratio based on variable fluorescence of chlorophyll. The number ranges between 0 and 1 and show how efficiently energy is transported after adsorption. Generally, high numbers indicate fit and active cells, while low numbers indicate stressed algae cells. Low N:C ratios are one stressor described to lead to inefficient energy transfer (low QY, Cleveland and Perry, 1987).

Cleveland, J. S., & Perry, M. J.: Quantum yield, relative specific absorption and fluorescence in nitrogen-limited Chaetoceros gracilis. Marine Biology, 94(4), 489-497, 1987.

Change:

"The maximum photosynthetic quantum yield (Fv/Fm) is commonly used as a proxy of photosynthetic fitness (high QY), indicating the efficiency of energy transfer after adsorption in photosystem II. Low values are typically related to stress, including for example nitrogen limitation (Cleveland and Perry, 1987). We found an increase in QY from approx. 0.62 to 0.67 d-1 in the exponential phase and a decrease to approx. 0.62 in the BAC+ treatment after 8 days and to approx. 0.58 in the BAC- treatment (Table A8)."

Pg. 8, ln. 279: This seems a pretty serious deficiency given the focus of this paper; I would interpret this as potentially a problem at the tuning stage; did you consider weighting fitting ammonium more heavily?

We suggest that the poor fit to ammonium is mainly related to the measurements rather than the model (immobilized ammonium in the biofilm). Hence, we did not put a strong weighing on ammonium for the fitting routines but fitted the parameter to DIN (NO3 + NH4) instead. When we did try to fit the model with heavy weighting on ammonium, we could still not reproduce the high ammonium concentrations in the stationary phase, while having a substantially worse fit for the other measured variables (RMSE=8.8). We discuss this limitation in the manuscript as follows:

Changes:

"The model was only fitted to total DIN, due to the potential uncertainties related to immobilized ammonium in the biofilm. In fact, a test run, fitting the EXT model to NO3 and NH4 separately lead to a substantially worse overall fit (RMSE=8.79)."

"While not all ammonium measured is also available for algae growth, discussion of the dynamics (decrease in the start, increase with the onset of the stationary phase), especially if also shown in the EXT model, are still useful to understand multinutrient dynamics (e.g. regeneration). Considering the overall higher concentrations of NO3, compared to NH4, discussions of total DIN dynamics, DIN.DIP ratios, and limitations are also meaningful."

"Fine scale DIN dynamics caused by ammonium – nitrate interactions were represented well (Fig. 6a). However, at the onset of the stationary phase, ammonium concentrations of the model were one order of magnitude lower than in the experiment, showing a major weakness (Fig. 6c). Increased weighting of ammonium during the model fitting led to a slightly better fit to ammonium, but a substantially worse fit of the model to POC, PON, and Chl ($RMSE_{EXT}$=8.79), indicating that the problem lies with the ammonium data (immobilized ammonium)."

Pg. 9, ln. 283: "complexity" is an unusual way to describe a lack of sensitivity (which is what you seem to be suggesting); also, given the extended model performs no better (worse?) than the G98 model is this not to be expected?; i.e. you've added a means for the model to be different, but this means is far less powerful than what the model already has

We agree that complexity is not the best fitting term and changed it to "sensitivity" or "added parameters". Changed to: "…was more sensitive than any of the original model parameters. Hence, the added parameters of the extended…"

Change:

"The sensitivity analysis (Fig. B1, Table A1) revealed that the sensitivity of the added parameters in EXT is overall comparable to the sensitivity of the original parameters in G98. The model outputs were most sensitive to P_C^{Ref} (L1=0.8, L2=1.5), which is a parameter in both G98 and EXT. The most sensitive added

parameters in EXT were the remineralisation rate of refractory DON (rem_d, L1=0.24), the half saturation constant for ammonium (K_{nh4}, L1=0.08) and the inhibition of photosynthesis under Si limitation ($Si_{PS,}$ L1=0.08), which was comparable to other sensitive parameters of the G98 model (Q_{max}, R_C, α_{Chl}, ζ, n, I, $\Theta_N{}^{max}$, Table A1). Small perturbations of the parameters only indirectly related to the fitted output variables did not lead to changes in POC, PON, Chl, or DIN."

Pg. 9, section 4: this discussion seems far too long for what's quite a simple set of Experiments

We agree that we originally thought this was simple set of experiments. However, the additional model interpretation, though very valuable we believe, does warrant a lengthier discussion. Also the detailed and thorough reviews for this manuscript made it impossible for us to substantially shorten the discussion and we believe that shortening the discussion would not be possible while addressing all comments of the three reviewers. We still tried to keep it as short as possible with the suggested changes.

Pg. 10, section 4.1: there's nothing in here about the (hard-wired!) 80% adjustment to growth rates caused by low silicate; this appear to be an unchangeable assumption

Firstly, we changed the 80% formulation into a tuneable parameter. Secondly, we added a sentence in the discussion. "Photosynthesis was reduced by 80% after silicate became limiting, which is in accordance with earlier experimental studies (Tezuka..)."

Pg. 10, ln. 310: do values of the f-ratio from bottle experiments relate well to those measured from the open ocean?; I can't think of any reason to suspect that they will, not least because there are no nitrifying bacteria including in the cultures here

We do not expect that the f-ratio in our bottle experiment is representative for the open ocean, but compare the values as starting point for discussing why. We argue that a discussion of the differences between the bottle experiment and open ocean values (e.g. grazing, nitrification) can show the limitations of the experiment and thereby the limitations of our model. We also add a reference to nitrification to the lacking processes.

Change:

"While we do not expect the f-ratio in our bottle experiment to be directly comparable to open ocean system, which does include a variety of algal taxa beyond C. socialis, a comparison can aid to identify limitations in our experiment and model. Regenerated production is significant in polar systems and our estimated experimental f-value of 0.31 is slightly below the average for polar systems (Harrison and Cota, 1990, mean f-ratio=0.54). Nitrification is a process supplying about 50% of the NO3 used for primary production in the oceans, which may lead to a substantial underestimation of regenerated production (Yool et al., 2007), inflating the f-ratio interpreted as estimate for new production, potentially also in the study by Harrison and Cota (1990). The absence of vertical PON export in our experiment may be another explanation…"

Pg. 11, ln. 357: ah-ha, computational cost is finally mentioned

We also added this information in previous formulations of “cost” and “complexity” in the corrected version of the MS.

Pg. 11, ln. 351: I don’t think it’s ever made clear why there may be a preference for NH4 over NO3; it would be good to include mention of this so that readers understand why this aspect may be important in the work here

The conversion of NH4 to biomass NH3 is energetically much cheaper, making it the preferred source. We added following information:

“Due to the metabolic costs related to nitrate reduction to ammonium, ammonium uptake is preferred over nitrate, potentially leaving more energy for other processes (Lachmann et al., 2019). Ammonium can even inhibit or reduce nitrate uptake over certain concentrations (Morris, 1974). The dynamics are mostly controlled by intracellular processes, such as glutamate feedbacks on nitrogen assimilation, cost for nitrate conversion to ammonium, or lower half saturation constants of ammonium transporters (Flynn et al., 1997).”

Lachmann, S. C., Mettler-Altmann, T., Wacker, A., & Spijkerman, E.: Nitrate or ammonium: Influences of nitrogen source on the physiology of a green alga, Ecology and evolution, 9(3), 1070-1082, 2019.

Pg. 12, ln. 360: the authors note different conceptual models for the Si:N relationship in this section, but stick instead with a highly simplified approach from a review almost 40 years old; and also remove this relationship from the tuning exercise undertaken; I would expect to see more justification for this - or potentially some form of model sensitivity analysis to evaluate how important it is

We argue that N dynamics are not directly coupled to Silicate limitation, but indirectly via reduced photosynthesis and inhibited chlorophyll production. The reference by Martin-Jezequel shows no direct coupling of N and Si, but overall different controls for Si and N/P, where Si is tighly linked to the cell cycle, fueled by heterotrophic respiration, while N/P are controlled by photosynthesis. Overall, Martin-Jezequel et al. supports our assumption of decoupled Si and N metabolism and is included in the manuscript as additional support.

However, we acknowledge that there is 1 study by Gilpin et al., 2004, discussing a coupling of N:Si. Hence, we added it in the discussion:

Change:

“Studies on the coupling of silicate limitation on C, N, and Chl show inconclusive patterns, including a complete decoupling (Claquin et al., 2002), a relation of N to Si (Gilpin et al., 2004) and reduction of photosynthesis (Werner, 1978; Gilpin et al., 2004) while no new chlorophyll is produced (Werner, 1978;

Gilpin et al., 2004).”…” Our cultivation study shows”…” ii) that coupling of Si:N:C:Chl is present. We do not expect a direct Si:N coupling, due to different controls of Si and N metabolism (Martin-Jézéquel et al., 2000.), but suggest indirect coupling via reduced photosynthesis.”

We agree that the 80% reduction should not be a fixed parameter, but tuneable. We adjusted the model accordingly. We also included the parameter to the sensitivity analysis and repeated the fitting routine.

Pg. 12, ln. 375: is a biofilm something one might expect in the natural system?; it doesn’t seem to be the sort of thing that would form in free water; also, it’s unclear from the methods whether there’s any agitation of the cultures to mimic ocean mixing

We would not expect biofilm formation in open oceans, but aggregation, which is commonly found in the end of spring blooms increasing the vertical export (e.g. Thornton, 2002). Both processes are similar in the way that algae aggregate via EPS facilitating a specific and active microbiome. We added a sentence about biofilm as proxy for marine snow in the discussion. Ocean mixing was mimicked by inverting all bottles 2-3 times a day (added to the methods).

Change in the discussion:

“While we would not expect biofilms in the open ocean, aggregation of algae cells, facilitated by EPS is common towards the end of spring blooms, increasing vertical export fluxes (e.g. Thornton, 2002). Chaetoceros socialis is in fact a colony forming diatom building EPS-rich aggregates in nature (Booth et al., 2002).”

Change in the methods:

“The cultures were incubated at 4°C and 100 µE m-2 s-1 continuous light and mixed 2-3 times a day to keep the algae and bacteria in suspension.”

Pg. 13, ln. 392: the value of the f-ratio has been questioned as the wider role of nitrifying bacteria has been recognised; perhaps rephrase talking instead about the balancing roles of export and remineralisation?

We replaced the term f-ratio by “regenerated production” and added that the higher regenerated production is due to increased remineralization compared to export.

Pg. 13, ln. 406: consider: Kamatani, A., Dissolution rates of silica from diatoms decomposing at various temperatures, Mar. Biol., 68, 91– 96, 1982

We included the reference

Pg. 14, ln. 426: model availability?; might be good to include the code too - it’s simple Enough

The R code is now available at github under https://github.com/tvonnahm/Dynamic-Algae-Bacteria-model.

Pg. 21, Figure 1: presumably the gap between (NOx + NH4) in the two experiments is due to N getting stuck in (dead) organic matter?; bar PON / POC, was anything about this recorded in the experiments?

We did not differentiate between life and dead organic matter, but assume mostly life organic matter until the stationary phase where biofilm formation played a role indicating EPS production, which can contribute to the measured PON and POC.

We agree that NH4 adsorption to organic matter (EPS) can play an important role and is likely one of the main explanations for the poor (lower) model fit of ammonium to the measured data. In addition, NH4 may be immobilized in micro-pockets of the biofilm unavailable for algae uptake.

However, we attribute the gap of DIN between the experiments mainly to a) increased NH4 regeneration in BACT+, with some ammonium likely immobilized in the biofilm (= higher NH4 concentrations), and b) preferred NH4 uptake over NO3 and NO3 uptake inhibition by NH4 leading to higher NO3 concentrations in the BACT+ treatment due to slower uptake. The PON/POC ratios change due to carbon overconsumption (Schartau et al. 2007), which is most relevant under N limitation, whule Si limitation has a more direct effect on photosynthesis (Lippemeier et al., 1999, Thangaraj et al., 2019; Liu et al., 2020). All these 3 dynamics are part of the extended model taking bacterial processes and NH4-NO3 interactions into account.

Pg. 21, Figure 1: the span of PO4 at day 14 (5-55) seems implausible given its narrow span at day 11 (30-35); especially as it narrows again at day 15 (5-18)

We argue that the large range is plausible since it is i) based on 1 data point, which may be an outlier and ii) it corresponds with high variation in bacteria abundances, which are ultimately responsible for the high PO4 value presumably originating from remineralization. Especially towards the end of the experiment it is not implausible that the different bottles behave differently.

Change in figure legend:

"c) PO42- with a potential outlier at day 14 leading to a negative peak"

Pg. 22, Figure 2: not so axenic, eh?; is this contamination in the axenic incubations from repeatedly opening the vessels?

As mentioned in the results, the bacteria growing towards the end are still in so low abundances compared to the bacteria enriched experiment, that it is effectively axenic. Obtaining and especially maintaining axenic diatom cultures is challenging and does typically not last very long. Since we used independent bottles during the experiment, contaminations during the course of the experiment are not possible (bottles were not opened before the sampling day). However, antibiotic treatments attack mostly active bacteria cells susceptible to the antiobiotics, while endospores and antibiotic resistant bacteria can survive. We believe that the bacteria starting to grow at day 14 originate form endospores activated by the high concentrations of DOM excreted by the stressed algae.

Change in the methods:

"We ensured sterile conditions during the experiment by keeping the cultivation bottles closed until sampling. However, endospores may survive the antibiotic treatment in low numbers and start growing especially towards the end of the experiment."

Pg. 23, Figure 3: so as well as having less NOx and NH4, the axenic experiments have less PON; where is the N going?

We suggest that the N is contributing to a higher DON pool (not measured) in the axenic experiments, which is not shown in Figure 1 and 3. The DON could be remineralized in the experiments with bacteria yielding higher NOx, NH4+ and PON.

We hope that our schematic representation of the model added to the methods helps to clarify it (See Fig. 1 above).

Pg. 24, Figure 4: it's idle curiosity, but what happens if you extend your model runs past the time point that the laboratory cultures ran?; the model should permit this
we added the output of a prolongated model run in the supplement (See above).

Pg. 24, Figure 4: the inflections on some of the model plots here look rather artificial; can you explain why there are such sharp transitions around the 8-day mark?
The sharp transition is due to the threshhold based formulation of reduced photosynthesis and inhibited Chl synthesis under Si limitation. At day 8, the silicate limitation threshold is reached and Photosynthesis is reduced and Chl synthesis inhibited. As describe dabove, threshold based modelling approaches are not uncommon.

Pg. 24, Figure 4: the spikes in chlorophyll in the cultures seem difficult to believe; do you think they are perhaps artifacts / measurement error?
Yes, as mentioned above these spikes represent a single data point that can be measurement artifacts and added this information to the figure legend.

Pg. 25, Figure 5: given that the key is the same in all of the plots, it would be better to not use it in plots where it interferes with the data (e.g. 5c)
We now only put the legend in plot a and mention that the legend is valid for all subplots.

Pg. 25, Figure 5: why are the fits without the excretion term all flat?; that's not what I'd expect at all; actually, I now realise that you're using two sets of dotted lines on this plot; one for the model output, one for the limiting concentration of the nutrients; this should be changed as it's a very confusing presentational choice

We changed the style of the lines.The fit without excretion is not the flat line, but the dotted line close to the +excr model. The –excr model simply modelled the excretion fraction of the +excr model into the maintenance respiration term (general loss without being available for remineralization). Since our system was highly affected by ambient DOM (likely terrestrial), the difference is little, showing that the regenerated production in our experiment is mostly caused by terrestrial DOM regeneration rather than freshly produced DOM regeneration.

Pg. 26, Table 1: the text reads as if these crosses denote both (a) remineralisation, and (b) variable stoichiometry?; that seems a lot for one cross to bear!; however, in the table, it looks like you separate out the stoichiometry - I think this sentence needs rewording

Pg. 26, Table 1: as a stylistic aside, a cross is not necessarily the best way to denote that a model includes something; conventionally, ticks are used, with ticks and crosses meaning opposite things

Pg. 26, Table 1: where other models are presented, these are often older versions of these models; might it be better to report their current versions?

We clarified the table caption and used ticks and crosses instead.
While the full ecosystem scale models may have more recent versions with updated formulations, we give the original reference to the biogeochemical compartment of the ecosystem scale models, which are still quite old. We will however, added references to the most recent full-scale models used in addition to the reference only describing the algae growth formulations. We added following references to more recent ecosystem scale model formulations:

BFM model: Smith, K. M., Kern, S., Hamlington, P. E., Zavatarelli, M., Pinardi, N., Klee, E. F., and Niemeyer, K. E.: BFM17 v1. 0: Reduced-Order Biogeochemical Flux Model for Upper Ocean Biophysical Simulations, *Geoscientific Model Development Discussions*, 1-35, 2020.

ReCom-2 model: Schourup-Kristensen, V., Wekerle, C., Wolf-Gladrow, D., and Völker, C.: Arctic Ocean biogeochemistry in the high resolution FESOM 1.4-REcoM2 model, Progress in Oceanography, 168, 65-81, doi:10.1016/j.pocean.2018.09.006, 2018.

MEDUSA model: Henson, S. A., Cole, H. S., Hopkins, J., Martin, A. P., and Yool, A.: Detection of climate change-driven trends in phytoplankton phenology, *Global Change Biology*, *24*(1), e101-e111, 2018.

NEMURO model: Anju, M., Sreeush, M. G., Valsala, V., Smitha, B. R., Hamza, F., Bharathi, G., and Naidu, C. V.: Understanding the Role of Nutrient Limitation on Plankton Biomass Over Arabian Sea Via 1-D Coupled Biogeochemical Model and Bio-Argo Observations, *Journal of Geophysical Research: Oceans*, *125*(6), 2020.

SINMOD model: Alver, M. O., Broch, O. J., Melle, W., Bagøien, E., and Slagstad, D.: Validation of an Eulerian population model for the marine copepod Calanus finmarchicus in the Norwegian Sea, *Journal of Marine Systems*, *160*, 81-93, 2016.

NPZD model: Gruber, N., Frenzel, H., Doney, S. C., Marchesiello, P., McWilliams, J. C., Moisan, J. R., Oram, J. J., Plattner, G., and Stolzenbach, K. D.: Eddy-resolving simulation of plankton ecosystem dynamics in the California Current System, Deep Sea Research Part I: Oceanographic Research Papers, 53(9), 1483-1516, 2006.

And we added and discussed following culture-scale model suggested by reviewer 4:

Flynn, K. J., Skibinski, D. O., and Lindemann, C.: Effects of growth rate, cell size, motion, and elemental stoichiometry on nutrient transport kinetics, PLoS computational biology, 14(4), 2018.

Pg. 27, Table A1: this is confusing; why not have separate columns for G98 and the extended model?; also, this table implies that some properties are not in the model, but you seem to have equations for them; meanwhile, there are other properties, e.g. dSi, for which no equation is presented

We had only a G98 column because all state variables are part of the extended model (The EXT model is the G98 model with added variables). We added a column for EXT besides G98 with ticks for every state variable for clarification. We also mentioned the equation for each state variable in table A6/A7 and added the missing equations.

Pg. 27, Table A1: you appear to be using underscores rather than minus signs in units at the base of this table

We changed it.

Pg. 30, Table A4: what do all of the columns mean here?; some explanation would be Useful

We shortened the table slightly and explained all columns in detail in the corrected version.

Change:

Table A4. Output of the sensitivity analysis (senFun of the FME package in R) with the value for each parameter and different sensitivity indices obtained after quantifying the effects of small perturbations of the parameters.on the output variables (POC, PON, Chl, DIN). The L1 and L2 norms are normalized sensitivity indices defined as $L1 = \sum \frac{|S_{i,j}|}{n}$ and $L2 = \sqrt{\frac{S_{i,j}^2}{n}}$ with $S_{i,j}$ being the the sensitivity of parameter i for model output j.

par	value	L1	L2	Mean	Min	Max
G98						
ζ	1.00	0.10	0.19	-0.02	-0.15	0.98
R^C	0.07	0.04	0.05	-0.03	-0.08	0.14

Pg. 31, Table A5: please choose a table size that doesn't line-break your units
We adjusted the table size

Pg. 32, Table A6: maybe pull the ODEs together in one place then follow-up with the separate terms afterwards?; it's a little difficult to parse the equations otherwise
we changed the order accordingly.

Pg. 33, Equation 1: if there's a conditionality on a single term in an equation (as here) better to have a single ODE and put the conditionality inside this term (i.e. it's this value if X, zero if Y); this is easier to follow and makes it much easier to see where the important parts of the model's behaviour lie; duplicating the equations for the sake of a single term in them does not make things clear
We changed the equations accordingly.

Pg. 33, Equation 1: you should note somewhere that organic C is removed from an unmodelled reservoir of DIC; unmodelled because it's always in excess of the ecosystem model's requirements
We added the information to the schematic figure in the methods and mentioned it next to the equation.

Pg. 35, Equation 15: the presentation of equations 14 and 15 around the 14e3 divisor is different; this is an unnecessary confounding factor that makes the equations less readable
We changed the form of eq 14 to the same format as in eq 15.

Pg. 35, Equation 16: why is this a hard-wired number (0.2) and not a parameter?; even if it's not something you change in your study (which seems a little strange given what you do change), having this as a clearly parameter rather than an undescribed constant is important
We changed this parameter to a tuneable parameter and included it into the sensitivity analyses and parameter fitting exercise.

Pg. 36, Figure B1: I don't understand what this plot is showing; please explain what it means for a line to deviate from zero here; also, why is sensitivity time-variable in any case?; and why is it not monotonically variable in time?; I also note that it looks like DIN is super-sensitive compared to the other properties - is that a correct interpretation of this plot?
The sensitivity analyses in the FME package tests the sensitivity of the model output (here DIN, POC, Chl, DIN) with changing parameter values within the predefined constraints. The plot shows the deviation from the model output towards the measured data over time. We realized that this figure is too complex while adding little information to the manuscript and removed it.

Pg. 37, Figure B2: a full explanation for what this plot is showing is critical; it is very difficult to understand what's being shown; also does the frequent occurrence of "NA" imply that some parameters should be excluded from this analysis?

The plot shows pairwise comparisons of parameter sensitivity/ sensitivity functions. On the upper right the pairwise data are shown for each tuneable parameter with the boundaries/constraints given in table A3. The sensitivity is given for POC (blue), PON (red) and Chl (green). The correlation coefficients are given in the lower left corner. NAs indicate no correlation because of low sensitivity. We realized that this figure is too complex while adding little information to the manuscript and replaced it with following table.

Change:

Table A8. Output of the collinearity or parameter identifiability analysis using the collin function of the FME R package (Soetaert et al., 2010b). A subset of any combinations of two parameter with a collinearity above 20, indicating non-identifiable parameter combinations is given (Brun et al., 2001).

ζ	R^C	θ^N_{max}	Q_{min}	Q_{max}	α^{Chl}	I	n	K_{no3}	P^C_{ref}	collinearity
1	0	1	0	0	0	0	0	0	0	31
1	0	0	0	1	0	0	0	0	0	59
1	0	0	0	0	1	0	0	0	0	42
1	0	0	0	0	0	1	0	0	0	42
1	0	0	0	0	0	0	1	0	0	74
0	1	0	0	0	0	0	0	0	1	22
0	0	1	0	1	0	0	0	0	0	32
0	0	1	0	0	1	0	0	0	0	26

Pg. 38, Figure B3: the key seems to omit reference to the bacterial model

We added the information in the legend.

Pg. 38, Figure B3: the failure of the model to capture the observed behaviour of the PON seems quite significant, but is not well-described in the text; it is also noticeably different from that of POC, which suggests interesting POM dynamics in the model that I would not expect; do the authors know what is going on here?

After G98 carbon is continuously fixed, even under nitrogen limitation (Carbon overconsumption, Schartau et al., 2007), while nitrogen is slowly used up for maintenance (maintenance respiration term), leading to a decoupling of POC and PON. The main reason for the failure of the G98 model is the neglection of bacterial DIN regeneration. Thus, the PON dynamics are quite well modelled for the BACT- experiment, while the BACT+ experiment shows severe limitations. In fact, this is one of the main arguments showing the need to include bacterial regeneration. The model may be tuned to an artificially better fit the BACT+ treatment by increased DIN uptake efficiencies, but this would lead to a substantially

poorer fit to the BACT- experiment. As discussed on p2 l.63 this fitting of the G98 model without a bacterial regeneration component on non-axenic culture experiment can lead to misleading interpretations and kinetic parameters (e.g. half saturation constants). We added this information to the discussion.

Schartau, M., Engel, A., Schröter, J., Thoms, S., Völker, C., & Wolf-Gladrow, D.: Modelling carbon overconsumption and the formation of extracellular particulate organic carbon, 2007.

Pg. 38, Figure B3: would quartile or decile range be better here?; this may make your experiments look more messy than they actually are (i.e. it looks like you may have an outlier experiment); this may not be possible given the number of replicates

With three measured values per day and treatment, we prefer to show all values separately instead of artificially calculating error estimates (e.g. quartiles, deciles, standard deviations).

Response to reviewer 3

Anonymous Referee #3
Received and published: 11 November 2020

In the manuscript, "Modelling Silicate – Nitrate - Ammonium co-limitation of algal growth and the importance of bacterial remineralisation based on an experimental Arctic coastal spring bloom culture study" by Vonnahme et al. the authors present a new model development for diatom co-limitation of nutrients. Based on experimental data they expand the classical model by Geider et al. (1998), which remains its feasibility for larger (ecosystem) models, while improving the representation of algae growth. Improving biological parameterizations in ecosystem models is important and contributes to improving their predicative capability. However, the authors should address some points listed below.

We want to thank the reviewer for the positive and very helpful review and addressed the points raised as described below.

The authors report that "With the onset of the stationary phase in the bacteria-enriched cultures about 30% of the cells formed biofilms on the walls of the cultivation bottles (estimated after sonication treatment)." (line 230). The formation of such biofilms has occurred in other experiments before and cannot always be avoided. However, it does potentially have a huge impact of microbial dynamics and interactions. Therefore only reporting (and discussing) it is insufficient, if one is to compare experimental results with a new modelling approach. I would suggest to run a model sensitivity analysis specifically targeting this.

We agree that the biofilm formation can have a substantial impact on some microbial dynamics and the identity of different carbon pools. Since, the biofilm only contributed to 30% of the cell counts we are still confident that the model is well suited to represent the dynamics of the experiment and coastal Arctic spring blooms. The current model has various dynamics changing after silicate limitation, especially in the presence of bacteria and NH4 regeneration. As outlined in the response to reviewer 2, we changed the reduction of photosynthesis after Si limitation from a hard-wired number (80%) to a tuneable parameter. Since Si limitation corresponds with the timing of biofilm formation, we assumed that the silicate limitation parameters (in particular Si_{PS} – reduction of photosynthesis after Si limitation) could describe the changed dynamics.

To test if this assumption holds true and to deepen the discussion of biofilm related dynamics, we suggest 3 potential dynamics which are likely different in the biofilm. Since the biofilm formation corresponds with Si limitation, we modelled changed dynamics after Si limitation to represent specific changes known to be different in biofilms.

1) DOC coagulation to EPS as part of the POC pool
   a. $dPOC_{EPS}/dt = x_f * x_{eps} * C$
   b. (x_{eps} – fraction of coagulation excreted DOC)
2) Increased DOM excretion in the stationary phase

a. IF (Si < lim){ $x_f = x_{f2}$ } else { $x_f = x_{f1}$}
b. (x_{f1}- excretion before Si limitation, x_{f2} –excretion after Si limitation)

3) Increased remineralization of excreted DON in the stationary phase
   a. IF (Si < lim){ rem= rem_2 } else { rem = rem_1}
   b. (rem_1- remineralization rate before limitation, rem_2 –remineralization rate after Si limitation)

We extended the model as described above and compared the new model output with the original fit. We changed tuned the new parameter manually until the model output showed substantial differences (approx. > 10% in POC, PON, Chl, or DIN). Eventually, we tested the effects of 100% DOM coagulation (x_{eps} = 1), 1000x higher remineralization rate after Si limitation (rem2 = 10000, rem1=10), 2x higher DOM excretion after Si limitation (xf1 = 0.06, xf2 = 0.12). The order of magnitude of perturbations needed to get changes of 10% of at least 1 output variable gives an indication of the parameters sensitivity. We then tried to tune the new model again to the initial fit by changing the Si_{PS} parameter. For each case, we were overall able to return to the original fit with less (1 & 2) or equal (3) perturbations of the SiPS parameter than was perturbed in the added parameters. This shows that SiPS is more sensitive and collinear (unidentifiable) with the added parameters, which shows clearly that the 3 suggested model extensions would not improve the model without additional data (e.g. EPS measurements). We added this discussion in a shortened way to the manuscript. More details are given below:

1) The POC can include not only algae biomass, but also EPS that holds the biofilms together. For estimating the potential importance of this POC pool, we added a model run, where all the excreted carbon (given by x_f * POC) is coagulating to EPS and thereby contributing to the POC pool. As shown in Fig 1 the outcome are ca 30% increased POC values in the stationary phase, which is in accordance with our estimate from the sonication treatment. However, it is unlikely that all excreted DOM aggregates to EPS (Schartau et al., 2007) and earlier studies describe much lower proportions of EPS being part of the EPS pool (up to 7% of an biofouling diatom biofilm, Khandeparker and Bhosle, 2009), with the highest fraction after nitrogen and silicate limitation in the stationary phase. A potential model extension to account for EPS aggregation that contributes to the EPS pool would be the approach, described by Schartau et al. (2007) who model carbon excretion (3 different DOC pools) and aggregation to TEP (transparent exopolymeric substances). However, since we did not measure cellular C and extracellular EPS separately, we argue that the extension requiring 11 additional parameters and 3 additional state variables of TEP (EPS) and 3 different DOC pools would i) not be in line with our goal to develop a simple model, and ii) would not be justified by the measured data, making the tuning process rather speculative (overfitting issue). Nevertheless, we acknowledge that this process needs to be discussed and we add the figure below to the supplement to show the maximum potential importance of EPS aggregation (assuming immediate aggregation of excreted DOC).

For estimating the importance of considering EPS aggregation, we also tested an extended model where a fraction of the excreted carbon is coagulated to EPS (xeps), contributing to the POC pool. We added following equation:

$$POC_{EPS} = x_{EPS}\, x_f\, POC$$

with a start value of 50% aggregation and no constraints (values between 0 % and 100 %). The difference between the extreme values of 0 % and 100 % are shown in Fig. 1 below and lead to 30 % difference in maximum POC. This makes the parameter quite insensitive. In fact the SensFUN of the FME package defines the sensitivity of the added xeps parameter close to 0. We also suggest, that the effect of xeps could be compensated by the SIPS term of the EXT model (% reduction in photosynthesis after Si reduction), leading to a very similar fit, indicating collinearity. This is mainly the cause since, EPS aggregation only has a major role with a linear response in the stationary phase when also Si is limiting. Thus, an additional xeps term would be unidentifiable with the current set of measured data.

We will add a more thorough discussion of the approach by Schartau et al. (2007) and the suggestion for a more simplified model extension described above (adding the xeps parameter), for experiments were EPS data are available, but with its limitations for the current model due to collinearity/unidentifiability issues.

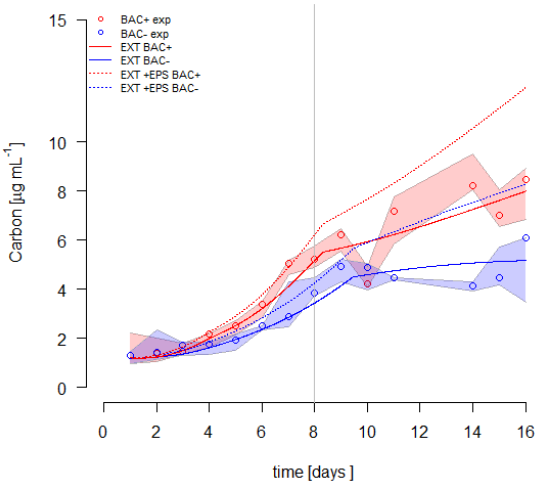

Figure 1. POC concentrations of the measured data and model, including a model run showing POC as originally modelled POC + excreted DOC assumed to aggregate immediately and completely to EPS (dashed line).

2) Another finding by Khandeparker and Bhosle (2009) is an increased DOM excretion after Si and N limitation, which is not yet part of our model. Hence, we added a second excretion term xf2 after silicate limitation. A doubling of the excretion rate after silicate limitation leads to slightly reduced POC and PON values (Fi. 2), but no changes in Chl and only small extra NH4, due to the higher importance of the ambient DON for NH4 regeneration. The lower POC values can be completely compensated by doubling the the Photosynthesis after Si limitation (SiPS 0.2 -> 0.4) parameter. The lower PON value can almost be compensated with the same parameter.
This little difference indicate, that a modelling approach with changing xf rates after Si or N limitation is not necessary, at least not in our model system with high refractory DOM concentrations. It may however, become important in open-ocean systems with less terrestrially derived DOM.

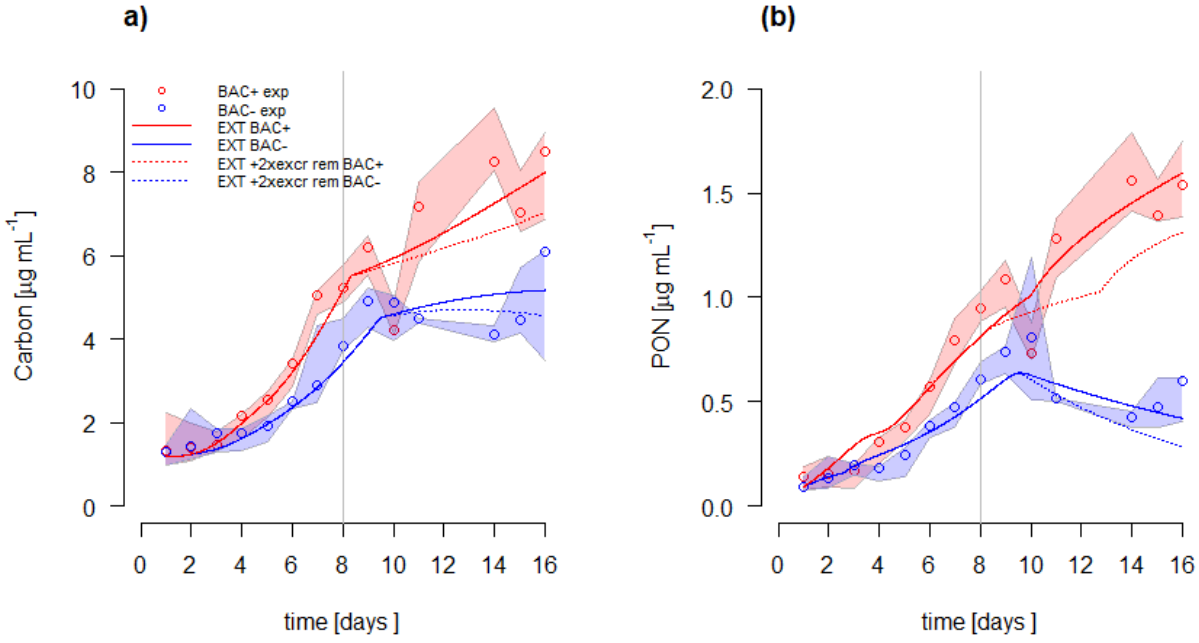

Figure 2. Impacts of a 2 times increased DOM excretion after Si limitation (dashed line).

3) The biofilm can also facilitate interactions between bacteria and algae due to the closer proximity. This increased interactions could be represented in an increased remineralization rate of excreted organic matter (rem) after N or/and Si limitation. A potential model extension accounting for it would include a second higher remineralization rate after Si or N limitation. However, the difference between the EXT model where C is excreted as DOC (xf), or simply lost for maintenance respiration (RC) is minor (Fig. 3).
After adding a second remineralization rate of labile DOM (rem2) an increase of 3 orders of magnitude is needed (rem2 = 10e03 rem) to show any visible effect on N assimilation and NH4

regeneration (Fig. 3), showing that this is a highly insensitive parameter. However, about 4 order of magnitude higher rates appear to bring the modelled NH4 concentrations closer to the measured data (while fits to POC, PON, and Chl become very different form the measured data), hinting that the poor model fit to NH4 may not only be related to immobilized NH4 in the measured data (e.g. NH4 adsorbed or trapped in EPS), but may also be related underestimated DON excretion or remineralization in the model.

However, more than 10% DOM excretion and a DON remineralisation rate 3-4 order of magnitude higher than remineralization of the ambient, likely terrestrially derived DOM is rather unlikely. A likely explanation of the low impact of increased remineralization in the biofilm of our experiment is the high ambient DOM concentrations, which are the main DON source for NH4 remineralization (See difference of the extended model with and without excretion in the manuscript). Since our model is supposed to represent coastal systems, we thus argue that only 2 different remineralization rates related to refractory and labile OM is sufficient. In more open ocean setting with less allochthonous DOM input, increased remineralization rate of algae EPS in the stationary phase, may be a useful addition.

We suggest therefore, that a higher remineralization rate is likely, but that a large part of the remineralized NH4 is not available for algae growth due to the biofilm. Thus, the modelled NH4 values represent the available NH4 for algae, which representation is the aim of this study.

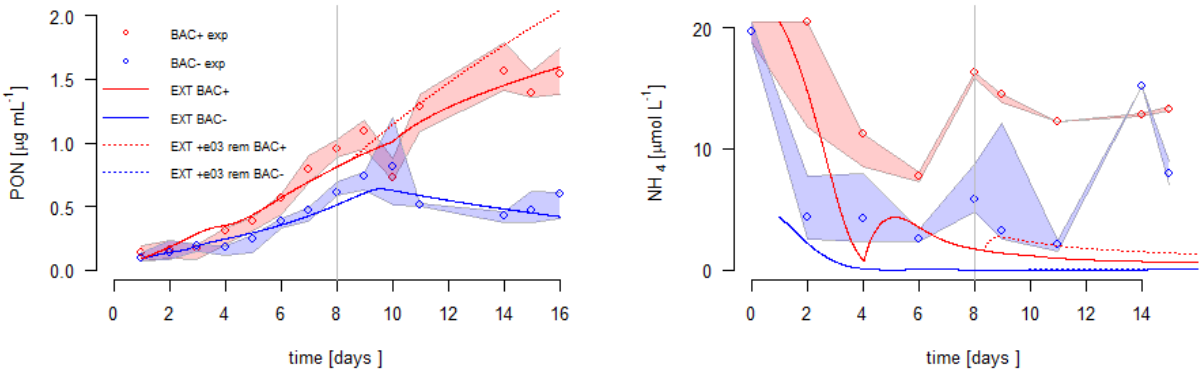

Figure 3. Comparison of the original model and a 3 orders of magnitude increased remineralization rate of excreted DON after Si limitation (dashed line).

We add Fig. 3 to the supplement and discuss the potential of increased bacterial remineralization in biofilms and why this is not quantitatively important in our experiment and model.

4) Another effect of the biofilm may be adsorption of ammonium to the EPS or concentration in pockets, not available for algae growth. In fact, this could be one of the explanation for the consistently high NH4 values in the stationary phase, which are poorly represented in the model (See response to Referee #2).

Response to Referee #2

"Ammonium is most likely immobilized in the biofilm via adsorption to the EPS and accumulation in pockets unavailable to diatoms. These immobile NH4 pools are still part of the measured data. With the model assuming all NH4 being available for algae growth, this is a problem."

This could, in particular, explain the high values of measured NH4 compared to the model results as shown in Figure 5c. In addition, the higher values of measured NH4 could be explained in terms of a potential pH dependence of NH4+ adsorption to the EPS in terms of the pKa values of NH4+ and carboxylic groups, which belongs to the acidic polysaccharides as a fraction of EPS:

- Carboxylic groups have a pKa < 5, i.e. far away from seawater pH ~ 8, which means that they are always in the deprotonized negatively charged form R-COO- in seawater.
- NH4+ has a pKa ~9 closer to seawater pH.
- Thus, the NH4+/NH3 ratio will be higher in more acidic microenvironments (pH ~7.5-8).
- Thus, a lower pH due to bacterial respiration would increase the concentration of NH4+ in comparison to the bulk medium, which results in a higher immobile NH4 pool due to adsorption to the EPS.
- This could explain the higher discrepancy between modelled and measured NH4+ values in the experiments with bacteria (as seen in Figure 5c).

Since the biofilm formation corresponds with silicate limitation, the reduced photosynthesis might of course be related to either the biofilm or the silicate limitation. But for untangling the effects of biofilm formation and silicate limitation, more experiments or data would be needed. However, only 30% of the culture was part of the biofilm and the best fit of an 80% reduction corresponds very well with an earlier study by Werner (1978), who did not have the issue of biofilm formation. Hence, we suggest that the main cause for the reduction of photosynthesis is related to Si limitation and not the biofilm.

We add this argumentation together with collinearity issues of SiPS with potential model extensions taking the biofilm into account to the discussion. We will also add the figures above showing the impact/sensitivity of potential model extensions to account for the changed dynamics in a biofilm to the supplement.

In the manuscript we add a discussion about the results explaining: i) the potential changes in a biofilm (increased DOM excretion, increased remineralization, trapped NH4), ii) the importance of the biofilm for our model run (POC as EPS instead of algae biomass, differences in fitting and sensitivity for the stationary or exponential phase, considerations of the biofilm being only 30%), iii) we also added a discussion of biofilms or aggregates/marine snow in the environment, which our study aims to represent.

Rakhee DS Khandeparker & Narayan B Bhosle (2001) Extracellular polymeric substances of the marine fouling diatom amphora rostrata Wm.Sm, Biofouling, 17:2, 117-127, DOI:10.1080/08927010109378471

The authors appropriately discuss quota models and their use. A different approach to model celluar nutrient kinetics, that has been argued to be more mechanistic, considers uptake sites for nutrients (Aksnes & Egge, 1991, Mar Ecol Prog Ser. 70:65-72). A good, though slightly technical, paper applying this approach and combining it with variable cellular stoichiometry is Flynn et al., 2018, PLoS Comput Biol 14(4): e1006118. Setting up a model like this for your data could be highly interesting, but beyond the scope of this study. However discussion the approach would provide a very useful context.

We want to thank the reviewer for the interesting suggestion and reference. We added the model by Flynn et al. to Table 1. We also included a discussion of the approach. We argue overall that the model is too complex for the aim of our study, which tries to keep the number of parameters as low as possible allowing scalability (similar to Flynn, 1997; Flynn, 2001), but acknowledge the important role of considering transporter densities, cell size, and mobility. Especially the importance of mobility is an interesting aspect, that we now discussed in the context of diatom sedimentation.

Change:

"The next step to quota based-models is the consideration of more detailed cell based characteristics, such as transporter density, cell size, and mobility, including sedimentation (Aksnes and Egge, 1991). Flynn et al. (2018) discuss a model with detailed uptake kinetics showing that large cells are overall in disfavored over small cells due to higher half saturation constant, but that they may still have competitive advantages due to lower investment in transporter production, and increased sedimentation, increasing the mobility that may offset the disadvantage of a larger size. While this extension is too complex for our aim of a simple model, the dynamics may become important when modelling different algae taxa."

In the introduction (line 46) and in the discussion the authors mention the role of the impact of climate change on coastal phytoplankton succession, including projected increased DOM inputs via river run off. Several studies have found and/or suggested a delayed bloom due to increase turbidity (e.g. Opdal et al. 2019, Glob Change Biol. 2019;00:1–8), which should be mentioned here.

We thank the reviewer for the suggestion, which is an important clarification to the introduction and in particular for the discussion.

Change in Introduction:

"… stratification in coastal Arctic systems is expected to increase… earlier stratified surface layer in spring, which may lead to an earlier spring bloom (Tremblay and Gagnon, 2009).""However, at the same time, brownification and increased sediment resuspension is already leading to light inhibition in spring, which may lead to a delayed spring bloom (Opdal et al., 2019)."

Change in Discussion:

"An earlier temperature driven water column stratification may also lead to an earlier bloom. However, due to increasing river and lake brownification and sediment resuspension, the spring bloom may be delayed (Opdal et al., 2019)." "With decreased light, carbon overconsumption as described by Schartau et al. (2007) may become less important due to decreased photosynthesis. An earlier, or later phytoplankton bloom can lead to a mismatch with zooplankton grazers (Durant et al., 2007;Sommer et al., 2007), which could decrease the fecal pellet driven vertical export and thereby increase the residence time of POM in the euphotic zone and the potential for ammonium regeneration, making the incorporation of bacterial recycling into ecosystem models even more imporatant as also evident from our experimental data and model output."

The authors mention both nitrate and ammonium as nitrogen sources. Additionally, urea can be a relevant nitrogen source in some systems. I am not sure how much of a role this plays in arctic ecosystems, but it should either be discussed or mentioned why it does not play a significant role.

We agree with the reviewer that urea may be another important nitrogen source, especially under nitrate limitation. In some Arctic systems it may reach concentrations of 2 uM. While bacteria may produce urea by ON degradation, the main source of urea is attributed to zooplankton excretion (Conover and Gustavson, 1999). Hence, it does not play a role in our experiment, but may play a role in nature. We added a discussion of urea as potential nitrogen source to the discussion of zooplankton NH4 excretion.

Change in introduction:

"Zooplankton may also release some ammonium and urea after feeding on phytoplankton, but we suggest that this process is likely far less important than bacterial regeneration (e.g. Saiz et al., 2013). Previously measured ammonium excretion of Arctic mesozooplankton is typically low compared to bacterial remineralization (Conover and Gustavson, 1999), with the exception for one study in summer in a more open ocean setting (Alcaraz et al., 2010). In some Arctic systems urea, excreted by zooplankton may be an important N source for regenerated algae production (Conover and Gustavson, 1999)."

Change in discussion:

"Another potentially important N source for regenerated production may be urea (Harrison et al., 1985), which would lead to an even higher importance of regenerated production as suggested by our study."

Conover, R. J., and Gustavson, K. R.: Sources of urea in arctic seas: zooplankton metabolism, Marine Ecology Progress Series, 179, 41-54, 1999.

Harrison, W. G., Head, E. J. H., Conover, R. J., Longhurst, A. R., and Sameoto, D. D.: The distribution and metabolism of urea in the eastern Canadian Arctic, Deep Sea Research Part A, Oceanographic Research Papers, 32(1), 23-42 1985.

Line 168: ": : :but the growth rate can be reduced (Hildebrand, 2002; Gilpin, 2004)". How can the growth rate be reduced? What can lead to this reduction?

We realized that growth rate is not the best term here and changed the sentence as follows:

Change:

"N and Si metabolism have different controls and intracellular dynamics, with N uptake fueled by photosynthesis (as PCref in G98) and Si mainly fueled by heterotrophic respiration (Martin-Jezequel et al., 2000). In general, we assume that nitrogen metabolism is not directly affected by silicate limitation (Hildebrand 2002, Claquin et al., 2002), but we expect cellular ratios to be affected by reduced photosynthesis and chlorophyll synthesis under Si limitation (Hildebrand, 2002; Gilpin, 2004)."

We also suggest to add more recent references on the effects of Si limitation on photosynthesis.

Gilpling (2004) only described the relationships of C,N,Chl production/assimilation under N and Si limition, but didn't give a physiological explanation.
Lippemeier et al. (1999) found a direct inhibition of the PSII reaction centre due to increased photochemical quenching, which is part of the explanation, but still rather descriptive. Our study confirmed lower efficiency of PSII (via Quantum yield measurements) after Si limitation, which is in accordance with Lippemeier et al. (1999) and supports our approach of reduced photosynthesis after Si limitation. Thus, we added a reference to this study in our discussion of the Quantum yield.
Another recent study by Liu et al. (2020) investigated gene expression patterns for C fixation related genes, and found reduced exression under Si limitation, but not under N or P limitation. The most detailed study has probably been done by Thangaraj et al. (2019), who used a metaproteomics approach and found not only downregulated photosynthetic proteins after silicate limitation, but also distracted protein production for mitochondria-chloroplast interactions, chlorophyll synthesis, and mechanisms compensating for disruption in electron transfer.

Lippemeier, S., Hartig, P., and Colijn, F.: Direct impact of silicate on the photosynthetic performance of the diatom Thalassiosira weissflogii assessed by on-and off-line PAM fluorescence measurements, Journal of Plankton Research, 21(2), 1999.

Liu, Q., Xing, Y., Li, Y., Wang, H., Mi, T., Zhen, Y., & Yu, Z. (2020). Carbon fixation gene expression in Skeletonema marinoi in nitrogen-, phosphate-, silicate-starvation, and low-temperature stress exposure. Journal of Phycology, 56(2), 310-323.

Thangaraj, S., Shang, X., Sun, J., and Liu, H.: Quantitative proteomic analysis reveals novel insights into intracellular silicate stress-responsive mechanisms in the diatom Skeletonema dohrnii, International Journal of Molecular Sciences, 20(10), 2540, 2019.

We added more details and references to the introduction, model description, and discussion.

Figure 6 and figure 7 do not exist.

We corrected the figure references.

Line 660: Table 1 is not the most up-to-date. Especially on the ecosystem model side it would be nice to see more recent developments reflected as well.

For the cultivation based models, we added the study by Flynn et al., 2018 as mentioned above. For the ecosystem scale models, we cited the original references of the algae growth and potential nutrient regeneration dynamics, which are often rather old, while the full-scale models are mostly updated in terms of physical formulations. We clarified this in the legend and added following more recent references to the ecosystem scale models:

BFM model:_ Smith, K. M., Kern, S., Hamlington, P. E., Zavatarelli, M., Pinardi, N., Klee, E. F., & Niemeyer, K. E. (2020). BFM17 v1. 0: Reduced-Order Biogeochemical Flux Model for Upper Ocean Biophysical Simulations. *Geoscientific Model Development Discussions*, 1-35.
ReCom-2 model: Schourup-Kristensen, V., Wekerle, C., Wolf-Gladrow, D., Völker, C. (2018): Arctic Ocean biogeochemistry in the high resolution FESOM 1.4-REcoM2 model, Progress in Oceanography, 168, 65-81,doi:10.1016/j.pocean.2018.09.006.

MEDUSA model: Henson, S. A., Cole, H. S., Hopkins, J., Martin, A. P., & Yool, A. (2018). Detection of climate change-driven trends in phytoplankton phenology. *Global Change Biology*, *24*(1), e101-e111.
NEMURO model: Anju, M., Sreeush, M. G., Valsala, V., Smitha, B. R., Hamza, F., Bharathi, G., & Naidu, C. V. (2020). Understanding the Role of Nutrient Limitation on Plankton Biomass Over Arabian Sea Via 1-D Coupled Biogeochemical Model and Bio-Argo Observations. *Journal of Geophysical Research: Oceans*, *125*(6), e2019JC015502.
SINMOD model: Alver, M. O., Broch, O. J., Melle, W., Bagøien, E., & Slagstad, D. (2016). Validation of an Eulerian population model for the marine copepod Calanus finmarchicus in the Norwegian Sea. *Journal of Marine Systems*, *160*, 81-93.

NPZD model: Gruber, N., Frenzel, H., Doney, S. C., Marchesiello, P., McWilliams, J. C., Moisan, J. R., Oram, J. J., Plattner, G., and Stolzenbach, K. D.: Eddy-resolving simulation of plankton ecosystem dynamics in the California Current System, Deep Sea Research Part I: Oceanographic Research Papers, 53(9), 1483-1516, 2006.

Especially in the abstract and the introduction there are several long (sometimes convoluted) sentences. To increase readability it would be could to rephrase these (Schachtelsaetze sind im Englischen nicht so hoch angesehen wie im Deutschen ;)).

We splitted the long sentences into shorter easier to read sentence in the revised version.

Revised Manuscript with tracked changes

Modelling Silicate – Nitrate - Ammonium co-limitation of algal growth and the importance of bacterial remineralisation based on an experimental Arctic coastal spring bloom culture study

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Abstract. Arctic coastal ecosystems are rapidly changing due to climate warming~~, which~~. This makes modelling their productivity crucially important to better understand future changes. System primary production in these systems is highest during the pronounced spring bloom, typically dominated by diatoms. Eventually the spring blooms terminate due to silicon or nitrogen limitation. Bacteria can play an important role for extending bloom duration and total CO_2 fixation through ammonium regeneration. Current ecosystem models often simplify the effects of nutrient co-limitations on algal physiology and cellular ratios and ~~neglect bacterial driven~~simplify nutrient regeneration. These simplifications~~,~~ may lead~~ing~~ to ~~an~~ underestimations of primary production. Detailed biochemistry- and cell-based models can represent these dynamics but are difficult to tune in the environment. We performed a cultivation experiment that showed typical spring bloom dynamics, such as extended algal growth via ~~bacteria~~bacterial ammonium remineralisation, ~~and~~ reduced algal growth and inhibited chlorophyll synthesis under silicate limitation, and gradually reduced nitrogen assimilation and chlorophyll synthesis under nitrogen limitation. We developed a simplified dynamic model to represent these processes. ~~The model also highlights the importance of organic matter excretion, and post bloom ammonium accumulation.~~ Overall, model complexity (number of parameters) is comparable to ~~other~~the phytoplankton growth and nutrient biogeochemistry formulations in common ecosystem models used in the Arctic while improving the representation of nutrient co-limitation related processes. Such model

enhancements that now incorporate increased nutrient inputs and higher mineralization rates in a warmer climate will improve future predictions in this vulnerable system.

1 Introduction

Marine phytoplankton ~~is~~are responsible for half of the CO_2 fixation on Earth (Field et al., 1998; Westberry et al., 2008). ~~Diatoms in~~In high latitude oceans, diatoms are an important group contributing 20-40% of the global CO_2 fixation (Nelson et al., 1995; Uitz et al., 2010). ~~-~~Marine primary production can be bottom-up limited by light and/or nutrients like nitrogen (N), phosphorous (P), silicon (Si), and iron (Fe). Their availability is affected by~~with~~ pronounced geographical and seasonal variations ~~in their availability~~ (Eilertsen et al., 1989; Loebl et al., 2009; Iversen and Seuthe, 2011; Moore et al., 2013). Arctic coasts are one of the fastest changing systems due to climate change. Thus,~~and~~ modelling their dynamics is difficult but crucial for predictions of primary production with climate change (e.g. Slagstad et al., 2015; Fritz et al., 2017; Lannuzel et al., 2020). In Arctic coastal ecosystems, primary production is typically highest in spring. ~~,~~In spring, previous ~~after~~ winter mixing supplied fresh nutrients~~, sea ice has melted, and combined with increasing temperatures, caused the formation of~~ and a stratified surface layer with sufficient light is facilitated by increasing temperatures and potentially sea ice melt (Sverdrup, 1953; Eilertsen et al., 1989; Eilertsen and Frantzen, 2007; Iversen and Seuthe, 2011). With increasing temperatures and runoff, stratification in coastal Arctic systems is expected to increase (Tremblay and Gagnon, 2009). This~~,~~ will lead~~leading~~ to decreased mixing and nutrient upwelling in autumn and winter and an earlier stratified surface layer in spring ~~(Tremblay and Gagnon, 2009)~~, which may lead to an earlier spring bloom (Tremblay and Gagnon, 2009). However, at the same time, brownification and increased sediment resuspension is already leading to light inhibition in spring, which may lead to a delayed spring bloom (Opdal et al., 2019). The spring bloom typically consists of chain-forming diatoms and is terminated by Si or N limitation (Eilertsen et al., 1989; Iversen and Seuthe, 2011). ~~Bacteria remineralisation~~Zooplankton grazing is typically of low importance for terminating blooms (e.g. Saiz et al., 2013), while inorganic nutrients are considered to drive bloom termination (Krause et al. 2019, Mills et al. 2018). Heterotrophic bacteria remineralisation of organic matter may supply additional N and Si (Legendre and Rassoulzadegan, 1995; Bidle and Azam, 1999; Johnson et al., 2007). ~~However,~~ N regeneration has been described as a mostly bacteria-related process (Legendre and Rassoulzadegan, 1995), while Si dissolution is mainly controlled by abiotic dissolution of silica (Bidle and Azam, 1999). Zooplankton may also release some ammonium and urea after feeding on phytoplankton, but we suggest that this process is likely far less important than bacterial regeneration (e.g. Saiz et al., 2013). Previously measured ammonium excretion of Arctic mesozooplankton is typically low compared to bacterial remineralization (Conover and Gustavson, 1999), with the exception for one study in summer in a more open ocean setting (Alcaraz et al., 2010). In some Arctic systems urea, excreted by zooplankton may be an important N source for regenerated algae production (Conover and Gustavson, 1999). A warmer climate will increase both

bacteria-related remineralisation rates (Legendre and Rassoulzadegan, 1995; Lannuzel et al., 2020) and abiotic silica dissolution (Bidle and Azam, 1999).~~,~~ However,~~But~~ the magnitude is not well understood.

Phytoplankton blooms may be dominated by a single or a few algal species, often with a similar physiology during certain phases of the bloom (e.g. Eilertsen et al., 1989; Degerlund and Eilertsen, 2010; Iversen and Seuthe, 2011). Chain-forming centric diatoms~~,~~ share~~ing~~ physiological needs and responses to nutrient limitations (e.g. Eilertsen et al., 1989; von Quillfeldt, 2005) and~~,~~ typically dominate these blooms. In some Arctic and sub-Arctic areas the Arctic phytoplankton species chosen for this model, *Chaetoceros socialis*, ~~is a~~can be dominant~~ species~~ during spring blooms (Rey and Skjoldal, 1987; Eilertsen et al., 1989; Booth et al., 2002; Ratkova and Wassmann, 2002; von Quillfeldt, 2005; Degerlund and Eilertsen, 2010). Such spring phytoplankton blooms are accompanied by heterotrophic bacterioplankton blooms also showing typical succession patterns and distinct re-occurring taxa that dominate the community (Teeling et al., 2012; Teeling et al., 2016). The importance of bacterial nutrient recycling for regenerated production has been recognized in several ecosystem models (e.g. van der Meersche et al., 2004; Vichi et al., 2007; Weitz et al., 2015) and algae bioreactor models focussing on nutrient conversions (e.g. Zambrano et al., 2016). However,~~,~~ these models are ~~but is~~ typically ~~neglected~~highly simplified or omitted in more sophisticated dynamic multi-nutrient, quota based models~~.~~ (e.g. Flynn and Fasham, 1997b.; Wassmann et al., 2006; Ross and Geider, 2009). These latter models have been often developed and tuned based on cultivation experiments in which microbial remineralization reactions were assumed to be absent (e.g. Geider et al., 1998; Flynn, 2001) despite the fact that most algae cultures, likely including Geider et al., (1998) and Flynn (2001) are not axenic~~ and~~. Parameters estimated by fitting axenic models ~~based~~ on ~~these experiments ignore bacterial contributions to nutrient recycling~~non-axenic experiments may be misleading, mostly by an inflated efficiency of DIN uptake. Additional positive effects of bacteria include vitamin synthesis (Amin et al., 2012), trace metal chelation (Amin et al., 2012), the scavenging of oxidative stressors (Hünken et al., 2008), and exchange of growth factors (Amin et al., 2015). ~~However, especially~~Especially in the stationary algal growth phase~~,~~ Christie-Oleza et al. (2017) found that marine phototrophic cyanobacteria cultures are dependent on heterotrophic bacteria contaminants mainly due to their importance in degrading potentially toxic DOM exudates and regenerating ammonium. The current study aimed to bridge the gap between detailed representations of algae physiology and the role of microbial activity in an accurate way while keeping model complexity low.

Most ecosystem models consider only a single limiting nutrient to control primary production after Liebig's Law of the minimum (Wassmann et al., 2006; Vichi et al., 2007). Yet we know that nutrient co-limitation is more complex.~~;~~ ~~i.e.~~For example, ammonium and glutamate can inhibit nitrate uptake (Morris, 1974; Dortch, 1990; Flynn et al., 1997), ~~iron has a strong control on silicate~~C and N uptake is reduced under Fe limitation, while Si uptake continues (Werner, 1977; ~~Hohn~~Firme et al., ~~2009~~2003), and the effects on photosynthesis differs ~~between~~for nitrogen and silicon limitations and for different algal groups (Werner, 1977; Flynn, 2003; Hohn et al., 2009). Complex interaction models considering intracellular biochemistry (NH_4-NO_3 co-limitation, Flynn et al., 1997), transporter densities and mobility (Flynn et al., 2018), and cell cycles (Si limitation, Flynn, 2001) can accurately describe these dynamics (Flynn, 2003), but are ultimately too ~~complex~~computationally expensive to be integrated and parameterized in large scale ecosystem models. Some models (Hohn et al., 2009, Le Quéré et al., 2016) implemented multinutrient (Hohn et al., 2009) and heterotrophic bacterial dynamics (Le Quéré et al., 2016) in Southern

Ocean ecosystem models, but have their limitations in representing bacterial remineralisation (Hohn et al., 2009), or ammonium and silicate co-limitations (Le Quéré et al., 2016). In contrast to Antarctica, DIN is the primary limiting nutrient for phytoplankton growth while iron ~~and phosphate are~~is not limiting in most Arctic ~~coastal~~ systems~~. Controlled~~ (Tremblay and Gagnon, 2009; Moore et al., 2013).

While simple lab experiments~~, representing the major~~ cannot represent all nutrient dynamics found in the environment ~~and predicted with climate change, are needed~~(e.g. N excretion by zooplankton), they can focus on the quantitatively most important dynamics, to facilitate the development of simple~~, but accurate~~ multinutrient models scalable to larger ecosystem models.

The current study investigated the relevance of silicate, ammonium - nitrate co-limitation, bacterial nutrient regeneration and changes in photosynthesis, nitrogen assimilation, and cellular quotas in response to the changing nutrient limitations based on data from a culture based Arctic spring bloom system. The culture consisted of an axenic isolate of *Chaetoceros socialis*, dominating a phytoplankton net haul of a Svalbard fjord. The culture was~~,~~ used experimentally either under axenic conditions or after inoculation with ~~its associated bacteria.~~bacteria cultures, isolated beforehand from the non-axenic culture. Parametrization and insights from these incubations were then used to develop and parameterize a simple Carbon quota based dynamic model (based on Geider et al., 1998), aiming to keep ~~complexity~~the number of parameters, and computational costs low to allow its use in larger ecosystem models.

The aims of the study was I) to study the bloom dynamics of simplified Arctic coastal pelagic system in a culture experiment consisting of one Arctic diatom species and ~~associated~~co-cultured bacteria, II) to develop a simple dynamic model representing the observed ~~cell~~ interactions, and III) to discuss the importance of more complex bloom dynamics and their importance for an accurate ecosystem model.

We hypothesize that: I) Bacterial regeneration ~~of ammonium will extend~~extends a phytoplankton growth period and gross carbon fixation; II) ~~Silicate or nitrogen limitations will have different physiological effects and physiological responses~~Diatoms continue photosynthesis under silicate limitation at a reduced rate if DIN is available; III) ~~A simple growth experiment and dynamic model with three nutrient pools and bacterial DON regeneration can adequately represent Arctic~~Cultivation experiments are powerful for understanding the major spring bloom dynamics.

2 Methods

2.1 Cultivation experiment

The most abundant phytoplankton species from a net haul (20µm mesh size) in April 2017 in van Mijenfjorden (Svalbard) *Chaetoceros socialis* was isolated via the dilution isolation method (Andersen et al., 2005) on F/2 medium (Guillard, 1975). Bacteria were isolated on LB-medium (evaluated by Bertani, 2004) Agar plates using the algae culture as inoculum and sequenced at GENEWIZ LLC using the Sanger method and standard 16S rRNA primers targeting the V1-V9 region (Forwards 5'-AGAGTTTGATCCTGGCTCAG -3', Reverse 5'- ACGGCTACCTTGTTACGACTT -3') provided by GENEWIZ LLC for identification via blastn (Altschul et al., 1990). Two strains of *Pseudoalteromonas elyakovii*, a taxon previously isolated from the Arctic (Khudary et al., 2008) and known to degrade algae polysaccharides (Ma et al., 2008) and to excrete polymeric substances (Kim et al., 2016), were

successfully isolated and used for the experiments. Before the start of the experiment, all bacteria in the algae culture were killed using a mixture of the antibiotics penicillin and streptomycin. The success was confirmed via incubation of the cultures on LB-Agar plates and bacterial counts after DAPI staining (Porter et al., 1980). The axenic cultures were diluted in fresh F/2 medium lacking nitrate addition (Guillard, -1975) using sterile filtered seawater of Tromsø sound (Norway) as basis. The algae cultures were transferred into 96 200ml sterile cultivation bottles with three replicates for each treatment. Half of the incubations were inoculated with bacteria cultures~~,~~ (BAC+), while the other half was kept axenic~~.~~ (BAC-). The cultures were incubated at 4°C and 100 µE m^{-2} s^{-1} continuous light~~.~~ and mixed 2-3 times a day to keep the algae and bacteria in suspension. We ensured sterile conditions during the experiment by keeping the cultivation bottles closed until sampling. However, endospores may survive the antibiotic treatment in low numbers and start growing especially towards the end of the experiment. Over 16 days three axenic and three ~~bacteria-enriched~~BAC+ bottles were sacrificed daily for measurements of chlorophyll a (Chl), particulate organic carbon (POC) and nitrogen (PON), ~~bacteria cell numbers, algae cell numbers~~bacterial and algal abundances, nutrients (nitrate, nitrite, ammonium, phosphate, silicate), dissolved organic carbon (DOC), and the maximum quantum yield (QY) of PSII (Fv/Fm) as a measure of healthy photosystems.

Chlorophyll a was extracted from a GF/F (50ml filtered at 200mbar) filter at 4C° for 12-24h in 98% methanol in the dark before measurement in a Turner Trilogy™ Fluorometer (evaluated by Jacobsen and Rai, 1990). POC and PON were measured after filtration onto precombusted (4h at 450°C) GF/F (Whatman) filters (50ml filtered at 200mbar), using a Flash 2000 elemental analyser (Thermo Fisher Scientific, Waltham, MA, USA) and Euro elemental analyser (Hekatech) following the protocol by Pella and Colombo (1973) after removing inorganic carbon by fuming with saturated HCl in a desiccator. Bacteria were counted after fixation of a water sample for 3-4h with 2% Formaldehyde (final concentration), filtration of 25ml on 0.2µm pore size Polycarbonate filter, washing with filtered Seawater and Ethanol, DAPI staining for 7 minutes after Porter et al. (1980), and embedding in Citifluor-Vectashield (3:1). Bacteria were counted in at least 20 grids under an epifluorescence microscope (Leica DM LB2, Leica Microsystems, Germany) at 10x100 magnification. In the same sample the average diameter of diatom cells at the start and end of the experiment was measured. Algae were counted in 2ml wells under an inverted microscope (Zeiss Primovert, Carl Zeiss AG, Germany) at 20x10 magnification after gentle mixing of the cultivation bottle. Algae cells incorporated in biofilms after day 9 in the ~~bacteria enriched~~BAC+ cultures were counted after sonication in a sonication bath until all cells were in suspension. Nutrient and DOC samples were sterile filtered (0.2µm) and stored at -20°C before measurements. Nutrients were measured in triplicates after using standard colorimetric on a nutrient analyser (QuAAtro 39, SEAL Analytical, Germany) using the protocols No. Q-068-05 Rev. 12 for nitrate (detection limit = 0.02 µmol L^{-1}), No. Q-068-05 Rev. 12 for nitrite (detection limit = 0.02 µmol L^{-1}), No. Q-066-05 Rev. 5 for silicate (detection limit = 0.07 µmol L^{-1}), and No. Q-064-05 Rev. 8 for phosphate (detection limit = 0.01 µmol L^{-1}). The data were analysed using the software AACE. The nutrient analyzer was calibrated with reference seawater (Ocean Scientific International Ltd., United Kingdom). Ammonium was measured manually using the colorimetric method after McCarthy et al., (1977) on a spectrophotometer (Shimadzu UV-1201, detection limit = 0.01 µmol L^{-1}). DOC was measured by high temperature catalytic oxidation (HTCO) using a Shimadzu TOC-5000 total C analyser following methods

for seawater samples (Burdige and Homstead, 1994). The photosynthetic quantum yield was determined using an Aquapen PA-C 100 (Photon Systems Instruments, Czech Republic).

Certain factors, such as grazing, settling out of the euphotic zone, and bacterial and algae succession were not included into the experimental set-up to reduce complexity, and focus on nutrient dynamics. Trace metals, phosphate, and Vitamin B12 in coastal systems are assumed to be not limiting in Arctic coastal systems and were supplied in excess to the culture medium. Realistic pre-bloom DOC concentrations were present in the medium as it was prepared with sterilized seawater from the Fjord outside Tromsø before the onset of the spring bloom (March 2018).

All plots were done in R. The f-ratio as indication for the importance of regenerated production (Eppley, 1981) was calculated based on the average PON fixation in the last three days of the experiment~~.~~ (Eq C1). Here, nitrogen assimilation in the ~~axenic~~BAC- culture was assumed to be based on new (nitrate based) production, while fixation in the ~~bacteria-enriched~~BAC+ experiment was assumed to also be based on regenerated (ammonium based) production.

2.2 ~~Modelling~~Model structure

This section outlines ~~briefly~~ the overall model structure followed by a ~~short~~ description of the chosen parametrization approach for each relevant process. Details regarding model equations are provided in the Appendix (Table A1~~).~~) and a schematic representation of the models is given in Figure 1. We used a dynamic cell quota model by Geider et al. (1998) to describe the BAC- experiment (G98). We then extended the G98 model to represent the role of silicate limitation, bacterial regeneration of ammonium, and different kinetics for ammonium and nitrate uptake (EXT) and fitted it to the BAC+ experiment while retaining the parameter values estimated for G98.

The Geider et al. (1998) model (G98) ~~was used as a simple cell quota model to describe~~describes the response of phytoplankton to different nitrogen and light conditions~~. The G98 model~~ and is based on both intracellular quotas and extracellular ~~nutrient~~dissolved inorganic nitrogen (DIN) concentrations, allowing decoupled C and N growth (Fig. 1). ~~. It~~Within this model, light is a control of photosynthesis and chlorophyll synthesis. C:N ratios and DIN concentrations control nitrogen assimilation, which is coupled to chlorophyll synthesis and photosynthesis. Chl:N ratios are controlling photosynthesis and chlorophyll synthesis. G98 has been used in a variety of large scale ecosystem models with some extensions representing the actual conditions in the environment or mesocosms (e.g. Moore et al., 2004; Schartau et al., 2007; Hauck et al., 2013).

Photoacclimation dynamics in Geider type models have been evaluated as quick and robust (Flynn et al., 2001), while the N-assimilation component has some shortcomings in regard to ammonium-nitrate interactions. The original model of Geider et al. (1998) for C and N was corrected for minor typographical errors (see Ross and Geider, (2009~~),~~); Appendix Tables A6 A7~~) and afterwards extended to represent dynamics and interactions of silicate, nitrate and ammonium uptake, carbon and nitrogen excretion and bacterial remineralisation. The~~).

One aim of the study was to develop a model (EXT) with simplified dynamics of nutrient co-limitation, which is suitable for future implementation in coupled biogeochemistry-circulation models. The EXT model keeps all formulations of the G98 and adds dynamics and interactions of silicate, nitrate and ammonium uptake, carbon and nitrogen excretion and bacterial remineralisation (Fig. 1). The aim of the

model was to describe the response in photosynthesis, chlorophyll synthesis and nitrogen assimilation with a minimal number of parameters. Hence, dynamics in silicate cycling and bacterial physiology were highly simplified. The limitations of these simplifications and the potential need for more complex models are discussed later.

Silicate uptake was modelled using Monod kinetics after Spilling et al. (2010). The response of silicate limitation on photosynthesis and chlorophyll synthesis was implemented after findings by Werner (1978), Martin-Jézéquel et al. (2000), and Claquin et al. (2002). Werner (1978) found that silicate limitation can lead to a 80% reduction in photosynthesis and a stop of chlorophyll synthesis in diatoms within a few hours. ~~Nitrogen~~Hence, we added a parameter for the reduction of photosynthesis under silicate limitation (Si_{PS}) and formulated a stop of chlorophyll synthesis under silicate limitations.

N and Si metabolism have different controls and intracellular dynamics, with N uptake fuelled by photosynthesis (as P_C^{ref} in G98) and Si mainly fuelled by heterotrophic respiration (Martin-Jezequel et al., 2000). Besides earlier cultivation studies, the reduction of photosynthesis after Si limitation has been shown via photophysiological (inhibited PSII reaction centre, Lippemeier et al., 1999 ~~-~~) and molecular (down-regulated photosynthetic proteins, Thangaraj et al., 2019) approaches.

In general, we assume that nitrogen metabolism is ~~typically~~ not directly affected by silicate limitation (Hildebrand 2002, Claquin et al., 2002), but ~~the growth rate can~~we expect cellular ratios to be affected by reduced photosynthesis and chlorophyll synthesis under Si limitation (Hildebrand, 2002; Gilpin, 2004). The algal respiration term included both respiration and excretion of dissolved organic nitrogen and carbon as a fraction of the carbon and nitrogen assimilated. For testing the importance of DON excretion we also ran the EXT model without DON excretion (EXT_{-excr}). Dissolved organic nitrogen (DON) was recycled into ammonium via bacterial remineralisation. It was assumed that this process is faster for freshly excreted DON compared to DON already present in the medium. Thus, we implemented a labile (DON_l) and refractory (DON_r) DON pool with different remineralization rates (rem, rem_d). We also assumed that excreted DON and DOC do not coagulate as extracellular polymeric substances (EPS) during the course of the experiment. After Tezuka (1989), net bacterial ~~remineralisation~~regeneration of ammonium occurs at ~~substrate~~DOM C/N mass ratio below 10 and is proportional to bacterial abundances. Higher thresholds up to 29 have been found (e.g. Kirchmann, 2000), but we selected a lower number to stay conservative. ~~Substrate~~DOM C/N ratios are assumed to be proportional to algae C/N ratios (van der Meersche et al., 2004), with algal C/N ratios below 10 representing substrate (DOM) C/N ratios below 10.5. Hence, we assume net bacterial ~~remineralisation~~ammonium regeneration to occur at POC/PON ratios below 10, while higher ratios lead to bacteria retaining more N for growth than they release. Bacteria abundance change was estimated using a simple logistic growth curve as a function of DOM since the number of parameters is low (2) and the fit sufficient for the purpose of modelling algae physiology.

Michaelis-Menton kinetics based on bacteria growth on DOM with different labilities kinetics could give a more accurate representation of bacterial growth, but would not change the fit of the other model parameters aiming for the best fit of the model output to algal PON, POC, Chl, and DIN. Algal nitrate uptake was modelled after the original model by Geider et al. (1998) and ammonium assimilation was based on the simplified SHANIM model by Flynn and Fasham (1997b), excluding the internal nutrient and glutamine concentrations. Ammonium uptake is preferred over nitrate (lower half saturation constant) and reduces nitrate assimilation if available above a certain threshold concentration of ammonium

(Dortch, 1990; Flynn, 1999). Ammonium is the primary product of bacterial regeneration N-compound after remineralization of DON. Nitrification was assumed to be absent, since the bacteria in our experiment are not known to be capable of nitrification.

2.3 Model fitting

The model was written as a function of differential equations in R ~~and all~~. All model equations are provided in the Appendix (Table A6~~).~~) and the R code is available in the supplement. The differential equations were solved using the ode function of the deSolve package (Soetart et al., 2010) with the 2nd-3rd order Runge-~~Kutte~~Kutta method~~. After sensitivity analyses using the sensFun function of the FME package (Soetart and Petzoldt, 2010) (Fig. B1), and collinearity tests using the collin function and pairs plots (Fig. B2) the parameters not available from the experimental data (14~~ with automated stepsize control. deSolve is one of the most widely used packages for ~~our~~solving differential equations in R. Parameter of the G98 model~~, 6 for G98)~~ were fitted ~~based on~~ to the BAC- experiment data and the ~~data of both~~ EXT model was fitted to the ~~axenic~~BAC+ experiment ~~(i.e.~~data. The G98 parameter values were fitted first and retained without ~~remineralisation) and~~ changes for the ~~bacteria enriched experiment (i.e. with remineralisation). The aim was to reach an optimal fit for both the axenic and bacteria enriched experiment using the same values for the parameters.~~

EXT model fitting. The ~~first parameter fitting was done using the traditional G98 model. The parameters~~ maximum ~~gChl:gN~~Chl:N ratio (θ^{N}_{max}), minimum and maximum ~~gN:gC~~N:C ratios (Q_{min}, Q_{max}), and irradiance (I) ~~were~~are given by the experimental data and needed no further ~~tuning.~~fitting (Table A2). The start values and constraints for the remaining six variables (ζ, R^{C}, α_{Chl}, n, K_{no3}, P^{C}_{ref}, Table A3) were based on model fits of G98 to other diatom cultures in previous studies (Geider 1998, Ross and Geider 2009). The parameters were first fitted manually via graphical comparisons with the experimental data (POC, PON, Chl, DIN, Fig. 5 and 5), and via minimizing the model cost calculated as the root of the sum of squares normalized by dividing the squares with the variance (RMSE Eq. C2, Stow et al., 2009). ~~Maximum C specific photosynthesis (P^{C}_{ref}) and C based maintenance metabolic rate (R^{C}) were collinear and only P^{C}_{ref} was fitted. Manual parameter fitting was done using constraints~~ The initial manual tuning approach allowed control of the model dynamics, considering potential problems with known limitations of the G98 model (e.g lag phase not modelled; Pahlow, 2005). The manual tuning also allowed obtaining good start parameters for the automated tuning approach and sensitivity/ collinearity analyses, which are sensitive to the start parameters.

After the manual tuning, an automated tuning approach was used to optimize the fits. The automated tuning was done using the FME package (Soetart et al., 2010b), a package commonly used for fitting dynamic and inverse models based on differential equations (i.e. deSolve) to measured data. The automated analyses were based on minimizing the model cost calculated as the sum of squares of the residuals (SSR, Fitted vs measured data). The experimental data were normalized so that all normalized data were in a similar absolute range of values. This involved increasing Chl and PON values by an order of magnitude while decreasing DIN (NH_4 + NO_3) data by one order of magnitude The data were not weighted, assuming equal data quality and importance. Prior to the automated fitting, parameters were tested for local sensitivity (SensFun) and collinearity, or parameter identifiability (collin; e.g. Wu et al., 2014). sensFun tests for changes in output variables at each time point based on local perturbations of the

model parameter. The sensitivity is calculated as L1 and L2 norms (Soetart et al. 2009; Soetart et al., 2010b). The sensFun output is further used as input for the collinearity, or parameter identifiability analyses. Parameters were considered collinear and not identifiable in combination with a collinearity index higher than 20 (Brun et al., 2001). In this case, only the more sensitive parameter was used for further tuning. Eventually, R^C, K_{no3}, n, and α_{Chl} were subject to the automated tuning approach using the modfit function, based on minimizing the SSR within the given ~~by earlier studies (Geider et al., 1998; Ross and Geider, 2009). The model was fitted manually to reach an optimal fit for both the axenic and bacteria enriched experiment using the same values for the parameters, (Fig. 3,4), considering known limitations in the lag and stationary phase. The modFit function, using the~~constraints. Parameters were first fitted using a Pseudorandom search algorithm~~, was based on~~ (Price, 1977) to ensure a global optimum. The resulting parameters were then fine-tuned using the ~~mod~~Cost function of Nelder-Mead algorithm (Soetart et al., 2010b) for finding a local optimum. A model run with the ~~FME package and used to test whether potential substantial improvements~~new parameters was then compared to the initial model via graphical comparisons of the model fit ~~using different~~to the experimental data, and via the RMSE value.

The parameter values ~~could be achieved, but this was not the case so manual fits~~obtained for the G98 fit to the BAC- experiment were retained~~. The G98 model based parameters were kept for the tuning of the~~ without changes or further fitting in the EXT model. The additional parameters of the ~~extended model aiming to use the same parameter values for both experiments.~~EXT model were then fitted to the BAC+ experimental data (POC, Chl, PON, DIN). The model was only fitted to total DIN, due to the potential uncertainties related to ammonium immobilization in the biofilm. In fact, a test run, fitting the EXT model to NO_3 and NH_4 separately lead to a substantially worse overall fit (RMSE=8.79). Otherwise, the data were not weighted. Since the aim of the study was to model the effects of silicate and bacteria on algae growth and not to develop an accurate model for bacteria biomass and silicate concentrations, the parameters μ_{bact}, $bact_{max}$, K_{si}, and V_{max} were only fitted to the corresponding data (Bacteria, Silicate) prior to fitting the other parameters of the EXT model. Bacterial growth parameters (~~μ~~$_{bact}$$\mu_{bact}$, $bact_{max}$) were ~~determined in~~fitted to the bacterial growth ~~experiment~~curve. Silicate related parameters (K_{si}, V_{max}~~, s_{min}~~) were constrained by the study of Werner (1978~~).~~) and fitted to the measured silicate concentrations. The remaining parameters were subject to the tuning approach described for G98. Ammonium related parameters (K_{nh4}, $nh4_{thres}$) were constrained ~~loosely~~ by measured ammonium concentrations, and ~~remineralisation~~constants available for other diatom taxa described by Eppley et al. (1969). Remineralization parameters for excreted (rem~~,~~) and background (rem_d) DOM were ~~left unconstrained. Collinearity tests, and manual~~constrained by the data with the limitation of rem > rem_d, assuming that the excreted DOM is more labile. The parameters related to the effect of silicate limitation on photosynthesis and ~~automated parameter fitting~~chlorophyll production (s_{min}, Si_{PS}) were ~~done~~constrained by the study of Werner (1978) and fitted as described for ~~the~~ G98 ~~model.~~. None of the added parameters were collinear/ unidentifiable or given by the measured data and thus retained for the automated tuning approach. Eventually, the ~~14~~15 parameters (Table A3) were fitted against 160 data points (Table A1).

Due to the biofilm formation in the stationary phase of the BAC+ experiment, we tested three additional modelling approaches representing different dynamics in biofilms: i) DOC coagulation to EPS as part of the POC pool (Schartau et al., 2007), ii) Increased DOM excretion in the stationary phase (e.g. Christie-Oleza et al., 2017), and iii) Increased bacterial regeneration in the biofilm due to closer contact between

algae and bacteria (Equations in Table S1). However, we suggest that the photosynthesis reduction term Si_{PS} can give very similar model outputs, while being similarly or more sensitive. Thus, we tested the sensitivity of the added parameters of the three extended biofilm models in comparison to Si_{PS} by testing the magnitude of perturbations of Si_{PS} needed to reverse the effects of the added biofilm parameter (Fig. S1-3). In every case, the effects could be reversed with similar or less perturbations of Si_{PS}. The main effect of the biofilm that we could not model with the available data appears to be ammonium immobilization in the biofilm, either due to adsorption, accumulation in pockets, or conversion to ammonia due to the locally reduced pH caused by increased bacterial respiration. Model stability was estimated by extending the model run for 30 days, to test for unrealistic model dynamics (Fig. S4).~~The model cost was estimated via calculating the root of the sum of squares normalized by dividing the squares with the variance (RMSE Eq. (C1), Stow et al., 2009).~~

3 Results

3.1 Cultivation experiment

The concentrations of nitrate and silicate declined rapidly over the course of the experiment (Fig. ~~1~~2). After eight days, silicate decreased to concentrations below 2 µmol L^{-1} a threshold known to limit diatom dominance in phytoplankton (Egge and Aksnes, 1992), while inorganic nitrogen (nitrate, nitrite, and ammonium) became limiting (<0.5 µmol L^{-1} , POC:PON >8-9 DIN:DIP<16) only in the ~~axenic~~BAC- culture. DIN:DIP ratios far below 16, or DIN concentrations below 2 µmol L^{-1} have been described as indication for DIN limitation (Pedersen and Borum, 1996), as well as POC:PON ratios >9 (Geider and La Roche, 2002). Phosphate was not potentially growth limiting with molar DIN to PO_4 ratios consistently far below 16 (Redfield, 1934) and concentrations around 15 µmol L^{-1}. Typically, phosphate concentrations below 0.3 µmol L^{-1} are ~~typically~~ considered limiting (e.g. Haecky and Andersson, 1999). Regeneration of ammonium and phosphate were important ~~by the start of the stationary phase~~after eight days as seen by increasing concentrations of both nutrients and showed ~~–~~higher concentrations in the ~~bacteria enriched~~BAC+ experiments compared to the ~~axenic~~BAC- cultures (Fig. ~~1a~~2a,b). Ammonium concentrations were consistently higher, and nitrate was removed more slowly in the presence of bacteria, especially during the exponential phase. With the onset of the stationary phase in the BAC+ experiment, PO_4 and NH_4 concentrations doubled within 2 to 4 days and stayed high with variations in phosphate concentrations, while they stayed low in BAC-. With depletion of NO_3 in BAC+, NH_4 concentrations remained high, while PO_4 concentrations dropped. While not all ammonium measured is also available for algae growth, discussion of the dynamics (decrease in the start, increase with the onset of the stationary phase), especially if also shown in the EXT model, are still useful to understand multinutrient dynamics (e.g. regeneration). Considering the overall higher concentrations of NO_3, compared to NH_4, discussions of total DIN dynamics, DIN.DIP ratios, and limitations are also meaningful. DOC values were very high from the start (approx. 2-4 mmol L^{-1}) and remained largely constant throughout the experiment (Table A8).

The diatom *Chaetoceros socialis* grew exponentially in both treatments until day 8 before reaching a stationary phase with declining cell numbers (Fig. ~~2~~3). The growth rate of the ~~axenic~~BAC- culture (0.36

d^{-1}) was slightly lower than in the treatment with bacteria present (0.42 d^{-1}) during the exponential phase. Algal cellular abundance was higher in the ~~bacteria-enriched~~BAC+ cultures. Towards the end of the exponential phase, the diatom started to form noticeable aggregates in cultures with bacteria present, but only to a limited extent in the ~~axenic~~BAC- cultures. Such aggregate formation with associated EPS production is typical for *C. socialis*. With the onset of the stationary phase in the ~~bacteria-enriched~~BAC+ cultures about 30% of the cells formed biofilms on the walls of the cultivation bottles (estimated after sonication treatment). ~~-~~Bacteria (Fig. ~~2~~3) continued to grow throughout the entire experiment, but growth rates slowed down from 0.9 to 0.6 after day 8. In the ~~axenic~~BAC- cultures, bacterial numbers increased after 8 days, but abundances remained two order of magnitude below the ~~bacteria enriched~~BAC+ cultures and effectively ~~axenic~~BAC- over the experimental incubation period. The maximum photosynthetic quantum yield (Fv/Fm) is commonly used as a proxy of photosynthetic fitness (high QY~~) increased~~), indicating the efficiency of energy transfer after adsorption in photosystem II. Low values are typically related to stress, including for example nitrogen (Cleveland and Perry, 1987), or silicate (Lippemeier et al., 1999) limitation. We found an increase in QY from approx. 0.62 to 0.67 d^{-1} in the exponential phase and ~~decreased~~a decrease to approx. 0.62 in the ~~bacteria-enriched~~BAC+ treatment after 8 days and to approx. 0.58 in the ~~axenic treatments~~BAC- treatment (Table A8).

During algal exponential growth, POC and PON concentrations followed changes in algal abundances increasing four, seven, and 19-fold respectively, within 8 days (Fig. ~~2a, 3~~3a, 4). Interestingly, with the beginning of the stationary phase, POC and PON continued to increase in the ~~bacteria-enriched~~BAC+ cultures, while their concentrations stayed constant (POC), or decreased due to maintenance respiration (PON) in ~~axenic~~BAC- cultures. POC and PON concentrations were consistently higher (1.2 times POC, 1.4 times PON) in ~~bacteria-enriched~~BAC+ cultures during the exponential phase. gC : gN ratios decreased in both cultures, but increased again after 11 days in the ~~axenic~~BAC- culture. Chlorophyll *a* concentrations also increased exponentially over the first eight days in both treatments, and thereafter decreased within the stationary phase in the ~~axenic~~BAC- cultures. In contrast, cell numbers remained nearly constant in the ~~bacteria-enriched~~BAC+ cultures, before declining at the last sampling day.

Overall, both experimental cultures showed similar growth dynamics until day 8, with silicate becoming limiting for both treatments and nitrogen only limiting in ~~axenic~~BAC- cultures. Algal growth with bacteria present was slightly, but consistently higher during this phase. After eight days, algae growth stopped in both treatments, but nitrogen and carbon were continuously assimilated in ~~bacteria-enriched~~BAC+ cultures. ~~Axenic~~BAC- cultures started to degrade chlorophyll, while it stayed the same in ~~bacteria-enriched~~BAC+ cultures. Algal abundances in the ~~bacteria-enriched~~BAC+ treatment at the end of the experiment were ca 30% higher due to biofilm formation, and considerably more carbon (2x total POC, or 10-20% per cell) and nitrogen (3x total PON) per cell had been assimilated, and considerably more chlorophyll (2-3x total chlorophyll) produced at day 16. Cell size differences were not detectable (ca 4µm diameter, Table A8). POC to PON ratios increased after 11 days in ~~axenic~~BAC- cultures to maximum values of 7.2 and 1.3 mmol L^{-1}, respectively, but showed no change in ~~bacteria-enriched~~BAC+ cultures. POC to Chl ratios were comparable in both treatments (Fig. ~~4~~5). Assuming ~~axenic~~BAC- N fixation was mostly based on new production (nitrate as N source), while the algal N fixation in bacterial enriched treatments was based on new and regenerated (ammonium as N source) production, two-thirds of the production was based on regenerated production (f-ratio = 0.31).

3.2 Modelling

A comparison of the traditional G98 model with the ~~extended~~EXT model allowed an estimate of importance of bacterial DIN regeneration and Si co-limitations for describing the experimental growth dynamics. The ~~extended~~EXT model led to ~~no~~a slightly improved fit to the ~~axenic~~BAC- experiment ($RMSE_{G98}$ = 3.64 $RMSE_{EXT}$ = 3.34, Fig. 5 & 6). The real strength of the ~~extended~~EXT model was in representing growth dynamics with bacteria present (Fig. 5 & 6). Here, the fitted lines mostly overlapped with the range of measured data and the RMSE was reduced by 55% from $RMSE_{G98}$ = 4.57 down to $RMSE_{EXT}$ = 2.12.

Both, the G98 and ~~extended~~EXT model fits of the ~~axenic~~BAC- experiment were equally good for POC and PON with a slightly lower modelled growth rate. However, both models had limitations in modelling chlorophyll production, which was underestimated by about ~~50~~20% at the onset of the stationary phase (Fig. ~~4c~~5c). The degradation of chlorophyll *a* in the stationary phase was not modelled either (Fig. ~~4c). The bacteria enriched~~5c). PON in the BAC+ experiment was poorly modelled without consideration of silicate limitation or regenerated production specifically towards the end of the exponential phase and during the stationary phase. Maximum ~~POC,~~ PON ~~and Chl~~ values were ~~2~~ about 3 times lower using the G98 model (Fig. B3). In addition, the start of the stationary phase in the ~~bacteria-enriched~~BAC+ experiment was estimated 3 days too late via G98, even though modelled DIN was depleted 2 days too soon (Fig. B3). Under ~~axenic~~BAC- conditions, where silicate limitation does not play a major role the G98 model appears sufficient.

The ~~extended~~EXT model allowed representing detailed dynamics in a bacteria influenced system such as the responses to silicate limitation with a decrease in POC production, continued PON production, and the stagnation of Chl synthesis (Fig. ~~4~~5). Apart from the lag phase, the mass ratios of C:N and C:Chl were represented accurately (Fig. ~~4~~5). The model fits without the separate carbon excretion term (x_f) were ~~almost identical~~overall similar to the model with excretion, indicating the importance of the high background dissolved organic matter (DOM) concentrations, rather than excreted DOM for the regenerated ammonium, and the lack of significant aggregation of excreted DOM (~~identical~~ ~~$RMSE_{Ex}$~~$RMSE_{EXT\text{-}exr}$ of 2.21).

DIN dynamics caused by ammonium – nitrate interactions were represented well (Fig. 6a). However, at the onset of the stationary phase, ammonium concentrations of the model were one order of magnitude lower than in the experiment, showing a major weakness (Fig. 6c). Increased weighting of ammonium during the model fitting led to a slightly better fit to ammonium, but a substantially worse fit of the model to POC, PON, and Chl ($RMSE_{EXT}$=8.79). This indicates that the problem lies with the ammonium data, which include immobilized ammonium in the biofilm, unavailable for diatoms growth, while the model assumes that all ammonium is available. Other potential differences in biofilms, were tested via different model extensions (DOC aggregation to EPS, increase DOM excretion, increased regeneration), but all dynamics (Table S1) could be explained by the Si_{PS} term of the EXT model (Fig. S1-3). ~~Fine-scale DIN dynamics caused by ammonium – nitrate interactions were represented well (Fig. 5a). However, at the onset of the stationary phase, ammonium concentrations of the model were one order of magnitude lower than in the experiment, showing a main weakness of the model (Fig. 5c).~~ The silicate uptake ~~is~~estimation was highly simplified using simple Monod kinetics, ~~with~~leading to too high modelled values in the

stationary phase and a too quick depletion in the start (Fig. ~~5d~~6d). Carbon excretion (x_f) did not have any effect on the model fit to nutrients.

The sensitivity analysis (Fig. B1, Table A1) revealed that the ~~extended model was most sensitive to R^C (max~~ sensitivity ~~>1).~~ ~~R^C is part of the original G98 model, showing that none~~ of the added parameters ~~was more complex than any of the original model parameters. Hence, the added complexity of the extended model does not create a strong new~~ in EXT is overall comparable to the sensitivity~~.~~ of the original parameters in G98. The model outputs were most sensitive to P_C^{Ref} (L1=0.8, L2=1.5), which is a parameter in both G98 and EXT. The most sensitive added parameters in EXT were the ~~maximum silicate uptake rate (V_{max}) and~~ remineralisation ~~rates (rem,~~ rate of refractory DON (rem_d~~) with values of the sensitivity analysis reaching ca 0.5, which is~~ , L1=0.24), the half saturation constant for ammonium (K_{nh4}, L1=0.08) and the inhibition of photosynthesis under Si limitation (Si_{PS}, L1=0.08), which was comparable to other ~~sensible~~sensitive parameters of the ~~original~~ G98 model (~~shape factor for photosynthesis (Q_{max}, R^C, α_{Chl}, ζ, n~~~~),~~, I, Θ_N^{max}~~). The most affected model~~, Table A1). Small perturbations of the parameters only indirectly related to the fitted output ~~by R^C and n was the~~ variables did not lead to changes in POC, PON, Chl, or DIN ~~concentration.~~ .

4 Discussion

The experimental incubations represented typical spring bloom dynamics for coastal Arctic systems, including an initial exponential growth phase terminated by N and Si limitation and the potential for an extended growth period via regenerated production. Our model incorporating these results was able to reflect these dynamics by adding NH_4-NO_3-$Si(OH)_4$ co-limitations and bacterial NH_4 regeneration to the widely used G98 model. In addition, bacteria-algae interactions and DOC and biofilm dynamics were important in the experiment, but those were not crucial for quantitatively modelling algal C:N:Chl quotas. While *C. socialis* may not be the dominant species in all coastal Arctic phytoplankton blooms, we argue that it is representative for chain-forming diatoms typically dominating these systems due to their shared needs and responses to nutrient limitations (e.g. Eilertsen et al., 1989; von Quillfeldt, 2005).

4.1 Silicon-nitrogen regeneration

Spring phytoplankton dynamics in Arctic and sub-Arctic coastal areas is typically characterised by an initial exponential growth of diatoms, followed by peaks of other taxa (like *Phaeocystis pouchetii*) soon after the onset of silicate limitation (Eilertsen et al. 1989). Thus, a shift in species composition for the secondary bloom is linked to silicate limitation prior to final bloom termination caused by inorganic nitrogen limitation. ~~The~~Photosynthesis was reduced by approx. 70% after silicate became limiting, which is comparable to earlier experimental studies (Tezuka, 1989). However, the secondary bloom was extended in time by bacterial regeneration of ammonium, allowing regenerated production to contribute about 69% of the total production (f-ratio=0.31) even during ~~a~~the diatom dominated scenario in our experimental incubation. With the start of the stationary phase, NH_4 and PO_4 concentrations doubled, presumably due to decreased assimilation by the silicate starved diatoms and increased regeneration by bacteria, supplied with increasing labile DOM (doubled remineralisation rate in EXT) excreted by the stressed algae. After NO_3 depletion at day 15, also PO_4 concentrations drop, indicating a coupling of N:P

metabolism. Excretion of organic phosphate by diatoms is also common in cultures with surplus orthophosphate (Admiraal and Werner, 1983), which can be another explanation of the phosphate peak after silicate becomes limiting. The presence of bacteria and –thus regenerated production allowed ~~algae~~diatom growth to continue 8 days after silicate became limiting (Figs. ~~1,~~2 ~~&,~~ 3 & 4), nearly doubling the growth period similar to observations in the field (e.g. Legendre and Rassoulzadegan, 1995; Johnson et al., 2007). ~~This extended production shows~~ The G98 model has its most severe limitation, the modelling of PON, simply due to the lack of the ammonium pool, supplied via bacterial regeneration. The substantially better fit of PON in the EXT model shows therefore clearly that bacterial remineralisation is crucial to successfully model spring bloom dynamics, especially near bloom termination. Many biogeochemical models used in the Arctic include remineralisation, but rely on fixed or temperature dependent rates and do not consider them bacteria-dependent (MEDUSA, LANL, NEMURO, NPZD, see Table 1). While this simplification allows modelling regenerated production, using bacteria-independent remineralisation rates does have limitations under spring bloom scenarios, where bacteria biomass can vary over orders of magnitudes (e.g. Sturluson et al., 2008) as also seen in our experimental study.

While we do not expect the f-ratio in our bottle experiment to be directly comparable to open ocean system, which does include a variety of algal taxa beyond *C. socialis*, a comparison can aid to identify limitations in our experiment and model. Regenerated production is significant in polar systems and our estimated experimental f-value of 0.31 is slightly below the average for polar systems (Harrison and Cota, 1990, mean f-ratio=0.54). Nitrification is a process supplying about 50% of the NO_3 used for primary production in the oceans, which may lead to a substantial underestimation of regenerated production (Yool et al., 2007), inflating the f-ratio interpreted as estimate for new production, potentially also in the study by Harrison and Cota (1990). The absence of vertical PON export in our experiment may ~~explain~~be another explanation for the above average fraction of regenerated production. In the ocean environment, regenerated production is also affected by vertical export (sedimentation) and grazing which are not represented in the experimental incubations. Via sedimentation, a fraction of the bloom either in the form of direct algal sinking of fecal pellets is typically exported to deeper water layers, reducing the potential for N regeneration within the euphotic zone (e.g. Keck and Wassmann, 1996). Larger zooplankton grazing can lead to increased export of PON via fecal pellet aggregation, or diel vertical migration (Banse, 1995), but may also release ammonium and urea (Conover and Gustavson, 1999, Saiz et al., 2013).

In contrast, bacterial death by microflagellate grazing and viral lysis may supply additional nutrients, or DON available for N regeneration in the euphotic zone (e.g. Goldman and Caron, 1985), which potentially leads to an overestimation of regenerated production. Another potentially important N source for regenerated production may be urea (Harrison et al., 1985), which would lead to an even higher importance of regenerated production as suggested by our study. Hence, ecosystem scale models will need to consider these dynamics regarding bacterial abundances, microbial networks and particle export in addition to bacterial remineralization in order to model realistic ammonium regeneration in the euphotic zone.

Bacteria-mediated silicate regeneration is absent from the ~~modeling~~modelling approach, as indicated by the identical silicate concentrations in both treatments and models (Fig. ~~1~~2). In the environment silicate dissolution is, in fact, mostly described as an abiotic process with temperature as the main control, and a minor contribution by bacterial remineralisation (Bidle and Azam, 1999). Our experiment indicates that

silicate dissolution for *Chaetoceros socialis* was negligible at cold temperatures and the time scale of the incubations and typical for bloom durations and residence times of algae cells in the euphotic zone (Eilertsen et al., 1989, Keck and Wassmann, 1996). We conclude that silicate dissolution in coastal Arctic systems happens most likely in the sediment or deeper water layers and is only supplied via mixing in winter. In Antarctica substantial silicate dissolution has been observed but not in the upper ~~100m~~100 m, which has been related to the low temperatures (Nelson and Gordon, 1981) in agreement with our conclusion. Hence, modelling silicate regeneration in the euphotic zone is not necessary in these systems.

4.2 Algal growth response to Si and N limitation

The response of diatoms to Si or N limitation is based on different dynamics and different roles of N and Si in diatom growth. N is needed for proteins and nucleic acids~~, while~~ and its uptake is mainly fueled by phototrophic reactions (Martin-Jézéquel et al., 2000). Si is only needed for frustule formation, mostly during a specific time in the cell cycle (G2 and M phase, Hildebrand, 2002~~).~~) and the assimilation mostly fueled by heterotrophic reactions (Martin-Jézéquel et al., 2000). Once N is limiting, growth rapidly stops (Geider et al. 1998). In the case of ~~SI~~Si limitation, however, growth can continue with a slower rate if N is still available (Werner, 1978~~).~~; Gilpin et al., 2004). Several studies found a reduced growth rate with weaker silicified cell walls (Hildebrand, 2002; Gilpin, 2004), but unaffected nitrogen assimilation under silicate limitation (Hildebrand 2002, Claquin et al., 2002) in accordance with our study. Claquin et al. (2002) found variable Si:C and Si:N ratios and highly silicified cells under nitrogen limitation, indicating uncoupled Si and N:C metabolism.

Nitrogen is a crucial element as part of amino acids and nucleic acids, which are necessary for cell activity and growth. If N becomes limiting major cellular processes are affected and growth or chlorophyll synthesis is not possible. Photosynthesis can continue for a while leading to carbon overconsumption (Schartau et al., 2007~~).~~), which is well modelled by G98 for both BAC+ and BAC-. A part of the excess carbon can be stored as intracellular reserves, and a part is excreted as DOC, which may aggregate as EPS, contributing to the total POC pool. The excess carbon can potentially be toxic for the algae and excretion and extracellular degradation by bacteria may be crucial for algal survival (Christie-Oleza et al., 2017). Quantitatively, N limitation is well modelled by G98 under ~~axenic~~BAC- conditions, if only one nitrogen source plays a role. However, under longer nitrogen starvation times or higher light intensities, alternative models that include carbon excretion and aggregation (Schartau et al., 2007) or intracellular storage in reserve pools (Ross & Geider 2009) might be needed. Our growth experiment shows clearly, that C:N ratios are not fixed and variable quotas are needed. Vichi et al. (2007) estimated that Carbon based models may underestimate net primary production (NPP) by 50%, arguing for the importance of quota based models (Fransner et al., 2018). However, most ecosystem scale models are simplified by using fixed C:N ratios (Table 1). The next step to quota based-models is the consideration of more detailed cell based characteristics, such as transporter density, cell size, and mobility, including sedimentation (Aksnes and Egge, 1991). Flynn et al. (2018) discuss a model with detailed uptake kinetics showing that large cells are overall disfavored over small cells due to higher half saturation constants, but that they may still have competitive advantages due to lower investment in transporter production. Also increased sedimentation in larger cells increases the mobility and may offset the disadvantage of a larger

size. While this extension is too complex for our aim of a simplified model, the dynamics may become important when modelling different algae taxa.

The type of inorganic nitrogen available also affects nitrogen uptake. ~~Ammonium~~ Due to the metabolic costs related to intracellular nitrate reduction to ammonium, ammonium uptake is ~~typically~~ preferred over nitrate ~~and can~~, potentially leaving more energy for other processes (Lachmann et al., 2019). Ammonium can even inhibit or reduce nitrate uptake over certain concentrations~~.~~ (Morris, 1974). The dynamics are mostly controlled by intracellular processes, such as glutamate feedbacks on nitrogen assimilation, cost for nitrate conversion to ammonium, or lower half saturation constants of ammonium transporters (Flynn et al., 1997). The most accurate representation of these dynamics are given in the ANIM model by Flynn et al. (1997), but the model is too complex for implementations in larger ecosystem models. The number of parameters is difficult to tune with the typically limited availability of measured data and its complexity makes it also computationally costly to scale the models up. Typically, modelling ammonium-nitrate interactions by different half-saturation constants and inhibition of nitrate uptake by ammonium appears sufficient (e.g. BFM, LANL, NEMURO, Table 1) and has been adapted in our model.

~~Silicate limitation affects mainly the cell cycle. Without silicate, diatoms cannot form new frustules needed for forming new cells. Nitrogen assimilation, photosynthesis, and synthesis of proteins and nucleic acids can continue at lower rates (Werner, 1978).~~Studies on the coupling of silicate limitation on C, N, and Chl show inconclusive patterns, including a complete decoupling (Claquin et al., 2002), a relation of N to Si (Gilpin et al., 2004) and reduction of photosynthesis without new chlorophyll is production (Werner, 1978; Gilpin et al., 2004). Cell size is limited by the frustules, but cells may become more nutritious (higher N:C ratio), or simply excrete more DOM, which may aggregate and contribute to the PON and POC pools. A detailed cell-cycle based model has been suggested by Flynn (2001), but ~~its complexity remains too high~~the number of parameters (30) makes the model too complex for ecosystem scale models. In ecosystem scale models Si limitation is modelled in various simplifications, such as thresholds ~~for absence~~triggering a stop (MEDUSA), ~~and reduced~~ or reduction (e.g. BFM, MEDUSA, SINMOD)of the Si dependent production (~~e.g. BFM, MEDUSA, SINMOD,~~ Table 1), or Si:N ratio scaled production (NEMURO, Table 1). ~~We~~

Our cultivation study shows i) that a threshold value in the model, leading to a stop or solely Si dependent photosynthesis has its limitations, since DIN controlled photosynthesis continues at lower rates, and ii) that coupling of Si:N:C:Chl is present. We do not expect a direct Si:N coupling, due to different controls of Si and N metabolism (Martin-Jézéquel et al., 2000.), but suggest indirect coupling via reduced photosynthesis. In fact, detailed photophysiological and molecular approaches under Si limitation found inhibited PSII reaction centers (Lippemeier et al., 1999) similar to the decreased QY in our experiment, and down-regulated photosynthetic proteins (Thangaraj et al., 2019) under Si limitation. Thus, we modelled the response of diatom growth to silicate limitation by reducing photosynthesis ~~by 80% and~~ through a parameterized fraction (Si_{PS}) and a stop of chlorophyll synthesis ~~under~~below a certain threshold, based on experimental studies (Werner, 1978, Lippemeier et al., 1999, Gilpin et al., 2004, Thangaraj et al., 2019) and in accordance to other ecosystem scale approaches. Automated fitting showed the same 80 % reduction of photosynthesis as described by Werner (1978). We suggest that this extension is more accurate than the typical threshold based dynamics, with one limiting nutrient controlling the growth equally for POC and ~~chl~~Chl production (e.g. SINMOD by Wassmann et al., 2006; BFM by Vichi et al.,

2007), while still keeping the ~~complexity low.~~number of parameters low compared to very detailed cell-cycle based models (e.g. Flynn, 2001, Flynn et al., 2018).

4.3 Importance of algae-bacteria interactions and DOC excretion

As described above, N or Si limitation can lead to excretion of DON and DOC, which can aggregate as EPS and be available for bacterial regeneration of ammonium. For including EPS dynamics in the model additional data would be needed. However, the importance of EPS formation is clear in the end of the ~~bacteria-enriched~~BAC+ experiment. Firstly, a biofilm was clearly visible containing about 30% of the algae cells. While we would not expect biofilms in the open ocean, aggregation of algae cells, facilitated by EPS is common towards the end of spring blooms, increasing vertical export fluxes (e.g. Thornton, 2002). *Chaetoceros socialis* is in fact a colony forming diatom building EPS-rich aggregates in nature (Booth et al., 2002). Secondly, POC and PON concentrations increased, while cell numbers and sizes stayed constant, showing that the additional POC and PON was most likely part of an extracellular pool. Silicate limitation could be one trigger for enhanced exudation. In fact, the three biofilm dynamics evaluated (DOC aggregation, increased excretion, increased regeneration) could all be modelled by the Si_{PS} term. Since the biofilm formation corresponds with silicate limitation, it is difficult to untangle the direct effects of the biofilm, or the indirect effects of silicate limitation, without additional data or experiments (e.g. EPS measurements, DOM characterization). However, only 30% of the culture was part of the biofilm and the best fit of 80% reduction for the Si_{PS} term corresponds very well with an earlier study by Werner (1978), who did not have biofilm formation. Hence, we suggest that the main cause for the reduction of photosynthesis is related to Si limitation and not the biofilm.

Interestingly, algae – bacteria interactions can be species specific with specific organic molecules excreted by the algae to attract specific beneficial bacteria (Mühlenbruch et al., 2018). Thereby bacteria are crucial for recycling ammonium, but also to degrade potentially toxic exudates (Christie-Oleza et al., 2017).

In the ~~axenic~~BAC- experiment, Carbon excretion after Carbon overconsumption could be expected after Schartau et al. (2007), but no indications, such as biofilm formation, or increased POC per cell were found. This indicates that carbon overconsumption has been of minor importance likely due to the low light levels. An alternative explanation is that bacteria and potentially chemotaxis are important controls on algal carbon excretion (Mühlenbruch et al., 2018). Overall, DOM excretion and EPS dynamics appear to play a minor role in quantitatively modelling C:N:Chl quotas in our experiment, with ~~identical RMSEs~~ (~~similar~~ $RMSE_{EXT-excr}$=2.21, $RMSE_{EXT}$=2.12) for a model run with and without the excretion term x_f. However, in systems with less allochthonous DOM inputs, such as open oceans compared to coastal sites, these dynamics will most likely play a more important role.

4.4 Considerations in a changing climate

Due to a rapid changing climate, especially in Arctic coastal systems, the dynamics addressed in this study will change (Tremblay and Gagnon 2009). With warmer temperatures, heterotrophic activities, and thereby bacterial recycling will increase (Kirchman et al., 2009). Our study showed that regenerated production is crucial for an extended spring bloom. Hence, higher heterotrophic activities may lead to

extended blooms (~~higher f-ratio).~~increased bacterial regeneration). At the same time, higher temperatures and increased precipitation will lead to stronger and earlier stratified water columns, which will lead to less nutrients reaching the surface by winter mixing, reducing new production (~~lower f-ratio~~decreased bacterial regeneration)(Tremblay and Gagnon, 2009; Fu et al., 2016). Consequently, the phenology of Arctic coastal primary production in a warmer climate will likely be increasingly driven by bacterial remineralization, showing the necessity to include this process into biogeochemical models. An earlier temperature driven water column stratification may also lead to an earlier bloom. However, due to increasing river and lake brownification and sediment resuspension, the spring bloom may also be delayed (Opdal et al., 2019)~~An earlier temperature driven water column stratification will also lead to an earlier bloom however with potentially lower light intensities~~. With decreased light, carbon overconsumption as described by Schartau et al. (2007) may become less important due to decreased photosynthesis. An earlier or later phytoplankton bloom can lead to a mismatch with zooplankton grazers (Durant et al., 2007; Sommer et al., 2007). Reduced zooplankton production would decrease the fecal pellet driven vertical export and thereby increase the residence time of POM in the euphotic zone and the potential for ammonium regeneration. Thus, the incorporation of bacterial recycling into ecosystem models may be even more important under this scenario.~~In this case, less light is available earlier in the Arctic spring season and carbon overconsumption as described by Schartau et al. (2007) may become less important. An earlier phytoplankton bloom can lead to a mismatch with zooplankton grazers (Durant et al., 2007; Sommer et al., 2007), which could decrease the fecal pellet driven vertical export and thereby increase the residence time of POM in the euphotic zone and the potential for ammonium regeneration, making the incorporation of bacterial recycling into ecosystem models even more imporatant as also evident from our experimental data and model output.~~ In fact, global climate change models agree that vertical carbon export is decreasing overall (Fu et al., 2016). Silicate regeneration is thought to be mostly controlled abiotically by temperature (Bidle and Azam, 1999). Thus, increasing temperature and a stronger stratification will allow recycling of silicate in the euphotic zone before sinking out and thus could cause a shift in the algal succession observed during spring with prolonged contributions of diatoms~~.~~ (Kamatani, 1982). Thus, a temperature dependent silica dissolution may need to be included for models in a substantially warmer climate in further model developments. Increased precipitation will also lead to increased runoff and allochthonous DOM inputs, increasing the importance of terrestrial DOM degradation and decreasing the relative importance of algal exudate regeneration (Jansson et al., 2008). The high fraction of regenerated production mostly based on allochtonous DOM degradation, the limited role of excreted DOM degradation, low light levels, and the absence of grazing and export fluxes are simplifications of our study, which are, however, expected to be realistic scenarios under climate change. Hence, we suggest that our experiment and model are well suited as a baseline for predictive ecosystem models investigating the impacts of climate change on coastal Arctic spring blooms. However, climate change may lead to shifts in algae communities with non-silicifying algae dominating over diatoms (e.g. Falkowski and Oliver, 2007), reducing the importance of silicate limitation. Thus, conducting similar experiments and modelling exercises with a wider range of algal taxa and different temperature and nutrient regimes is suggested.

Acknowledgements

The project was supported by ArcticSIZE - A research group on the productive Marginal Ice Zone at UiT (project number 01vm/h15). We want to thank Paul Dubourg and Elzbieta Anna Petelenz-Kurdziel for the help with Nutrient and POC/PON analyses. DOC analyses was supported through a Fulbright Distinguished Scholar Award to HRH.

Authors contributions

TRV designed the experiment with contributions by RG and ML. TRV isolated and identified the cultures. ML performed the experiment with contributions of TRV and UD. RH measured DOC and SK measured the Nutrients. The other parameters where measured by ML and TRV. TRV programmed the model with contributions of CV, ST and DvO. TRV wrote the manuscript with contributions from all co-authors.

Data availability

The experimental data are archived at DataverseNO under the doi number doi.org/10.18710/VA4IU9. The Rscripts for the model are available ~~from the corresponding author upon request~~at github under https://github.com/tvonnahm/Dynamic-Algae-Bacteria-model.

Competing interests

The authors declare that they have no conflict of interest.

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Figures

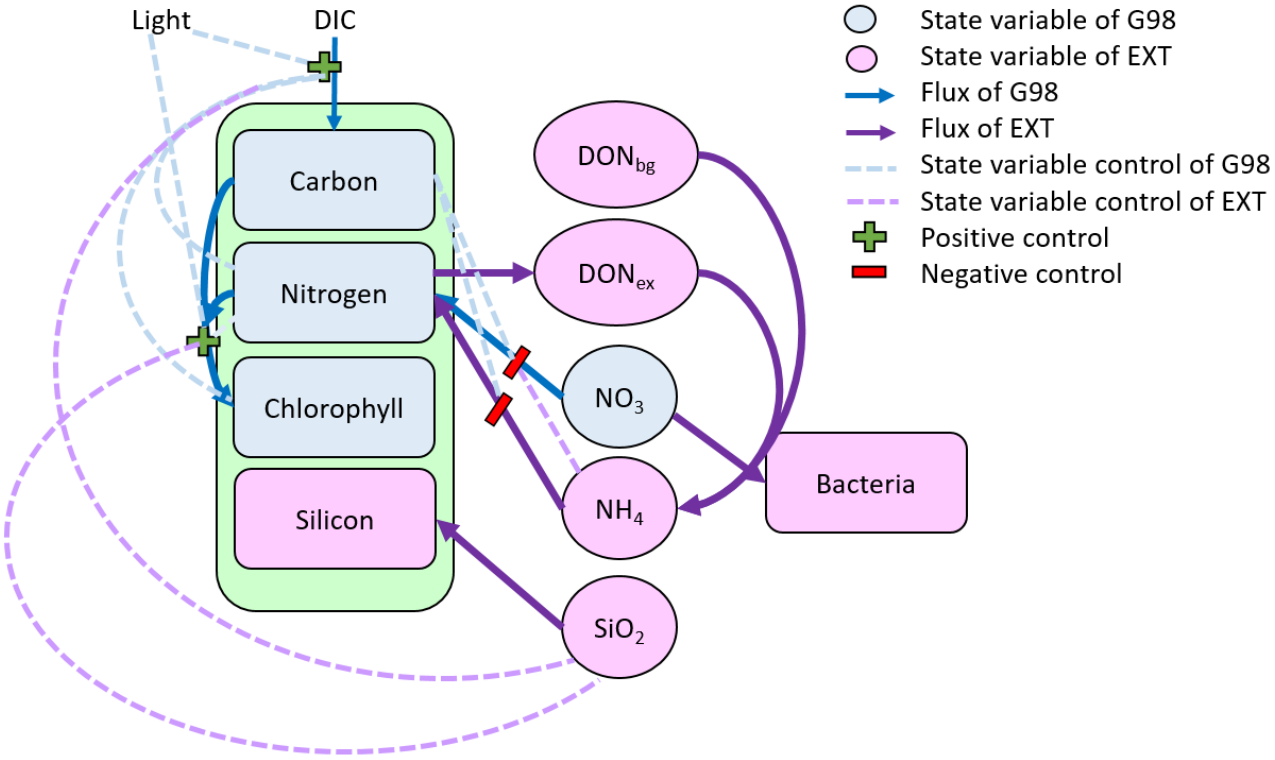

Figure 1. Schematic representation of the state variables and connections and controls in the G98 model (blue) and EXT model (purple). The EXT model has the same formulations as G98 with the additions shown in purple.

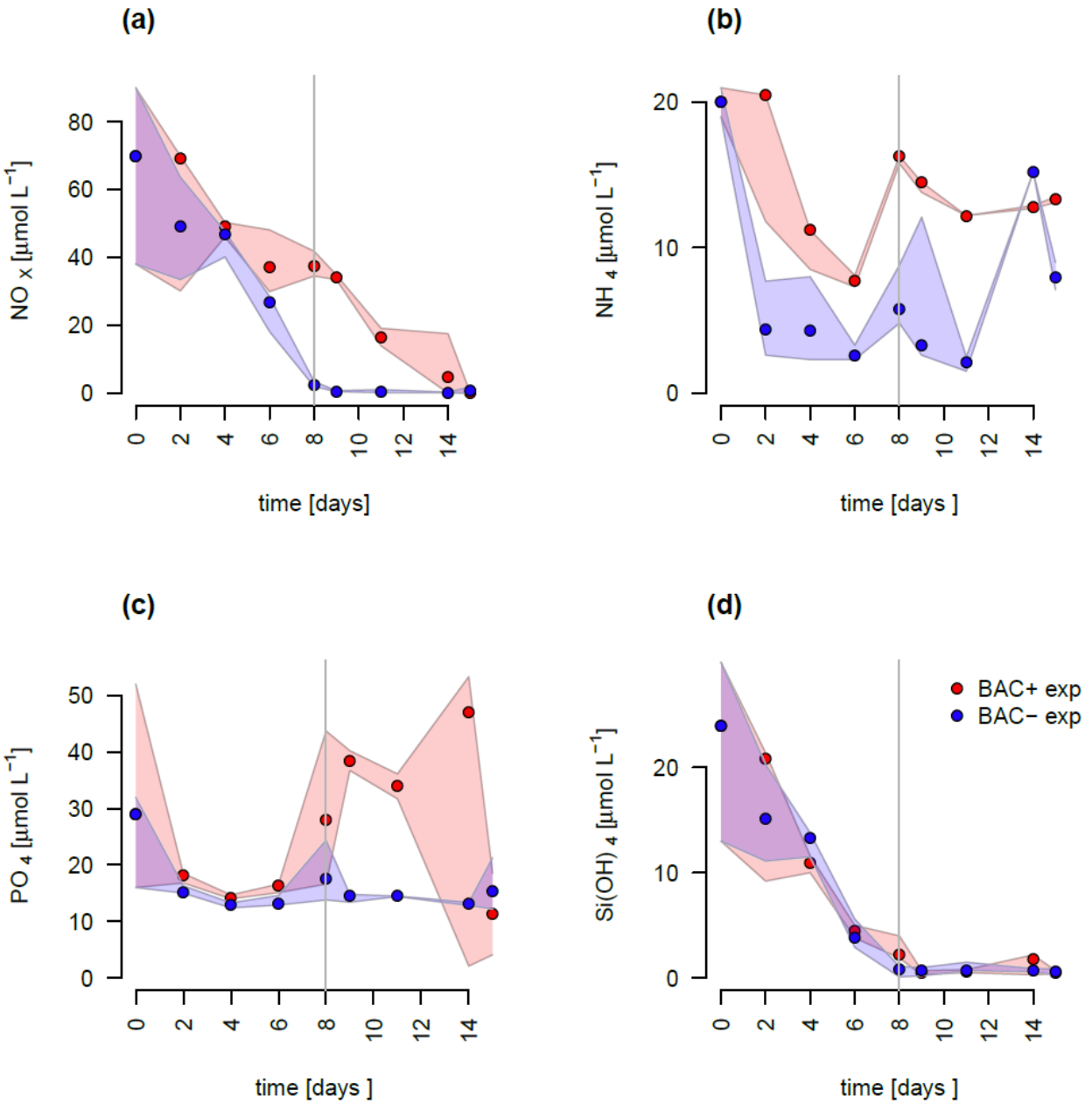

Figure 2. Nutrient measurements over the experimental incubations of a) NO_x, (NO_3^- + NO_2^-) b) NH_4^+, c) PO_4^{2-}~~,~~ with a potential outlier at day 14 leading to a negative peak, d) Silicate, red circles are ~~axenic~~BAC- cultures and green symbols are ~~bacteria-enriched~~BAC+ cultures. Circles show median values (blue = ~~axenic,~~BAC-, red = ~~bacteria)~~BAC+) and the ~~coloured~~colored polygons show ~~the total range~~maximum and minimum of measured data~~.~~ (n=3). The grey line shows the beginning of the stationary growth phase of *Chaetoceros socialis* ~~and the dotted horizontal line the threshold under which NO_x or silicate are limiting, or the threshold of NH_4^+ under which nitrate uptake is not inhibited~~.

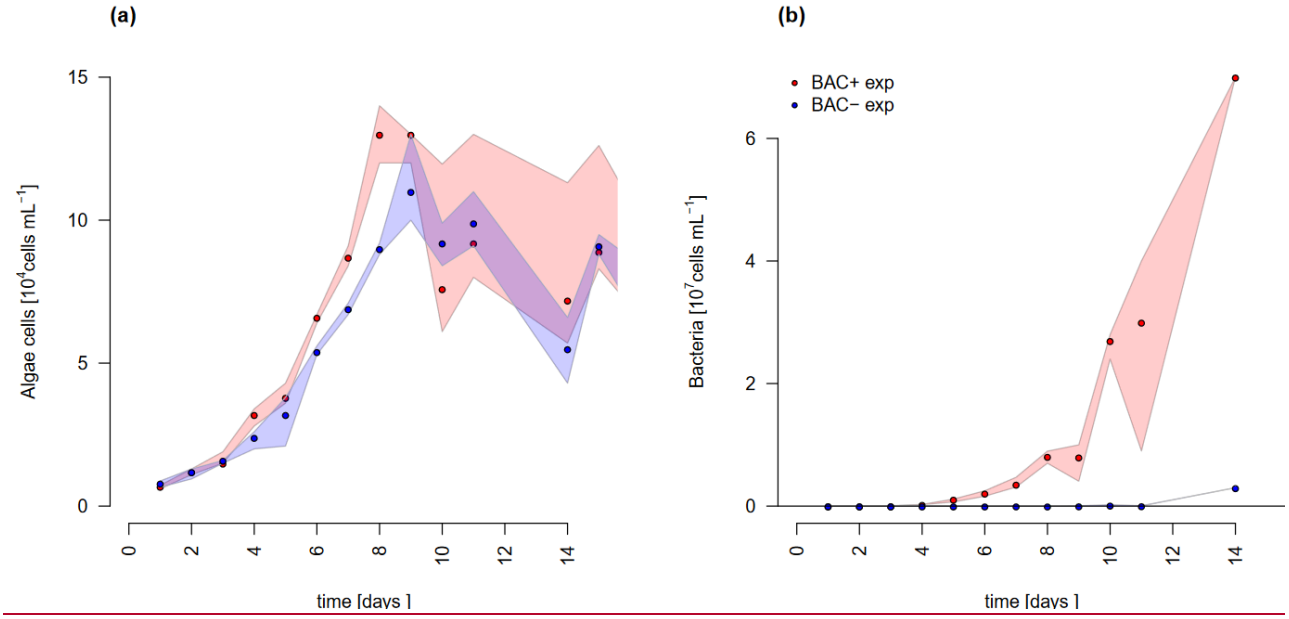

Figure ~~2~~3. Abundances of a) *Chaetoceros socialis* and b) bacteria over the 14 day experimental period. Blue data are from ~~axenic~~BAC- cultures and red from ~~bacteria-enriched~~BAC+ cultures. Circles represent median values (blue= ~~axenic,~~BAC-, red = ~~bacteria-enriched)~~BAC+) and the ~~coloured~~colored polygons show ~~the total range~~maximum and minimum of measured data (n=3) (Not visible for bacteria counts in ~~axenic~~BAC- cultures due to very small range). The maximum values of the ~~bacteria enriched~~BAC+ experiment includes algae cells in the biofilm (after day 9). ~~The grey line indicates the start of the stationary growth phase of *C. socialis*.~~

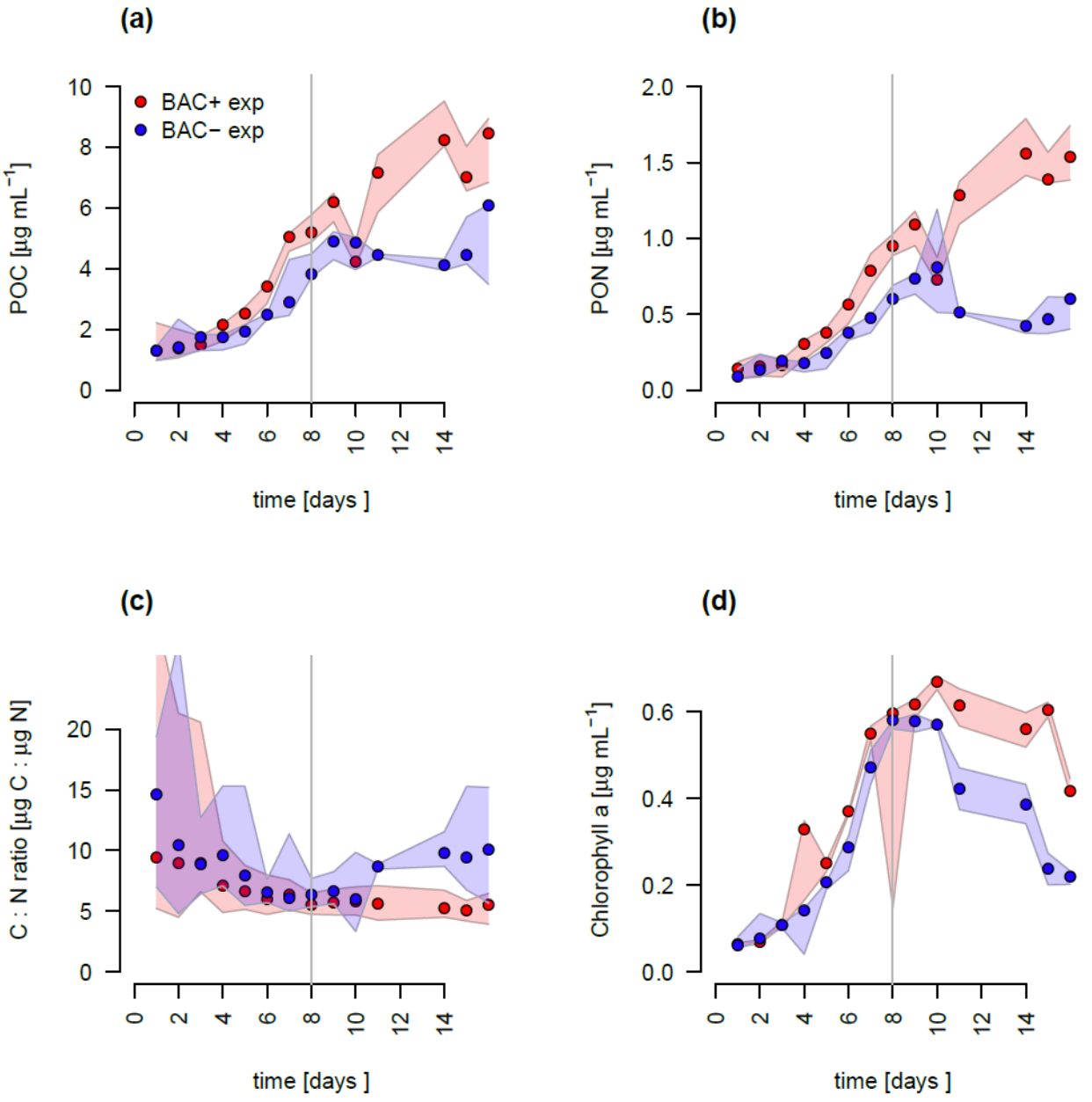

Figure ~~3~~4. Total particulate organic a) Carbon (POC) b) Nitrogen (PON), c) C : N ratios, and d) Chlorophyll a concentration in experimental cultures~~.~~ with a potential outlier at day 8, presumably due to photodegradation, causing a negative spike. Blue symbols are ~~axenic~~BAC- cultures and red show ~~bacteria-enriched~~BAC+ cultures. Circles show median values (blue = ~~axenic,~~BAC-, red = ~~bacteria enriched)~~BAC+) and the ~~coloured~~colored polygons show the ~~total range~~maximum and minimum of measured data~~.~~ (n=3). The grey line indicates the start of the stationary phase.

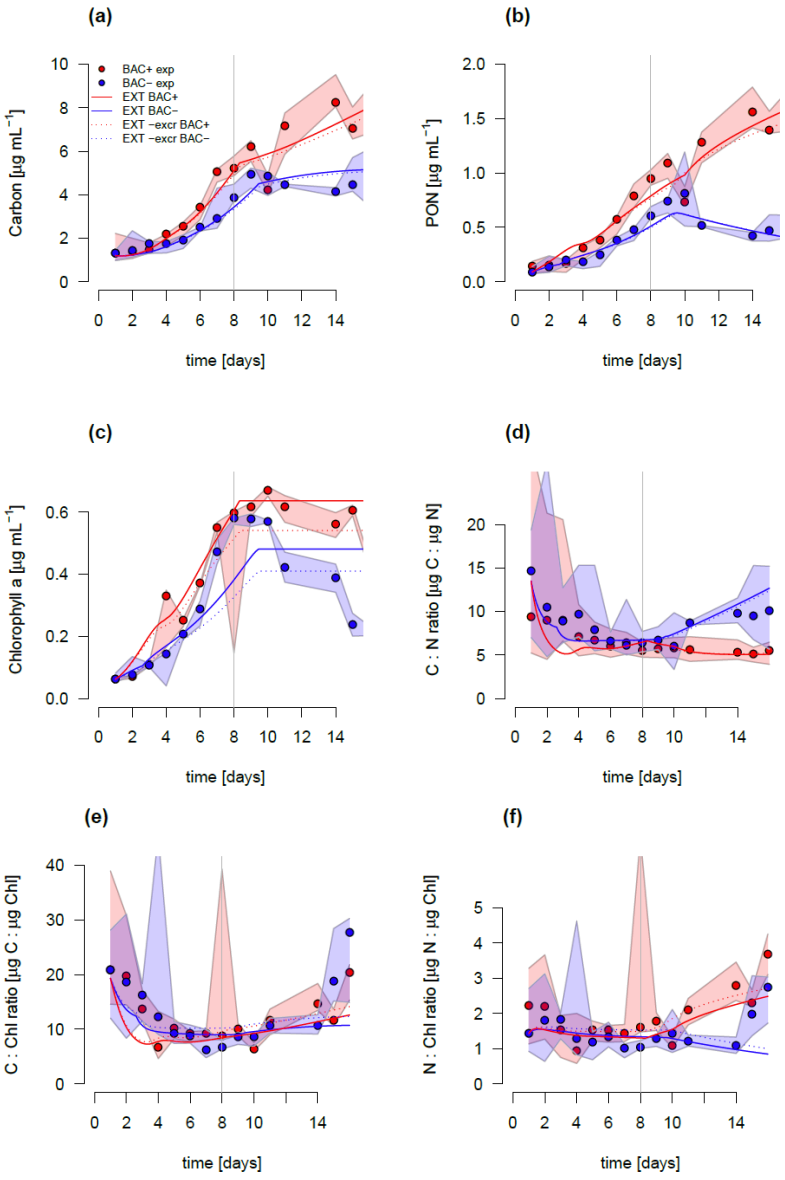

Figure ~~4:~~5. Model fit of the ~~extended~~EXT model to the ~~axenic~~BAC- (blue) and ~~bacteria enriched~~BAC+ (red) experiment. Circles show median values and the ~~coloured~~colored polygons show the ~~total range~~maximum and minimum of measured data~~.~~ (n=3). Solid lines show the model outputs of a) POC, b) PON, c) Chl~~,~~ (including an outlier at day 9 BAC+), d) C:N, e) C:Chl, and f) N:Chl. Dotted lines show the model fit without ~~the~~ additional Carbon excretion term x_f. At day 8 the threshold for silicate limitation is reached leading to reduced photosynthesis (by the factor given by Si_{PS}) and inhibited Chl synthesis, which is visible as sharp transitions in POC and Chl.

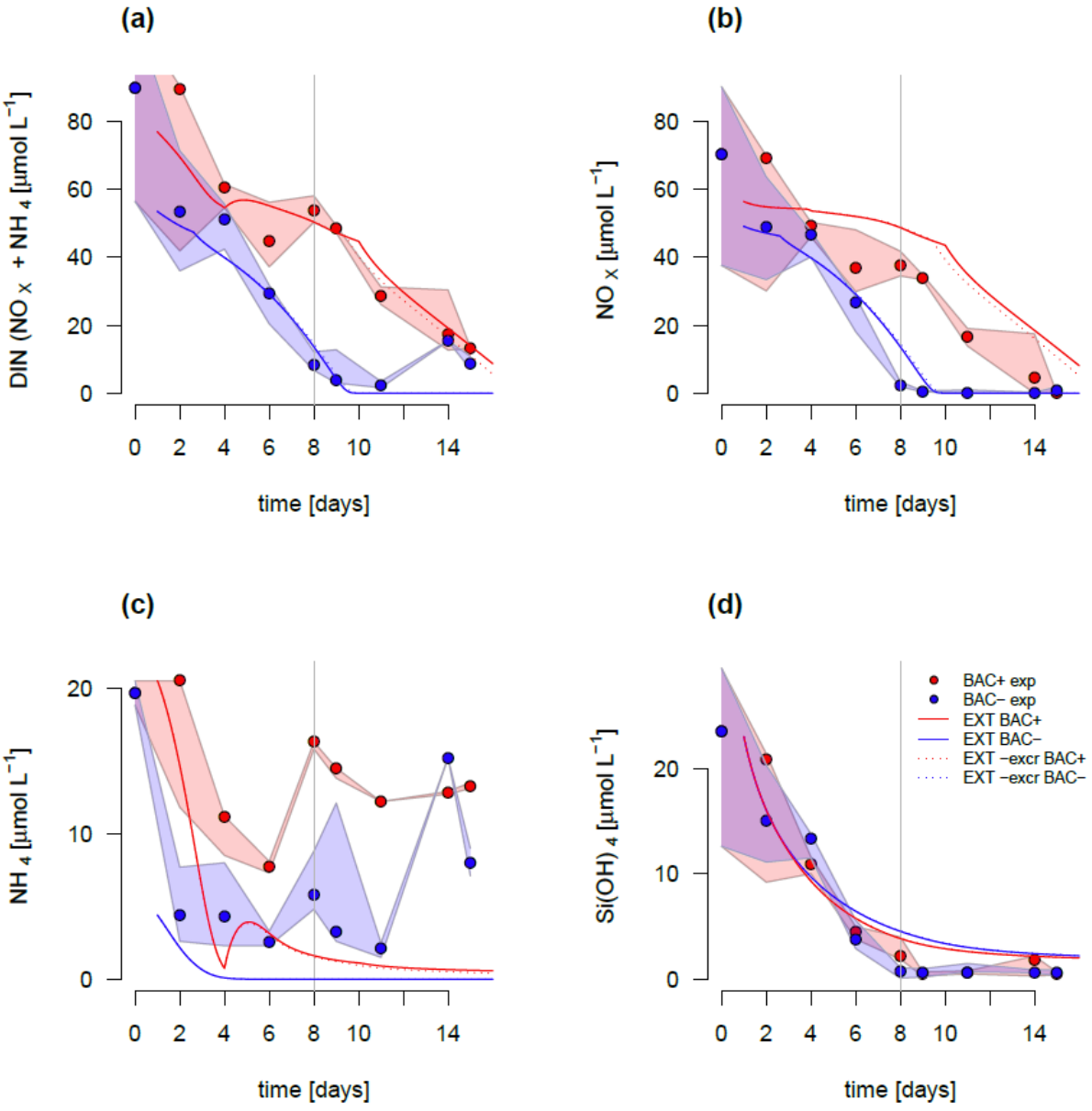

Figure ~~5.~~6. Model fit of the ~~extended~~EXT model to the ~~axenic~~BAC- (blue) and ~~bacteria enriched~~BAC+ (red) experiment. Circles show median values and the ~~coloured~~colored polygons show the ~~total range~~maximum and minimum of measured data~~.~~ (n=3). Solid lines show the model outputs of a) DIN (NOX and NH4), b) NO_X, c) NH_4, and d) $Si(OH)_4$ (All model fits overlap).

Table

Table 1~~:~~. A comparison of major components contributing to the complexity of different models discussed. #param is the number of parameters. In case of ecosystem models (SINMOD, BFM, MEDUSA, LANL, NEMURO, NPZD only the ~~components~~model formulations representing the components of the current model (phytoplankton growth, remineralisation, nutrient dynamics) are considered. For the full ecosystem scale models we give the original reference to the biogeochemical compartment of the ecosystem scale models and examples for more recent versions with updated formulations of other model compartments (e.g. physical drivers). REM designates those models that include Remineralisation (Rem~~), or allow~~) marked with V is present and X is absent. Ratios shows if the stoichiometry in the model considers variable or fixed ratios of intracellular elements (C:N:Si:P:Fe). The Nutrients considered are given under Nutrients. If DIN is considered as both NH_4 and NO_3, N is shown as N^2. MEDUSA has Fe dependent Si:N ratios, which makes them fixed in the Arctic (fixed*).

Model	Reference	#param	Rem	ratios	Nutrients
		Culture scale			
EXT	This study	21[*1]	V	variable	N^2, Si
G98	Geider et al., 1998	10[*2]	X	variable	N
ANIM	Flynn, 1997	30	V	variable	N^2
SHANIM	Flynn and Fasham, 1997	23	X	variable	N^2
Flynn01	Flynn, 2001	54	X	variable	N^2, Si, P, Fe
Flynn18	Flynn et al., 2018	27	X	variable	N
		Ecosystem scale			
BFM	Vichi et al., 2007	54	V	variable	N^2, Si, P, Fe
BFM17	Smith et al., 2020	24	V	variable	N^2, P
REcoM-2	Hauck et al., 2013 Schourup-Kristensen et al. 2018	28	X	variable	N, Si, Fe
MEDUSA	Yool and Popova, 2011 Henson et al., 2018	21	V	fixed*	N, Si, Fe
LANL	Moore et al., 2004	15	V	fixed	N^2, Si, P, Fe
NEMURO	Kishi et al., 2007 Amju et al., 2020	21	V	fixed	N^2, Si
NPZD	Gruber et al., 2006	9	V	fixed	N^2
SINMOD	Wassmann et al., 2006 Alver et al., 2016	12	X	fixed	N^2, Si

Degrees of freedom after constraints by the measured data are [*1]14 and [*2]6

Appendix

Tables

Table A1. State variables of the G98 model and the ~~extended~~EXT model (marked with V if present and X if absent) with units and designation if these state variables had been measured in the experiment.

variable	Description	G98	EXT	Measured	Unit
DIN	Dissolved inorganic nitrogen	V	V	V	mgN m^{-3}
~~pC~~C	Particulate organic carbon	V	V	V	mgC m^{-3}
~~pN~~N	Particulate Nitrogen	V	V	V	mgN m^{-3}
Chl	Chlorophyll a	V	V	V	mgChl m^{-3}
~~dSi~~Si_d	Dissolved Silicate	X	V	X	µmol L^{-1}
~~pSi~~Si_p	Particulate/biogenic Silicon	X	V	V	mgSi m^{-3}
Bact	Bacteria cells	X	V	V	10^r. cells mL^{-1}
DONr	refractory dissolved organic nitrogen	X	V	V	mgN m^{-3}
DONl	labile dissolved organic nitrogen	X	V	X	mgN m^{-3}
NH4	Ammonium	X	V	V	µmol L^{-1}
NO3	Nitrate	X	V	V	µmol L^{-1}
Q	Particulate N : C ratio	X	V	X	gN gC^{-1}
θ^C	Chl to POC ratio	X	V	X	gChl gC^{-1}
θ^N	Chl : phytoplankton nitrogen ratio	X	V	X	gChl gN^{-1}

Table A2. ~~parameters~~Parameters of the original G98 model and the model extension with associated units.

parameter		Unit
	G98	
ζ	cost of biosynthesis	gC gN^{-1}
R^C	The carbon-based maintenance metabolic rate	d^{-1}
θ^N_{max}	Maximum value of Chl:N ratio	gChl gN^{-1}
Q_{min}	Min. N:C ratio	gN gC^{-1}
Q_{max}	Max. N:C ratio	gN gC^{-1}
α^{Chl}	Chl-specific initial C assimilation rate	gC m^2 (gChl µmol photons)$^{-1}$
I	Incident scalar irradiance	µmol photons s^{-1} m^{-2}
n	Shape factor for V^N_{max} max photosynthesis	-
K_{no3}	Half saturation constant for nitrate uptake	µmol L^{-1}
P^C_{ref}	Value of max C specific rate of photosynthesis'	d^{-1}
	Extension	
x_f	Carbon excretion fraction	-
K_{si}	Half saturation constant for Si uptake	µmol L^{-1}
V_{max}	maximum Si uptake rate	mol Si d^{-1} mg C^{-1}
s_{min}	minimum Si required for uptake	µmol L^{-1}
rem	remineralisation rate of excreted don	$bact^{-1}$ d^{-1}
rem_d	remineralisation rate of refractory don	$bact^{-1}$ d^{-1}
μ_{bact}	bacteria growth rate	mio. cells mL^{-1} d^{-1}
$bact_{max}$	Carrying capacity for bacteria	mio. cells mL^{-1}
K_{nh4}	Half saturation constant for ammonium uptake	µmol L^{-1}
$nh4_{thres}$	threshold concentration for ammonium uptake	µmol L^{-1}
Si_{PS}	Fraction of photosynthesis possible after Si lim.	-

Table A3. Parameters of the original G98 model and the ~~extended~~EXT model with initial values used in the model and the lower and upper value constraints used for model fitting, unless the parameter was already defined by the data (measured). The constraints are either based on G98 fits to other diatom species, to present experimental data, or to typical values found in the literature.

parameter	value	lower	upper	constrained by
G98				
ζ	1	1	2	G98
R^C	0.~~02~~01	0.01	0.05	G98
θ^N_{max}	1.7	measured		Data
Q_{min}	0.05	measured		Data
Q_{max}	0.3	measured		Data
α^{Chl}	0.~~1~~1076	0.075	1	G98
I	100	measured		Data
n	3.~~7~~745	1	4	G98
K_{no3}	~~5~~2	~~2~~1	10	G98
P^C_{ref}	0.8	0.5	3.5	G98
Extension				
x_f	0.06	0.01	0.3	Schartau et al., 2017
K_{si}	~~10~~7.6	0.5	~~10~~8	Werner 1978
V_{max}	0.~~33~~1	0.~~32~~05	0.9	~~Werner 1978~~Data
s_{min}	1.82	1.5	6	Werner 1978
rem	~~5.6~~10	~~0.1~~10	~~10~~20	open (rem > rem_d)
rem_d	4.55	0.1	10	open (rem_d < rem)
μ_{bact}	0.04	0.01	0.79	Data
$bact_{max}$	0.015	0.005	0.1	Data
K_{nh4}	6.7~~4~~	~~2~~0.5	~~10~~9.3	~~open~~Eppley 1969
$nh4_{thres}$	~~8~~1.12	0.1	10	open
Si_{PS}	0.2	0	0.5	Werner 1978

Table A4. Output of the sensitivity analysis (senFun of the FME package in R~~).~~) with the value for each parameter and different sensitivity indices obtained after quantifying the effects of small perturbations of the parameters.on the output variables (POC, PON, Chl, DIN). The L1 and L2 norms are normalized sensitivity indices defined as $L1 = \sum \frac{|S_{i,j}|}{n}$ and $L2 = \sqrt{\frac{S_{i,j}^2}{n}}$ with $S_{i,j}$ being the the sensitivity of parameter i for model output j.

par	value	L1	L2	Mean	Min	Max
			G98			
ζ	1.00	0.10	0.19	-0.02	-0.15	0.98
R^C	0.07	0.04	0.05	-0.03	-0.08	0.14
θ^N_{max}	1.70	0.23	0.34	0.14	-1.00	0.58
Q_{min}	0.05	0.06	0.08	-0.04	-0.14	0.22
Q_{max}	0.30	0.34	0.47	-0.24	-1.90	0.28
α^{Chl}	0.08	0.20	0.29	-0.10	-1.10	0.20
I	100	0.20	0.29	-0.10	-1.10	0.20
n	3.40	0.33	0.75	0.03	-0.47	4.07
K_{no3}	2.00	0.01	0.02	0.00	-0.01	0.09
P^C_{ref}	0.80	0.82	1.48	0.16	-7.70	1.04
			EXT			
x_f	0.06	0.19	0.27	-0.10	-0.37	1.10
K_{si}	7.6	0.00	0.00	0.00	0.00	0.00
V_{max}	0.1	0.00	0.00	0.00	0.00	0.00
s_{min}	1.82	0.00	0.00	0.00	0.00	0.00
rem	10	0.00	0.00	0.00	0.00	0.00
rem_d	4.55	0.24	0.31	0.24	0.00	0.65
μ_{bact}	0.04	0.00	0.00	0.00	0.00	0.01
$bact_{max}$	0.015	0.00	0.00	0.00	0.00	0.01
K_{nh4}	6.74	0.08	0.11	-0.03	-0.25	0.46
$nh4_{thres}$	1.19	0.00	0.00	0.00	0.00	0.00
Si_{PS}	0.2	0.08	0.24	-0.02	-1.40	0.31

Table A5. Other parameters calculated and used in the model equations

parameter	Description	Unit
P^{C}_{phot}	C-specific rate of photosynthesis	d^{-1}
P^{C}_{max}	Maximum value of P^{C}_{phot} at temperature T	d^{-1}
R^{Chl}	Chl degradation rate constant	d^{-1}
R^{N}	~~R~~N remineralization rate constant	d^{-1}
V^{C}_{nit}	~~Phytoplankton~~Diatom carbon specific nitrate uptake rate	gN $(gC\ d)^{-1}$
V^{C}_{ref}	Value of V^{C}_{max} at temperature T	gN $(gC\ d)^{-1}$
p_{Chl}	Chl synthesis regulation term	-
μ	specific growth rate of algae	cells d^{-1}

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Table A6. Model equations from G98 (Geider et al., 1998) corrected for typographical errors by Ross and Geider (2009) with extensions.

1)	Carbon synthesis (C originates from unmodelled excess pool of DIC)	$\frac{dC}{dt} = \left(P^C - \zeta V_N^C - R^C\right)C = \mu C$
2)	Chl synthesis	$\frac{dChl}{dt} = \left(\frac{\rho_{Chl} V_N^C}{\Theta^C} - R^{Chl}\right) Chl$
~~4~~3)	Nitrogen uptake	$\frac{dN}{dt} = \left(\frac{V_N^C}{Q} - R^N\right) N$
4)	from Eq. (1) and (2)	$\frac{dQ}{dt} = V_N^C - \mu Q$
5)	from Eq. (1) and (2)	$\frac{d\Theta^C}{dt} = V_N^C \rho_{Chl} - \Theta^C \mu$
~~5~~6)	Photosynthesis	$P^C = P_{max}^C \left[1 - exp\left(-\frac{I}{I_K}\right)\right]$
~~6~~7)	Max. N uptake	$V_N^C = V_{ref}^C \left[\frac{Q_{max} - Q}{Q_{max} - Q_{min}}\right] \frac{DIN}{DIN + K_{no3}}$
8)	with	$\rho^{Chl} = \Theta_{max}^N \left[1 - exp\left(-\frac{I}{I_K}\right)\right]$
~~7~~9)	~~with~~	$V_{ref}^C = P_{ref}^C Q_{max}$
~~8~~10)		$P_{max}^C = P_{ref}^C \frac{Q - Q_{min}}{Q_{max} - Q_{min}}$
~~9~~11)		$I_K = \frac{P_{max}^C}{\alpha^{Chl} \Theta^C}$

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Table A7. Model equations of the EXT model based on G98

1a)	Carbon synthesis *(Reduced C synthesis under Si limitation after Werner 1978)*	$IF\ (Si_d < 2\ s_{min})$ $Si_{PS} = Si_{PS}$ $ELSE$ $Si_{PS} = 1$
1b)		$\frac{dC}{dt} = Si_{PS}\left(P^C - \zeta V_N^C - R^C - xf\right)C = \mu C$
2)	Chl synthesis *(Chl synthesis stops under Si limitation after Werner 1978)*	$IF\ (Si_d < 2\ s_{min})$ $\frac{dChl}{dt} = 0$ $ELSE$ $\frac{dChl}{dt} = \left(\frac{\rho_{Chl} V_N^C}{\Theta^C} - R_{Chl}\right) Chl$
~~10~~3)	from Eq. (1~~) and~~ (& 2)	$\frac{dQ}{dt} = V_N^C - \mu Q$
~~11~~4)	from Eq. (1~~) and~~ (& 2)	$\frac{d\Theta^C}{dt} = V_N^C \rho_{Chl} - \Theta^C \mu$
5)	Nitrogen uptake	$\frac{dN}{dt} = \left(\frac{V_N^C}{Q} - R^N - xf\right) N$
6)	Bacteria biomass production *(Logistic growth)*	$\frac{dBact}{dt} = Bact\ \mu_{Bact}\ (Bact_{max} - Bact)$

7a) Silicate uptake

(Monod kinetics after Spilling et al., 2010)

$$\frac{dSi_d}{dt} = V_S^C = \left(V_{max} Si_d \frac{Si_d - S_{min}}{K_{si}\, S_{min}}\right) C$$

7b)

$$\frac{dSi_p}{dt} = -\frac{dSi_d}{dt}\ 14$$

8) Ammonium uptake and production

(Threshhold after Tezuka 1989, and Gilpin 2004)

$$IF\ (\frac{C}{N} < 10)$$

$$\frac{dNH4}{dt} = \frac{-\left(\frac{V_{NH4}^C}{Q}\right) N + Bact\ xf\ N\ rem + Bact\ DON\ rem_d - \frac{Bact}{16})}{14\ 10^3}$$

$$ELSE$$

$$\frac{dNH4}{dt} = \frac{-\left(\frac{V_{NH4}^C}{Q}\right) N + Bact\ xf\ N\ rem - \frac{Bact}{16})}{14\ 10^3}$$

9) DON uptake and production

$$IF\ (\frac{C}{N} < 10)$$

$$\frac{dDON}{dt} = -\frac{Bact\ xf\ N\ rem + Bact\ DON\ rem_d + xf\ N}{14\ 10^3}$$

$$ELSE$$

$$\frac{dDON}{dt} = -\frac{Bact\ xf\ N\ rem + xf\ N}{14\ 10^3}$$

10) DIN uptake

$$IF\ (NH4 > nh4_{thresh})$$

$$\frac{dDIN}{dt} = \frac{-\left(\frac{V_{NO3}^C}{Q}\right) N - \frac{Bact}{16}}{14\ 10^3}$$

$$ELSE$$

$$\frac{dDIN}{dt} = \frac{-0.2\left(\frac{V_{NO3}^{C}}{Q}\right)N - \frac{Bact}{16}}{14\,10^{3}}$$

11) Photosynthesis

$$P^{C} = P_{max}^{C}\left[1 - exp\left(-\frac{I}{I_K}\right)\right]$$

12a) Max NO3 uptake

$$V_{NO3}^{C} = V_{ref}^{C}\left[\frac{Q_{max} - Q}{Q_{max} - Q_{min}}\right]\frac{NO3}{NO3 + K_{no3}}$$

12b) Max NH4 uptake

(based on SHANIM Eq4 by Flynn and Fasham, 1997)

$$V_{NH4}^{C} = (0.01\,Q)\,0.0021\,\frac{NH4}{NH4 + K_{nh4}}$$

13) Max N uptake

(Based on Flynn and Fasham, 1997 and Flynn, 1999 showing no total inhibition in cold water)

$$IF\ (NH4 > nh4_{thresh})$$

$$V_N^C = V_{NH4}^C + 0.2\,V_{NO3}^C$$

$$ELSE$$

$$V_N^C = V_{NH4}^C + V_{NO3}^C$$

14) with

$$\rho^{Chl} = \Theta_{max}^{N}\left[1 - exp\left(-\frac{I}{I_K}\right)\right]$$

15)

$$V_{ref}^{C} = P_{ref}^{C} Q_{max}$$

16)

$$P_{max}^{C} = P_{ref}^{C}\frac{Q - Q_{min}}{Q_{max} - Q_{min}}$$

17)

$$I_K = \frac{P_{max}^{C}}{\alpha^{Chl}\Theta^{C}}$$

Table A8. Output of the collinearity or parameter identifiability analysis using the collin function of the FME R package (Soetart et al., 2010b). A subset of any combinations of two parameter with a collinearity above 20, indicating non-identifiable parameter combinations is given (Brun et al., 2001).

ζ	R^C	θ^N_{max}	Q_{min}	Q_{max}	α^{Chl}	I	n	K_{no3}	P^C_{ref}	collinearity
1	0	1	0	0	0	0	0	0	0	31
1	0	0	0	1	0	0	0	0	0	59
1	0	0	0	0	1	0	0	0	0	42
1	0	0	0	0	0	1	0	0	0	42
1	0	0	0	0	0	0	1	0	0	74
0	1	0	0	0	0	0	0	0	1	22
0	0	1	0	1	0	0	0	0	0	32
0	0	1	0	0	1	0	0	0	0	26
0	0	1	0	0	0	1	0	0	0	26
0	0	1	0	0	0	0	1	0	0	41
0	0	0	0	1	1	0	0	0	0	49
0	0	0	0	1	0	1	0	0	0	49
0	0	0	0	1	0	0	1	0	0	81
0	0	0	0	0	1	1	0	0	0	1756319
0	0	0	0	0	1	0	1	0	0	60
0	0	0	0	0	0	1	1	0	0	60

Figure

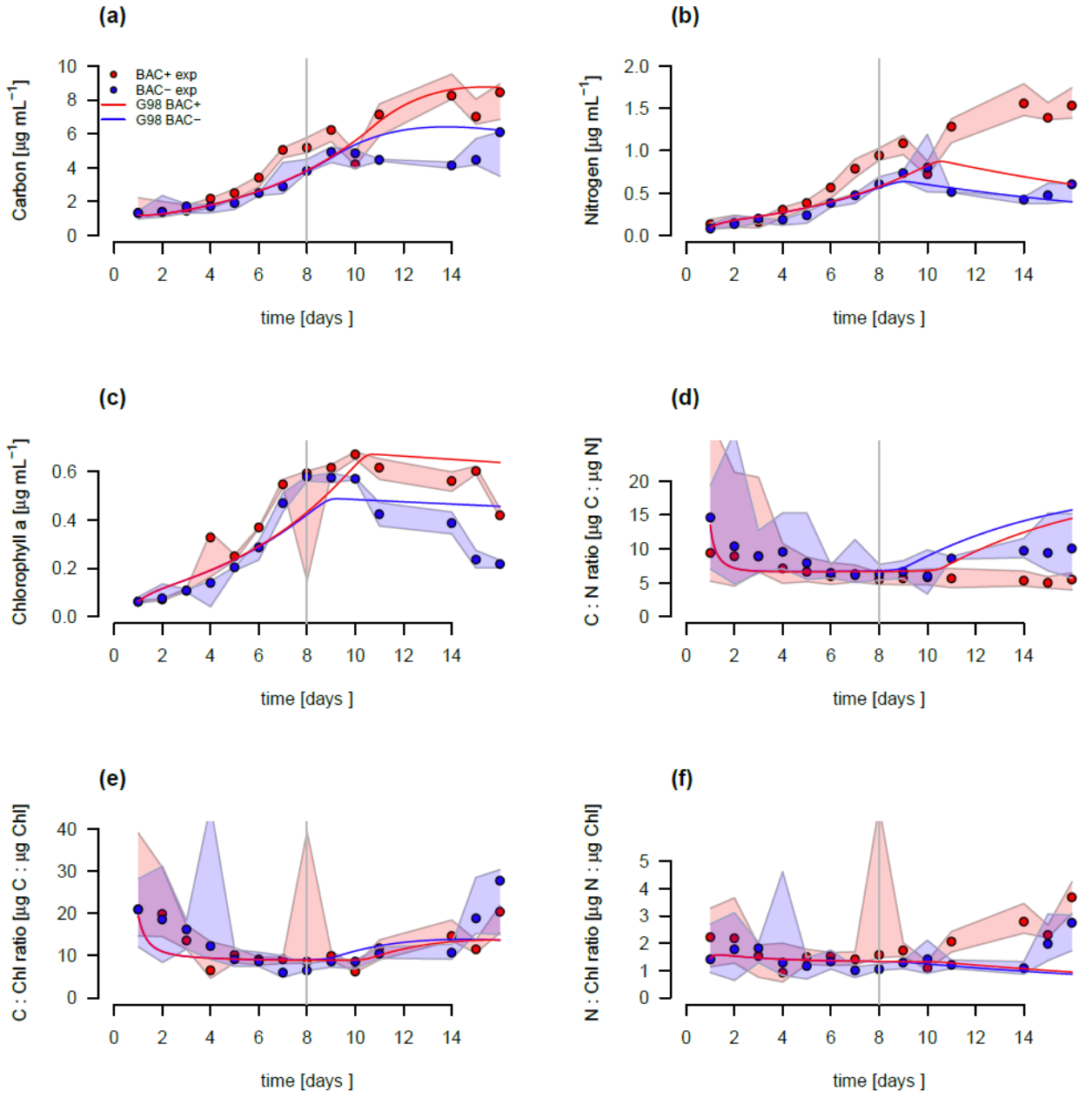

Figure B1: Model fit of the G98 model to the ~~axenic~~BAC- (blue) and ~~bacteria enriched~~BAC+ (red) experiment. Circles show median values and the ~~coloured~~colored polygons show the ~~total range~~minimum and maximum of the measured data~~,~~ (n=3). Solid lines show the model outputs of a) POC, b) PON, c) Chl~~,~~ (including outlier at day 8 in BAC+), d) C:N, e) C:Chl, and f) N:Chl.

Equations

Equation C1. F-ratio estimation in the cultivation experiments with the average PON concentrations at day 13 to 15 ($PON^{d13\text{-}15}$) for the BAC- and BAC+ treatments.

$$f - ratio = \frac{PON_{BAC-}^{d13-15}}{PON_{BAC-}^{d13-15} + PON_{BAC+}^{d13-15}}$$

Equation C2. normalized RMSE with i being the different variables (POC, PON, Chl, DIN), and j the different values of each state variable. Predicted values are given as P and observed values as O.

$$RMSE = \sqrt{\sum_{\substack{i=1 \\ j=1}}^{n,p} \frac{(P_{i,j} - O_{i,j})^2}{Var(O_i)}}$$