Response to referee O.P. Savchuk

We again thank the referee for his valuable comments on the manuscript. In addition to our previous response we will here provide som further comments on the changes that have been made in accordance with the referee feedback.

- 1.1 We have removed implications of causality where possible. We do however retain the word "causes" in the title with reference to our earlier reply.
- 1.2 We have removed the observational part. We have instead chosen to present the time-series and the wavelet spectrum of the simulated phytoplankton biomass together with the month of maximum chlorophyll maxima in Sect. 3.1. We have tried to make it clear that we are using only simulated variables.
- 1.3 We have changed to "phytoplankton biomass" as well as added a comment on the constant C:Chl ratio in Sect. 2.2.
- 1.4 We have removed discussions around the seasonal time-scale where possible. We have kept comments on the seasonal scale for the clear regime shift shown in current Fig. 10.
- 1.5 Again, we have removed the observational parts from the manuscript.
- 1.6 We have tried to improve the structuring and motivation for the manuscript mainly in the introduction and throughout Sect. 3. Much of the justification certainly boils down to the use of a relatively new tool. However, as a similar analysis has not previously been done for simulated biogeochemical variables we feel that an illustration of its uses is valuable.
- 2.1 We have changed the title to: Causes of simulated long-term changes in phytoplankton biomass in the Baltic Proper: A wavelet analysis.
- 2.2 We have removed "internal loads" where possible.
- 2.3 As stated in 1.6, we have tried to rework the introduction and section 3 in accordance with the referee comments.
- 2.4 This has been corrected.
- 2.4.1 We have removed unimportant equations from section 2. We have also removed the faulty comment on phosphate and salinity.
- 2.4.2 We have clarified that it is only around the study area.
- 2.5.1 The comparison with observations have been removed
- 2.5.2 We have removed discussion on the seasonal scale for the riverine input. We have further rewritten the section so that it is clear that we do not imply causality. We have also removed previous Figs 6 and 7.



Figure 1: Nitrogen or phosphate limitation as calculated with N/P ratios.

2.5.3 We have added a comment on that Nlim can become larger than one (but not NUTLIM). However, as NUTLIM is what directly affects the phytoplankton growth in the model we have kept this formulation. We have added a discussion on that N/P ratios gives a different result more inline with observations in Sect. 3.2 (see Fig. 1).

We have also added a note in the figure caption that simultaneous N and P limitation is not possible.

- 2.5.4 For some quantities this might be helpful but we prefer the mixed layer concept, as is much more straight forward when it comes to the physical quantities. The sharp pycnocline inhibits vertical transfer, and is therefore a more natural choice for studying variations in N and P concentrations.
- 2.5.5 We have tried to rework the section (now 3.2) so that the purpose of the section is more clear. We have kept the figure showing our model results for anoxic volume and deep water nutrients (now Fig. 5) since we believe it to be necessary for the discussion.
 - 3. We have addressed the minor comments.

Response to Referee #3

We again thank the referee for the valuable comments. Referee comments in italics.

We have shortened section 2.2 and removed unnecessary equations.

• Already in the abstract combination of words mixed layer parameter concentrations appears as solid term. However I did not find in the text how it was defined. Is it mean value of horizontal mean parameter in horizontal mean mixed layer? or it is integrated characteristic?

We have clarified that the parameters are horizontally and depth *averaged* and not integrated.

• Salinity in the Baltic Sea and in the Baltic Proper have strong lateral gradient. However, mixed layer depth (MLD) was defined as constant density difference. Could it be that with decrease of salinity MLD will increase? Could it be that seasonal variability in surface effects MLD and at the end all results? The part with mixed layer definition should be extended and some how emphasized. Maybe it makes sense to include it as additional subsection.

Changes in salinity will effect the mixed layer depth due to its effect on density, but this is captured in the definition of mixed layer as long as the equation of state used to calulate the density difference depends on salinity. The definition also captures the seasonal changes in mixed layer depth when monthly mean profiles of salinity and temperature are used in the calculation. The mixed layer definition is also standard and frequently used both in models and on observational data.

• The basin integrated approach was used here (line 61). Would be good to see in the text why this is acceptable (preferably in more than one sentence, line 6

We have added a comment in section 2.1.

• While SCOBI model is 1D model (line 67), I would suggest to show results of wavelet analysis for idealized 1D cases. So it could be seen how certain changes are reflected in final results of wavelet analysis. For my opinion such sensitivity test could enhance conclusions. Otherwise, section 2.4 should be extended with some aspects of wavelet coherence.

We have improved section 2.4 to provide a better description of the wavelet transform and wavelet coherence.

• Analysis focuses mainly on river loads and its changes. Other nutrient sources like atmospheric deposition, exchange with other Baltic Sea regions and there possible effect should be mention somehow

We have tried during the revision work to include the atmospheric deposistion. Sadly, we only had yearly averaged values of the deposition to work with and that is not good enough for the wavelet analysis. The horizontal transports suffers from a similar problem. Here we have the velocity fields and concentration fields but not their products, and we thus do not really know the transports. In future work we plan to close these nutrient budgets using online calculations, but in this current effort we have settled to look at correlations with some of the most important forcings.

• It could be considered to include wavelet analysis in to the title to my opinion application of this method is among the most interesting aspects of this manuscript

We have changed the title in accordance with the review comment.

• Line 75: eq. 1. NFIX is nitrogen fixation term, in all phytoplankton groups it looks strange. Is it a misprint?

We found the equation to be unnecessary and have therefore removed it.

- Line 78: SINKIphy / SINKOphy is it sinking of phytoplankton? We have removed the equation.
- Lines 177 181: Paragraph is confusing. It starts with sentence about open boundary, but last two sentences are probably about river loads. Please specify in more details: what these assumption were applied to We have rewritten.

Causes of simulated , longterm long-term changes in chlorophyll concentrations phytoplankton biomass in the Baltic SeaProper: A wavelet analysis

Jenny Hieronymus¹, Kari Eilola¹, Magnus Hieronymus¹, H. E. Markus Meier^{2,1}, and Sofia Saraiva¹

¹Research and Development Department, Swedish Meteorological and Hydrological Institute, Norrköping, Sweden ²Department of Physical Oceanography and Instrumentation, Leibniz Institute for Baltic Sea Research Warnemünde, Rostock, Germany.

Correspondence to: Jenny Hieronymus (jenny.hieronymus@gmail.com)

Abstract. The co-variation of key variables with modelled phytoplankton concentrations simulated phytoplankton biomass in 1 2 the Baltic proper has been examined using wavelet analysis and results of a long-term simulation for 1850-2008 with a high-3 resolution, coupled physical-biogeochemical circulation model for the Baltic Sea. By focusing on interannual-inter-annual variations it is possible to track effects acting on decadal time scales such as temperature increase due to climate change as 4 well as changes in nutrient input. The results indicate the largest inter-annual coherence of phytoplankton biomass with the 5 limiting nutrient. However, after 1950 the coherence is reduced due to high mixed layer nutrient concentrations diminishing 6 7 the effect of smaller long-term variations. Furthermore, the inter-annual coherence of mixed layer nitrate with riverine input of nitrate is much larger than the coherence between mixed layer phosphate and phosphate loads. This indicates a greater 8 9 relative importance of internal loads i.e. mixing of phosphate from deeper layers. In addition, shifts in nutrient patterns give rise to changes in phytoplankton nutrient limitation. The modelled pattern shifts from purely phosphate limited to a seasonally 10 11 varying regime. The results further indicate some effect of inter-annual temperature increase on cyanobacteria and flagellates. Changes in mixed layer depth affect mainly diatoms due to a high sinking velocity while inter-annual coherence between 12 13 irradiance and phytoplankton is not observed. found.

14 1 Introduction

The Baltic Sea is a semi-enclosed brackish water body separated from the North Sea and Kattegat through the Danish Straits. It stretches from about 54° to 66° N and the limited water exchange with the ocean in the south gives rise to a large meridional salinity gradient. The circulation is estuarine with a salty <u>deepwater deep-water</u> inflow from the ocean and a fresher surface outflow. The Baltic Sea comprises a number of sub-basins connected by sills further restricting the circulation.

The limited water exchange and the long residence time of water have consequences for the functioning of the biology and the biogeochemistry. The Baltic Sea is naturally prone to eutrophication and organic matter degradation keeps the deep water oxygen concentrations generally low in between deep water renewal events. In turn, this leads to complex nutrient cycling with different processes acting in oxygenized vs low oxygen environments. The Baltic Sea has experienced <u>extensive</u> anthropogenic pressure over the last century. After 1950an, intensive use of agricultural fertilizer greatly enhanced the nutrient loads. <u>Due to great improvements in sewage treatment the loads decreased</u>

25 again after 1980 (Gustafsson et al., 2012).

26 The intensification in nutrient loads This led to an expansion of hypoxic bottoms (Carstensen et al., 2014). This has had effects on, in turn affecting the cycling of nutrients through the system. Anoxic sediments have lower phosphorus retention 27 28 capacity resulting in increased deep water phosphate concentrations. Thereby, the flux of phosphate to the surface intensifies even though the external loads have decreased after 1980 in response to improved sewage treatment. Furthermore, as the 29 anoxic area increases, the boundary between anoxic and oxic sediments area of interface between oxic and anoxic zones 30 31 where denitrification occurs also increases. This results in a loss of nitrogen. Vahtera et al. (2007) described these processes as generating a "vicious circle" where decreased DIN concentrations together with increased phosphate enhance the relative 32 33 importance of nitrogen fixation by cyanobacteria.

The importance of this coupling between oxygen and nutrients have been further examined in models. Gustafsson et al. (2012) confirmed, using the model BALTSEM, that internal nutrient recycling has increased due to reduced phosphate retention capacity, implicating a self sustained eutrophication where enhanced internal loads outweigh external load reductions.

37 In addition to the biogeochemical shifts in the Baltic Sea environment during the 20th century, sea surface temperatures have

38 increased (Siegel et al., 2006). This has an effect on the growth rate of phytoplankton as well as the speed of other biological

39 processesSatellite monitoring has made it possible to observe changes in several physical and ecological surface variables

40 during the past three decades. Significant changes in seasonality have been observed, such as earlier start of phytoplankton

41 growth season and timing of chlorophyll maxima (Kahru et al., 2016).

42 From satellite data, Kahru et al. (2016) detected a prolonged productive season as well as a chlorophyll maxima shifted

43 towards the maximum cyanobacteria concentration in July. The effect of temperature on the growth rate and stratification is

44 likely to have positively affected the strength of cyanobacteria blooms as well as the length of the growth season.

45 Schimanke and Meier (2016) analyzed multidecadal variations in Baltic Sea salinity and the coherence with different physical

46 drivers. They used the wavelet transform to identify periodicities and wavelet conherency to analyse the driving mechanisms Although

47 the satellite record is already substantial and growing, interannual shifts and variations over the past century can not be

48 investigated in this way. Furthermore, the satellite record is restricted to a few surface variables. Shifts in nutrient composition

49 and deep water variables remain difficult to evaluate using observations. Even though the Baltic Sea has a dense observational

50 record from ships, stations and satellites, the longest nutrient records comprise station data from the early 70s (HELCOM, 2012).

51 For multidecadal periods of gap free data the use of a model is required.

52 In this paper we construct a thorough analysis of the co-variation of phytoplankton concentration biomass with key variables

53 that have been affected by anthropogenic change over the 20th century. Using the biogeochemical model SCOBI (Eilola et al.,

54 2009; Almroth-Rosell et al., 2011) coupled to the 3d circulation model RCO we scrutinize the effect of nutrient loads, nutrient

55 concentration, temperature, irradiance and mixed layer depth on the modelled phytoplankton community.

56 The effect of anoxia on the nutrient limitation and on the primary production is complex. In addition to decreased phosphorus

57 retention capacity and denitrification, nitrification ceases in anoxic environments ultimately resulting in increased ammonium

58 concentrations (Conley et al., 2009). To elucidate the effect on the primary production, we calculate the degree of nutrient

59 limitation and its correlation with phytoplankton. gap-free dataset provided by the model lets us decompose the variables in

60 time-frequency space using the wavelet transform. Two variables may than be compared using wavelet coherence (eg. Torrence and Compo

61

62 We have chosen to use a model run spanning 1850-2009. Thereby, we capture conditions relatively unaffected by anthro-

63 pogenic forcing as well as current conditions of eutrophication and climate change. Furthermore, we limit our investigation to

64 the Baltic Proper so as to capture relatively homogenous conditions with regards to the functioning of the biology. Our main

65 focus lies in inter-annual variations although some seasonal shifts will be investigated. biology.

66 Schimanke and Meier (2016) analyzed multidecadal variations in Baltic Sea salinity and the coherence with different physical

67 drivers. They used the wavelet transform to identify periodicities and wavelet coherency to analyse the driving mechanisms.

68 2 Methods

69 2.1 Study area

The Baltic Sea contains several different sub-basins with different characteristics in salinity and nutrient loads. We have here chosen to focus on the Baltic Proper. To obtain homogenous conditions we focus on the open ocean away from coasts. Areas where the depth is less than 20m are therefore removed. The study area is displayed in Fig. 1.

73 We have chosen to use a basin integrated averaged approach. All variables have thus been horizontally integrated averaged

74 over the study area. This way we aim to gain an remove local variability and hope to gain a better understanding of the overall

75 functioning of the system.

76 2.2 Model

We have used a run with the model RCO-SCOBI spanning 1850-2009. RCO (Rossby Centre Ocean model) is a threedimensional regional ocean circulation model(Meier et al., 2003). It is a z-coordinate model with a free surface and an open boundary in the northern Kattegat. The version used here has a horizontal resolution of 2nm with 83 depth levels at 3m intervals.

80 The biogeochemical interactions are solved by the Swedish Coastal and Ocean Biogeochemical model (SCOBI) (Eilola et al.,
81 2009; Almroth-Rosell et al., 2011)is a one dimensional biogeochemical model that. The model solves for three different water

82 column and benthic nutrients (phosphate, nitrate and ammonia) as well as plankton functional types representing diatoms,

83 flagellates and others (will be referred to as flagellates from here on) and cyanobacteria. Furthermore, the model contains

84 nitrogen and phosphorus in one active homogenous benthic layer.

The model equations can be found in Eilola et al. (2009). Since we are exploring the effect of different variables on the growth of phytoplankton we will, for clarity, repeat some of them here.

87 The time rate of change of the concentration of phytoplankton chlorophyll in units of mg Chl m³ day⁻¹ is described by-

- 88 $S_{PHY} \equiv GROWTH_{PHY} + NFIX + SINKI_{PHY}$
- $89 \qquad -SINKO_{PHY} MORT_{PHY} GRAZE_{PHY},$
- 90 where subscript PHY stands for phytoplankton 1 (diatoms), 2 (flagellates) or 3 (cyanobacteria). GROWTH_{PHY} describes the
- 91 growth of phytoplankton, NFIX the production by nitrogen fixation, SINKIPHY/SINKOPHY the flux of phytoplankton into /out
- 92 of the current layer, MORT_{PHY} the mortality and GRAZE_{PHY} grazing by zooplankton phytoplankton biomass is described in
- 93 terms of chlorophyll and with a constant C:Chl ratio. The model thus does not take into account seasonal changes in C:Chl as
- 94 was found by Jakobsen and Markager (2016).
- 95 The net growth of phytoplankton is described by the following expression,
- $96 \quad GROWTH_{PHY} = ANOX \cdot LTLIM \cdot NUTLIM_{PHY} \cdot GMAX_{PHY} \cdot PHY,$ (1)

where subscript PHY indicates the plankton funktional type (diatoms, flagellates or cyanobacteria). ANOX is a logarithmic
expression that approaches zero as the oxygen concentration becomes small. ANOX also contains a switch that sets it equal to
zero when the oxygen concentration is zero so that no phytoplankton growth can occur in anoxic environments.

LTLIM expresses the phytoplankton light limitation and NUTLIM describes the nutrient limitation. Nutrient limitation follows Michaelis-Menten kinetics where constant Redfield ratios are assumed in nutrient uptake. NUTLIM and LTLIM-is further described in Sects. 2.2.1 and 2.2.2. GMAX is temperature dependent and describes the maximum phytoplankton growth rate.

104 The difference between diatoms and flagellates are present in halfsaturation constants, maximum growth rate, temperature 105 dependence and sinking rate. Flagellates are more sensitive to a change in temperature than diatoms. Furthermore, the sinking 106 rate of diatoms is five times larger than that for flagellates.

The difference between cyanobacteria and the other phytoplankton species is more pronounced. Cyanobacteria can grow
 either according to Eq. (1) or using nitrogen fixationaccording to-

109 $\underline{\text{NFIX}} = \underline{\text{ANOX} \cdot \text{NF} \cdot \text{A3}}$

110 where NF is the . The rate of nitrogen fixation as a function of the phosphate concentration and temperature, and A3 is the 111 concentration of cyanobacteria. Both NFIX and GROWTH of cyanobacteria is zero if the salinity is above 10. Furthermore, 112 cyanobacteria is the most temperature sensitive of the phytoplankton groups and no sinking velocity is assumed.

Other processes important for our results involves chemical reactions occurring in the water column or in the sediment.
Denitrification occurs in both the water column and the benthic layer and constitutes a sink for nitrate in case of anoxia.
Nitrification transforms ammonium into nitrate as long as oxygen is present. Phosphorus is adsorbed to the sediment and the

116 benthic release capacity of phosphate is a function of the oxygen concentration where more oxygen implies less release. The

117 phosphorus release capacity is also dependent on salinity where higher salinity means more less phosphate is retained in the

118 benthic layer.

119 2.2.1 Nutrient limitation

120 Estimating nutrient limitation in nature is difficult. Usually this is done, either by comparing nutrient ratios to Redfield in eg.121 the surface water or external supply or by some nutrient enrichment experiment (Granéli et al., 1990).

The idea of nutrient limitation as often used is based on that the primary production is directly limited by the nutrient concentration in the ambient water and that the internal nutrient ratios in the phytoplankton are constant, i.e. in accordance with a Redfield-Monod model (Redfield, 1958). However, cell-quota type models (Droop, 1973) are being increasingly implemented and the use of constant internal nutrient ratios are becoming more and more questioned (Flynn, 2010).

Furthermore, N vs P limitation is a long standing debate. Tyrrell (1999) uses a box-modelling approach to show that in steady state, nitrogen becomes slightly deficient while it is the external input and removal of phosphate that ultimately controls the production.

Here, nutrient limitation is traditionally expressed assuming constand Redfield ratios and phytoplankton growth is limitedby either nitrogen or phosphate. The degree of nutrient limitation is described by:

$$131 \quad \text{NUTLIM}_{\text{PHY}} = \min(\text{NLIM}_{\text{PHY}}, \text{PLIM}_{\text{PHY}}) \tag{2}$$

where $NLIM_{PHY}$ and $PLIM_{PHY}$ are the nitrogen and phosphate limitation respectively. In addition, $NLIM_{PHY}$ contains the sum of the nitrate and ammonium limitation, i.e.

$$134 \text{ NLIM}_{PHY} = \text{NO}_3 \text{LIM}_{PHY} + \text{NH}_4 \text{LIM}_{PHY}, \tag{3}$$

135 where

136 NO₃LIM =
$$\frac{\text{NO3}}{\text{KNO3}_{\text{PHY}} + \text{NO3}} \cdot \exp(-\phi_{\text{PHY}} \cdot \text{NH}_4),$$
 (4)

$$137 \text{ NH}_4 \text{LIM} = \frac{\text{NH4}}{\text{KNH4}_{\text{PHY}} + \text{NH4}},$$
(5)

where NO3 and NH4 are the concentrations of nitrate and ammonium and $KNO3_{PHY}$ and $KNH4_{PHY}$ are the halfsaturation constants for nitrate and ammonium respectively. The exponent in (4) represents preferential ammonium accounts for inhibition of nitrate uptake (eg. Dortch (1990); Parker (1993)).

141 PLIM_{PHY} is modelled as,

142
$$PO_4LIM = \frac{PO4}{KPO4_{PHY} + PO4}$$
 (6)

143 Nutrient limitation is thus described by a number between 0 and 1 where 1 is no limitation. Note that NLIM in Eq. (3) may

144 obtain values larger than 1. However, as NUTLIM is calculated as the minimum of NLIM and PLIM, NLIM larger than one

145 will always mean P limitation.

- 146 The constants KNO3_{PHY}, KNH4_{PHY} and KPO4_{PHY} are the half saturation constants and differs between the different
- 147 phytoplankton groups. The constant ϕ_{PHY} in Eq. (4) determines the strength of ammonium inhibition of nitrate uptake. The 148 values of the constants for each phytoplankton type are given below.
- 149 $KNO3_{PHY} \equiv 0.5/0.25/0.25$ 150 $KNH4_{PHY} \equiv 0.5/0.25/0.25$
- 151 $KPO4_{PHY} \equiv 0.1/0.05/0.05$
- 152 $\phi_{PHY} \equiv 1.5/1.5/1.5$

153 Note that the half-saturation constants for flagellates and cyanobacteria are equal which means that in absence of nitrogen
154 fixation, the nutrient limitation for the nitrogen fixing species is equal to that of flagellates.

155 In addition to the above given nutrient limitation of phytoplankton growth there exists a similar nutrient dependency on

156 nitrogen fixation. In the model this dependency reads-

157
$$\underline{\text{NUTLIM}_{\text{NF}}} \equiv \frac{\text{aNFC}}{\text{aNFC} + \left(\frac{\text{NO}_3 + \text{NH}_4}{\text{PO}_4 \text{cNFC}}\right)} \cdot \frac{\text{PO}_4}{\alpha \text{NF} \cdot \beta \text{NF} + \text{PO}_4},$$

158 where aNFC, bNCF, cNFC and dNFC are constants used for calculating the nitrogen fixation capacity which in turn is a 159 function of the ratio of inorganic nitrogen to phosphate. α NF and β NF are constants determining the half-saturation for nitrogen 160 fixation. Again, NUTLIM_{NF} approaches one if the nitrate and ammonium concentrations are zero and for large concentrations 161 of phosphate.

162 2.2.2 Effect of physical parameters

163 Changes in cloud-cover affect the incoming solar radiation and thereby the phytoplankton growth. The effect of light shows up164 in the LTLIM term of Eq. (1).

$$165 \quad \underline{\text{LTLIM}} \equiv \frac{\text{fracI}_{PAR} I_{opt} \cdot \text{EXP} \left(1 - \frac{I_{PAR}}{I_{opt}}\right),}{166 \quad \underline{I}_{PAR}(z)} \equiv \frac{\alpha_{PAR} I_0 \cdot \text{EXP} \left(-\text{Kd} \cdot z\right)}{167 \quad \underline{I}_{opt}} \equiv \frac{\max(I_{opt,\min}, \alpha_{opt} I_0)}{168 \quad \underline{\text{Kd}}} \equiv \frac{\text{Kd}_w + \text{Kd}_{PHY} + \text{Kd}_Y + \text{Kd}_D}{169 \quad \underline{\text{Kd}}_{PHY}} \equiv \frac{\alpha_{Kd}(A1 + A2 + A3)}{169 \quad \underline{\text{Kd}}_{PHY}} = \frac{\alpha_{Kd}(A1 + A2 + A3)}{169 \quad \underline{\text{Kd}}_{PHY}}$$

170 where I_{PAR} is the photosynthetic available radiation and I_{opt} is the optimum irradiance for phytoplankton growth. $I_{opt,min}$ is 171 a constant minimum optimum irradiance, I_0 is the surface irradiance and Kd is the vertical attenuation. Kd_w is the background 172 attenuation, Kd_{PHY} is the light attenuation due to the concentration of phytoplankton, Kd_Y the attenuation due to humic 173 substances (calibrated) and Kd_D the attenuation due to detritus. α_{Kd} is a constant vertical attenuation per unit chlorophyll. 174 A1/2/3 is the concentration of the respective phytoplankton type.

The mixed layer depth has been defined as the depth where a density difference of 0.125 kg m^{-3} from the surface is reached in accordance with what was previously done by e.g. Eilola et al. (2013). The density was calculated from modelled temperature and salinity using the matlab routines by algorithms from Jackett et al. (2006).

178 2.3 Forcing

The study use reconstructed (1850-2008) atmospheric, hydrological and nutrient load forcing and daily sea levels at the lateral boundary as described by Gustafsson et al. (2012) and Meier et al. (2012). Monthly mean river flows were merged from reconstructions done by Hansson et al. (2011) and by Meier and Kauker (2003) and hydrological model data by Graham (1999), respectively. For further details about the physical model setup used in the present study the reader is referred to Meier et al. (2016) and references therein.

The nutrient loads from rivers and point sources were (1970-2006) compiled from the Baltic Environmental and HELCOM databases (Savchuk et al., 2012). Estimates of pre-industrial loads for 1900 were based upon Savchuk et al. (2008). The nutrient loads were linearly interpolated between selected reference years in the period between 1900 and 1970. Similarly, atmospheric loads were estimated (Ruoho-Airola et al., 2012). Nutrient loads contain both organic and inorganic phosphorus and nitrogen, respectively. For riverine organic phosphorus and nitrogen loads bioavailable fractions of 100 and 30% are assumed, respectively.

Figure 2 shows the loads of Dissolved Inorganic Phosphorus (DIP, top) and Dissolved Inorganic Nitrogen (DIN, bottom) to the Baltic Proper as used in the model defined in Fig. 1. The loads are shown together with the corresponding simulated mixed layer concentration. The loads are calculated from the runoff and annual mean nutrient concentrations (Eilola et al., 2011). Thus the seasonal cycle in river loads is determined by the runoff. After a spin-up simulation for 1850-1902 utilizing the reconstructed forcing as described above, the calculated physical and biogeochemical variables at the end of the spin-up simulation were used as initial condition for 1850. The open boundary conditions in the northern Kattegat were based on climatological (1980-2000) seasonal mean nutrient concentrations (Eilola et al., 2009). The bioavailable fraction of organic phosphorus was assumed to be 100% in accordance with the phosphorus supply from land runoff. Similar to Gustafsson et al. (2012) a linear decrease of nutrient concentrations back in time was added assuming that climatological concentrations in 1900 amounted to 85% of present day concentrations (Savchuk et al., 2008). The bioavailable fraction of organic phosphorus at the boundary was assumed to be 100% in accordance with the organic phosphorus supply from land runoff. Organic nitrogen was implicitly added because of the Redfield ratio of model detritus (Eilola et al., 2009).

- 203 **2.4** The wavelet transform and wavelet coherence
- Several references explain the wavelet transform and its application in depth (e.g. Lau and Weng (1995), Torrence and Compo (1998),
 Carey et al. (2016), Grinsted et al. (2004) and we will here provide a brief introduction.

206 The continuous wavelet transform provides a method to decompose a signal into time-frequency space. In contrast to the 207 Fourier transform, the wavelet decomposition thus provides time localization and the means to see how periodicities change with time. Wavelet coherence further expands the usefulness of the approach by allowing for calculating the time resolved 208 209 coherence between two that it is similar to the windowed Fourier transform where the signal is decomposed within a fixed time-frequency window which is then slided along the time-series. For all wavelet calculations we use the Matlab wavelet 210 211 package of described in Grinsted et al. (2004), which is freely available at http://www.glaciology.net/wavelet-coherence. However, the fixed width of the window leads to an underestimation of low frequencies. In comparison, the wavelet transform 212 utilizes wavelets with a variable time-frequency window. Wavelets can have many different shapes and the choice is not 213

- arbitrary. We have chosen the commonly used Morlet wavelet providing good time and frequency localization (Grinsted et al., 2004).
- 215

In time-series with clear periodic patterns that is affected by environmental variables such as population dynamics and 216 217 ecology the benefits with this approach are significant (Cazelles et al., 2008). Several studys have implemented wavelet analysis to plankton dynamics. Winder and Cloern (2010) applied the technique to time-series of chlorophyll-a from different localities 218 and discussed the annual and seasonal periodicities In recent years, several references have highlighted the usefulness of wavelet 219 220 analyses in plankton research (Winder and Cloern, 2010; Carey et al., 2016). The focus have been the increased availability of long observational data sets making it possible to use the wavelet transform for investigation of changes in seasonality. 221 222 Carey et al. (2016) discussed how the wavelet transform can be used to track interannual changes in phytoplankton biomass 223 and applied it to a 16-year time series of phytoplankton in Lake Mendota, USA. In doing this they were able to identify 224 periods when the annual periodicity was less pronounced. They discuss the benefit of this technique in scrutinizing changes to the seasonal succession due to changes in external drivers. Winder and Cloern (2010) applied the technique to time-series of 225 226 chlorophyll-a from marine and freshwater localities and discussed the annual and seasonal periodicities. Wavelet coherence further expands the usefulness of the wavelet approach by allowing for calculating the time resolved 227

228 coherence between two time-series (Grinsted et al., 2004; Cazelles et al., 2008). In this way, it is possible to identify transient

229 periods of correlation over different periodicities. The result is given as coherency as a function of time and period as well as a

230 phase lag between the two time-series.

- The problem with the wavelet transform is that it requires a dataset without gaps. The time-series also needs to be sufficiently long compared to the investigated periods. This makes it difficult to use the method to scrutinize the <u>effect coherence</u> of processes acting on longer time-scales, such as climate change, since long enough observational datasets are scarce. Hence, for our purpose only a model based approach is feasible.
- 235 Here we use wavelet coherence to Schimanke and Meier (2016) used wavelet coherency on a multi-centennial model run to

236 evaluate the correlation of different forcing variables with the Baltic Sea salinity. We will here scrutinize the coherence between

- 237 the three different phytoplankton groups (diatoms, flagellates, and cyanobacteria) and nutrients, temperature, irradiance and
- 238 mixed layer depthmodelled phytoplankton biomass and a few key modelled and forcing variables.
- For all wavelet calculations we use the Matlab wavelet package of described in Grinsted et al. (2004), which is freely
 available at http://www.glaciology.net/wavelet-coherence.

241 2.5 Observations

242 Oxygen and nutrient concentrations from the SCOBI model have been extensively evaluated against observations (Eilola et al., 2009, 2011,

243 well as other models (Eilola et al., 2011). Phytoplankton observations are more difficult to come by and our basin integrated

244 approach makes it difficult to compare with observations from individual stations.

245 We have used a basin integrated dataset of monthly Chl-a for the Baltic Proper previously published in HELCOM (2012).

246 The dataset includes all data from the Data Assimilation System (DAS) which is a database of Baltic Sea monitoring data

247 hosted by the Baltic Nest Institute, Stockholm University, completed with data from the EUTRO-PRO project and HELCOM

248 Indicator Fact Sheets (HELCOM, 2012) . The surface layer was defined as the top 10m of the water column and coastal areas

249 were removed.

250 3 Results and discussion

251 The model results shown are monthly means integrated averaged over the basin. The different variables have also been vertically

252 integrated averaged over the mixed layer and/or from the mixed layer down to a depth of 150m. The first 20 yrs of the model 253 run is excluded to minimize spinup effects.

254 We start out will begin in Sect. 3.1 by scrutinizing the modelled concentration of phytoplankton and its seasonal cycle by

255 comparison with observations. In Sect. 3.3, the coherence between nutrient loads and mixed layer nutrient concentrations as

- 256 well as phytoplankton concentrations will be examined. Section 3.2 presenting the model results of phytoplanton biomass.
- 257 In Section 3.2 we will consider the composition of nutrients and its effect on the phytoplankton concentrations. The effect
- 258 of temperature and irradiance is serutinized the coherence with the phytoplankton biomass. Coherence between riverine
- 259 loads and mixed layer nutrients will be discussed in Sect. 3.4 and in Sect. 3.5 3.3. Section 3.4 examines the coherence of

260 the phytoplankton with temperature and irradiance. Finally, the coherence between mixed layer depth with phytoplankton is

261 examined. and phytoplankton biomass is considered in Sect. 3.5.

262 3.1 Phytoplankton - model and observations biomass

Figure ?? shows the model results of basin integrated Chl-a concentration (the sum of the three different phytoplankton) over 0-10m Fig. 3 shows the time-series of phytoplankton biomass (a) together with the observations described above. The results are thus here integrated over a fixed depth rather than the mixed layer to better compare with the observations. The top panel of Fig. ?? displays observations and model results for the period 1990-2009. In order to illustrate the difference from pre-industrial, model results for the period 1880-1999 are also shown, corresponding wavelet spectrum (b).

The top panel reveals that the largest values representing the spring bloom are underestimated in the model results compared to the observations. The model implements a constant C:Chl ratio of 50 in phytoplankton, while Jakobsen and Markager (2016) found that this ratio, in reality, varies throughout the year. The underestimation of the spring bloom in the model may therefore, at least in part, be explained by this simplified assumption. Furthermore, the wavelet transform reveals a strengthening in the model of The wavelet power (variance) of the 6 month period relative to the annual compared to the early period (panel (c) and

273 (d)decomposed signal (in color) is displayed as a function of time (x-axis) and period (y-axis). The black curves in Fig. ??).

274 This is caused by the large increase in cyanobacteria resulting in a stronger late summer bloom. The half year period is much

275 weaker in the observations. In the upper panel of Fig. ??, this is visable as a greater observed difference between 3(b) show the

276 95% confidence level relative to red noise.

Averaging over time generates the global power spectrum displayed in Fig. 3 (c). The wavelet spectrum clearly reveals two main periodicities - the annual and the semi-annual representing the spring and late summer blooms. The smaller difference in magnitude between the two blooms in the model results reflects as stronger signal with a 6 month periodicity in the wavelet spectrum (panel (c) in Fig. ??). autumn blooms. It is also clearly visable that the power on both periodicities increases markedly after 1950.

Kahru et al. (2016) found a shift in chlorophyll maxima from the diatom dominated spring bloom to the cyanobacteria
 summer bloom. Fig. 4 shows that a similar pattern emerges from our model run with five years of cyanobacterial chlorophyll
 maxima occurring after 1998.

285 3.2 Nutrient loads

To determine the effect of the riverine loads on the mixed layer nutrient concentrations we perform wavelet coherence. The
 result is shown in Fig. 12. We have used riverine DIN and DIP loads in the results presented below. The use of instead total
 bioavailable nutrient loads does not change the results.

The results show the clear annual cycle in riverine inputs and mixed layer nutrient concentrations. The phosphate loads show
 little coherence on any other periodicity but DIN displays strong coherence on longer periods. Furthermore, there is a tendency
 for a enhanced coherence during the later part of the run most likely caused by increased DIN loads.

The phase arrows on the annual scale points to the right during most of the run indicating that the seasonal peak in nutrient loads and mixed layer concentrations are concurrent. However, during the period 1900-1920 the direction of the phase arrows shifts upwards. This is a result of a persistent shift in the runoff maxima of about two months over the period. During this period the peak in mixed layer nutrient concentrations thus precedes the runoff peak. The interpretation of this is not straight forward but most probably it has to do with the searcity of observations and the use of an integrated Baltic Sea runoff dataset

297 3.2 Nutrients and nutrient limitation

The extent of anoxic bottoms in the Baltic Sea has increased markedly over the past century. Carstensen et al. (2014) found a 10-fold increase in the hypoxic area since the beginning of the 20th century. They explained this to be primarily due to increased nutrient loads causing increased primary production and resulting in an enhanced deep water respiration.

To further investigate the lack of inter-annual coherence between riverine phosphate loads and mixed layer phosphate, the wavelet coherence between mixed layer salinity and nutrients are examined and displayed in Fig.13. Mixed layer salinity is affected by freshwater input from land, precipitation, evaporation and mixing with deeper layers. The coherence spectrum reveals higher coherence between mixed layer salinity and phosphate (top) on interannual periodicities than between salinity and DIN (bottom) Changing nutrient patterns in the Baltic Sea due to spreading hypoxia have been discussed by e.g. Conley et al. (2002); Sa Anoxia causes sedimentary phosphate release. A clear relationship between hypoxia and total basin averaged phosphate was first shown by Conley et al. (2002) (and later expanded by Savchuk (2010)) on observational data from the Baltic Proper.

The effect of hypoxia on DIN is less straight forward. Expanding hypoxia increases the boundary area between anoxic and oxic water where denitrification occurs resulting in a further loss of nitrate. Furthermore, hypoxia induced reduction in nitrification results in a loss of nitrate. Valtera et al. (2007) found a negative relationship between basin averaged DIN and hypoxic area in observations from the Baltic sea. The coherence existing between salinity and DIN on periodicities longer than one year is antiphase i. e. low salinity here coheres with high DIN concentrations. In contrast, the in-phase coherence between salinity and phosphate suggests that the reason for the coherence is a greater importance of the internal source i. e. phosphorus release from the sediments that eventually reaches the mixed layer through mixing with deeper layers.

Figures ?? and ?? show the coherence between the riverine input of phosphate/DIN and mixed layer chl concentrations of 315 316 diatoms (top), flagellates (middle) and cyanobacteria (bottom). There is again a strong annual coherence. There seems to be a quite strong coherence between mainly diatoms and both nutrients on a 16 year period. However, given that the length We 317 318 illustrate the changing nutrient patterns for our model run in Fig. 5. In conjunction with the increased anoxic volume we find a clear increase in ammonium and a decrease in nitrate. This is due to a decrease in nitrification and an increase in denitrification. 319 320 The phosphate concentration increases from the mid 20th century through the rest of the model run does not even give room for ten 16-year periods, this probably reflects the overall pattern of simultaneous increase in riverine loads and chlorophyll 321 eoncentrations over the second half of the 20th century as a combined effect of the accumulated terrestrial inputs and hypoxic 322 323 sedimentary release.

324 3.3 Nutrients and nutrient limitation

We will here assess the coherence of nutrients with the phytoplankton concentrations. Furthermore, as described above, the The 325 effect of nutrients on the primary production is in the model controlled by the term NUTLIM, or degree of nutrient limitation, 326 in Eq. (1). We thus NUTLIM can be viewed as a measure of the nutrient composition that linearly affects the phytoplankton 327 328 growth in the model. We will examine this term in and below the mixed layer. Even though there is no primary production in 329 the deep water and thus the nutrient limitation term has no effect here, a shift in the composition of nutrients in the deep water 330 will affect also the mixed layer. NUTLIM for the different plankton groups diatoms and flagellates has been calculated offline from the monthly means according to EqsEq. (2)and (7). 331 332 The evolution of NUTLIM in the surface layer and the deep water for diatoms and flagellates is shown in Fig. 6. There is a

333 clear increase over the 20th century and a shift towards less limited conditions (NUTLIM approaching 1).

Nitrogen has been shown to most often be limiting in the Baltic Proper, while phosphate is limiting in the northern basins (Granéli et al., 1990; Tamminen and Andersen, 2007). HoweverSchernewski and Neumann (2004) showed through a reconstruction of the Baltic Sea trophic state in the early 1900 that N/P ratios in the Baltic Proper have decreased but that much of the domain still indicated N limitation.

Using the models definition of nutrient limitation, our model results, displayed shown in Fig. 7, show display phosphate limitation for both diatoms and flagellates for the earlier part of the run. After 1980, seasonality appears in the mixed layer. Phosphate is still limiting during winter while nitrogen becomes limiting after the spring bloom. Calculating N/P ratios as a more conventional measure of nutrient limitation, our model results diplay instead a shifting pattern until 1976 whereafter persistant N limitation develops (not shown).

The extent of anoxic bottoms in the Baltic Sea has increased markedly over the past century. By compilation of a large amount of temperature, salinity and oxygen observations Carstensen et al. (2014) found a 10-fold increase in the hypoxic area since the beginning of the 20th century. They explained this to be primarily due to increased nutrient loads from land causing increased deep water respiration but also due to increased temperatures resulting in reduced oxygen solubility.

In order to understand the limitationpatterns found in our model run, we view the evolution of different nutrient concentrations.
Figure 5 shows the anoxic volume together with the below mixed layer nutrient concentrations. In conjunction with the increased anoxic volume we find a clear increase in ammonium concentration. This is due to a decrease in nitrification and is seen also as a decrease in the nitrate concentration. Furthermore, expanding anoxic bottoms increase the boundary area
between anoxic and oxic water where denitrification occurs resulting in a further loss of nitrate.

Figure 5 also shows that the phosphate concentration increases from the mid 20th century through the rest of the model run.
 This is a combined effect of increased riverine loads and enhanced sedimentary release due to anoxia.

The mixed layer displays corresponding patterns of increased phosphate and decreased nitrogen (Fig. ??). The seasonal variations are however much greater since the majority of the primary production occurs here and since the mixed layer is directly affected by riverine input. The mixed layer also comprises a smaller volume of water. Despite quite high wintertime concentrations, the spring bloom almost completely depletes the nitrogen. The seasonality that appears after 1980 in mixed layer nutrient limitation with nitrogen limitation after the spring bloom is thus a results of the larger relative increase in phosphate compared to nitrogen. 360 The sum of the effects on the nutrient concentrations shows up in the nutrient limitation expressions (Eqs. (3)-(6)).

361 The evolution of NUTLIM in the surface layer and the deep water for the three phytoplankton is shown in Fig. 6. There

362 is a clear increase over the 20th century and a shift towards less limited conditions. The changing nutrient patterns affects the

363 phytoplankton growth. We analyse the wavelet coherencies of phytoplankton biomass with mixed layer phosphate and DIN in

364 Figs. 8 and 9.

After 1980 there is a shift in the variability of nutrient limitation for diatoms and flagellates most clearly visable in the deep water. This shift is also visable in the lower two panels of Fig. 7 which show that deepwater NUTLIM shifts towards a purely nitrogen limited regime while NUTLIM for flagellates mostly display a seasonal pattern. The lower variability is due to the characteristics of the nitrogen limitation Eq. (3). The concentrations of nitrate and ammonium at the end of the model run corresponds to a minimum in Eq. (3). Therefore, even though the concentrations change, NUTLIM changes very little.

To see how the phytoplankton concentrations are connected to nutrient concentrations and nutrient limitation, we continue
 by scrutinizing the wavelet coherencies.

372 Figure 8 and 9 show the wavelet coherencebetween mixed layer phosphate and DIN and phytoplankton. Diatoms which

373 are the most Coherency is shown in color as a function of year (x-axis) and period (y-axis). More yellow indicates stronger

374 coherence. The arrows reveal the phase-lag between the two time-series. The line plots on the right show the time averaged

375 <u>coherence. As the strongest</u> nutrient limited groupshow strong, diatoms show persistant inter-annual coherence with phosphate
 376 during the first, consistently phosphate limited part of the run (see Fig. 7). During the later part of the run the nutrient and
 377 phytoplankton concentrations are so high high enough that smaller inter-annual variations have little effect.

Since nitrogen limitation in the model only occurs after 1980 and after the spring bloom and thus only affects the much
smaller diatom and flagellate autumn blooms no coherence between phytoplankton and nitrogen is visable in Fig. 9.

To further illustrate the shift from the more nutrient limited regime of the first part of the run we calculate the wavelet
 coherence between NUTLIM for the different phytoplanktonand the result is displayed in Fig. ??. Again, diatoms show strong
 coherence during the first, more nutrient limited part of the run.

In Fig. 10-To scrutinze the shift in deep water nutrient composition and the coherence with phytoplankton, we calculate the 383 384 wavelet coherence between below mixed layer NUTLIM and the three types of phytoplankton. Again, the coherence spectrum 385 shows the most inter-annual coherence for the more nutrient limited diatoms. However, the phase arrows diatom and flagellate 386 biomass. The result is shown in Fig. 10. The phase arrows here display some interesting features. After 1980 the phase arrows 387 within the annual coherence period change direction to the opposite direction. For diatoms, the phase shifts from NUTLIM 388 preceding diatoms by three months to diatoms preceding nutlim by the same amount. Flagellates display a similar shift. This occurs both for diatomswhere they shift from downward, indicating that the annual NUTLIM periodicity precedes the annual 389 390 diatom periodicity by 90 degrees, i.e. 3 months, to upwards, instead indicating that the diatoms precedes NUTLIM. A similar 391 pattern is visable also in flagellates.

To investigate the reasons for this, we have plotted the month of maximum NUTLIM in Fig. 11. The figures show a clear shift occuring after 1980 correlating with a strengthening of cyanobacteria blooms. The deep water 1980. Below the mixed layer, NUTLIM changes its maxima to the late summer months from December and January to July, August and September while a slight shift from February to March is apparent for diatoms. Mixed layer NUTLIM for flagellates displays no clear shift.

397 Figure 4 shows the timing of the maximum chlorophyll concentration for the different phytoplanktons as well as their sum.

398 Flagellates displays a weak shift towards May after 1960 but no other shifts are visable in the individual phytoplankton types.

399 However, the total chlorphyll concentration (Diatoms + Flagellates + Cyanobacteria) displays a few years at the very end of the

400 run where the chlorophyll maximum corresponds to the maximum for cyanobacteria. From satellite data, Kahru et al. (2016) found

401 a similar shift in chorophyll maximum from the spring bloom in May to the cyanobacteria bloom in July

402 3.3 Nutrient loads

403 The wavelet coherence between mixed layer nutrients and riverine input is shown in Fig. 12. We have used riverine DIN and

404 DIP loads in the results presented below. The use of instead total bioavailable nutrient loads does not change the results.

405 The phosphate loads show little coherence on periodicities longer than one year but DIN displays strong inter-annual

406 coherence. The phase-arrows indicate a phase-lag of about minus 45° on all inter-annual periodicities. For an 8 year period this

407 means that riverine input precedes DIN by about 1 yr.

408 To further investigate the lack of inter-annual coherence between riverine phosphate loads and mixed layer phosphate, the

409 wavelet coherence between mixed layer salinity and nutrients are examined and displayed in Fig. 13. Mixed layer salinity

410 is affected by freshwater input from land, precipitation, evaporation and mixing with deeper layers. The coherence spectrum

411 reveals higher coherence between mixed layer salinity and phosphate (top) on interannual periodicities than between salinity

412 and DIN (bottom). The coherence existing between salinity and DIN on periodicities longer than one year is antiphase i.e. low

413 salinity here coheres with high DIN concentrations. In contrast, the in-phase coherence between salinity and phosphate suggests

414 that the reason for the coherence might be a greater importance of phosphorus release from the sediments that eventually

415 reaches the mixed layer through mixing with deeper layers.

416 Riverine nutrient loads show little inter-annual coherence with phytoplankton biomass (not shown) other than on a 16 yr

417 period which probably reflects the overall pattern of simultaneous increase in riverine loads and phytoplankton biomass over

418 the second half of the 20th century.

419 3.4 Temperature and irradiance

The mixed layer temperature has increased over the 20th century. Figure 14 shows the 2-yr moving average of mixed layer temperature. To scrutinize the effect of temperature on the concentration of phytoplankton, the wavelet coherence between temperature and phytoplankton have been plotted in Fig. 15. The results suggest that the temperature increase after 1990 might have had an effect on cyanobacteria and flagellates. It is also noticable that the temperature increase observed between 1900 and 1940 probably had an effect on cyanobacteria. This is also in agreement with the model formulation where cyanobacteria are the most sensitive to temperature followed by flagellates. Light impacts primary production through the term LTLIM in Eq. (1). However, irradiance display very little variation on any other periodicity than the annual as can be observed in a wavelet power spectrum (not shown). Therefore there exists almost no coherence between phytoplankton and irradiance apart from the annual and semiannual.

429 3.5 Mixed layer depth

The lower panel of Fig. 14 shows the two year moving average of mixed layer depth averaged over the basin. We calculate thecoherence between mixed layer depth and diatoms, flagellates and cyanobacteria in Fig. 16.

Apart from the annual cycle there is a strong coherence between mixed layer depth and diatoms, and to some extent flagellates, on shorter periodicities as well. That is, the concentration of diatoms residing in the mixed layer seems to covary quite well on periodicities equal to or shorter than one year. The model value for diatom sinking rate is five times higher than that for flagellates while cyanobacteria is assumed to have no sinking rate. In a shallow mixed layer the diatom concentration decreases faster than in a deep mixed layer because of the large sinking rate. In the wavelet coherence spectrum we thus see in-phase short term coherence.

438 4 Summary and conclusions

With a main focus on inter-annual variations, the coherence of the mixed layer concentrations of phytoplankton phytoplankton
 biomass with key variables affecting the primary production has been examined for the Baltic Proper.

Riverine input of nutrients is an extremely important variable in the Baltie Sea and the large increase during the 20th century has initiated spreading of anoxic bottoms (Carstensen et al., 2014). We found quite strong coherence between riverine input of DIN and mixed layer DIN but not a similar relationship between riverine phosphate input and the corresponding mixed layer concentration. As mixed layer salinity displayed in-phase inter-annual coherence with phosphate and only weak anti-phase coherence with DIN we conclude that this is most probably due to a greater importance of the internal source of phosphate from lower layers.

We further found that the We found that the pattern of nutrient limitation in and below the mixed layer have changed in the model since 1980. Below the mixed layer, the limitation pattern changes from phosphate to nitrogen for diatoms and to seasonally shifting between phosphate and nitrogen. Within the mixed layer, the pattern changes from pure phosphate limitation to seasonally shifting for both diatoms and flagellates. This is due to decreased deep water oxygen concentrations and a rapid expansion of anoxia after 1970. The phosphate concentrations increase due to enhanced sedimentary release, denitrification results in loss of nitrate and reduced nitrification decreases the transformation of ammonium to nitrate. The combined effect results in nitrogen limitation after the spring bloom which benefits cyanobacteria.

The mixed layer concentrations of nutrients affect the primary production in the model through the nutrient limitation term, NUTLIM. The phytoplankton group most strongly limited by nutrients in the model is diatoms. The connection between primary production and the nutrient limitation term is visable as a strong inter-annual coherence between diatoms and phosphate as well as NUTLIM before 1940. After 1940 NUTLIM as well as and the concentrations of the individual phytoplankton 458 species has have gained such high values that smaller inter-annual variations have little effect on the production. Similarly, the 459 less nutrient sensitive group flagellates shows much smaller inter-annual coherence with phosphate even before 1940. NUTLIM 460 for this group is already high enough so that small long-term variations do not reflect strongly in the results.

Very little inter-annual coherence is visable also between phytoplankton and nitrogen. The spring bloom is phosphate limited throughout the run except for a few years after 1990 where diatoms display nitrogen limitation. The much weaker diatom and flagellate autumn bloom displays no inter-annual coherence with DIN most likely due to the high NUTLIM levels.

The shift in nutrient limitation patterns is also visable in a slight forward shift in the month of maximum mixed layer NUTLIM for diatoms after 1980, although a similar shift cannot be seen for flagellates. Below the mixed layer, maximum NUTLIM shifts significantly towards late summer for both diatoms and flagellates. Furthermore, the annual maximum of total chlorophyll concentration (Diatoms + Flagellates + Cyanobacteria) displayed a few years at the end of the run where the maximum corresponded to the autumn bloom due to the large increase in cyanobacteria. This is in agreement with Kahru et al. (2016) who found from satellite data that the annual chlorophyll maximum has shifted from the spring bloom maximum in May to the cyanobacteria bloom in July.

471 Riverine input of nutrients is an extremely important variable in the Baltic Sea and the large increase during the 20th century

472 has initiated spreading of anoxic bottoms (Carstensen et al., 2014). We found quite strong coherence between riverine input of

473 DIN and mixed layer DIN but not a similar relationship between riverine phosphate input and the corresponding mixed layer

474 concentration. As mixed layer salinity displayed in-phase inter-annual coherence with phosphate and only weak anti-phase
475 coherence with DIN we hypothesise that this is due to a greater importance of the flux of phosphate from lower layers.

The mixed layer temperature in the Baltic Proper has increased during the 20th century. We found some response of this mainly from the most temperature sensitive phytoplankton group cyanobacteria during periods of large interannual temperature increases. Flagellates, being more temperature sensitive than diatoms, seems to display a coherence with the temperature increase occuring after 1980.

Variations in mixed layer depth affects mainly diatoms as these have a high sinking speedvelocity. In-phase coherence on periodicities shorter than one year indicates that large seasonal changes in the mixed layer depth significantly affects the mixed layer concentrations while smaller interannual variations are of little consequence.

Finally, the effect of irradiance on primary production was scrutinized. However, interannual irradiance variations have very
little effect on the primary production Irradiance displayed very little coherence with phytoplankton biomass.

In conclusion, interannual variations have affected the primary production mostly through the limiting nutrient phosphate 485 before 1950 in our model run, through studying inter-annual wavelet coherence between simulated phytoplankton biomass and 486 key variables we have found that phytoplankton showed strong coherence with the limiting nutrient before 1950. After that 487 nutrients and phytoplankton exists in the water column at such high concentrations that smaller interannual variations have 488 489 much less effect. Furthermore, the mixed layer concentrations of DIN show strong interannual coherence with riverine DIN input while riverine phosphate displays almost no coherence with the corresponding mixed layer concentration. Instead, in-490 phase coherence with mixed layer salinity indicates a stronger importance of mixing with lower layers. Expanding low oxygen 491 conditions in the deep water has resulted in a shift in the composition of nutrients. In the model, this results in seasonality in 492

493 the nutrient limitation pattern of the mixed layer with phosphate limitation in the spring and nitrogen limitation after the spring

494 bloom Temperature displays some inter-annual coherence with the more temperature sensitive flagellates.

495 5 Data availability

496 The model data on which the results in the present study are based on are stored and available from the Swedish Meteorological 497 and Hydrological Institute. Please send your request to ocean.data@smhi.se.

Acknowledgements. This work was funded by the Swedish Research Council (VR) within the project "Reconstruction and projecting Baltic
 Sea climate variability 1850-2100" (Grant 2012-2117).

500 Funding was also provided by the Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning (FORMAS)

501 within the project "Cyanobacteria life cycles and nitrogen fixation in historical reconstructions and future climate scenarios (1850-2100) of

the Baltic Sea" (grant no. 214-2013-1449). The study contributes also to the BONUS BalticAPP (Wellbeing from the Baltic Sea - applications

combining natural science and economics) project which has received funding from BONUS, the joint Baltic Sea research and developmentprogramme.

This research is also part of the BIO-C3 project and has received funding from BONUS, the joint Baltic Sea research and development programme (Art 185), funded jointly from the European Union's Seventh Programme for research, technological development and demon-

507 stration and from national funding institutions.

508 We thank Brbel Muller-Karulis for providing the observational data.

509 References

- Almroth-Rosell, E., Eilola, K., Meier, H. E. M., and Hall, P. O. J.: Transport of fresh and resuspended particulate organic material in the
 Baltic Sea a model study, Journal of Marine Systems, doi:doi:10.1016/j.jmarsys.2011.02.005, 2011.
- Carey, C. C., Hanson, P. C., Lathrop, R. C., and St. Amand, A. L.: Using wavelet analyses to examine variability in phytoplankton seasonal
 succession and annual periodicity, Journal of Plankton Research, 38, 27–40, doi:10.1093/plankt/fbv116, http://www.plankt.oxfordjournals.
 org/lookup/doi/10.1093/plankt/fbv116, 2016.
- Carstensen, J., Andersen, J. H., Gustafsson, B. G., and Conley, D. J.: Deoxygenation of the Baltic Sea during the last century, Proceed ings of the National Academy of Sciences, 111, 5628–5633, doi:10.1073/pnas.1323156111, http://www.pnas.org/cgi/doi/10.1073/pnas.
- 517 1323156111, 2014.
- Cazelles, B., Chavez, M., Berteaux, D., Ménard, F., Vik, J. O., Jenouvrier, S., and Stenseth, N. C.: Wavelet analysis of ecological time series,
 Oecologia, 156, 287–304, doi:10.1007/s00442-008-0993-2, 2008.
- Conley, D. J., Humborg, C., Rahm, L., Savchuk, O. P., and Wulff, F.: Hypoxia in the Baltic Sea and Basin-Scale Changes in Phosphorus
 Biogeochemistry, Environ. Sci. Technol., 36, 5315–5320, doi:10.1021/es025763w, 2002.
- 522 Conley, D. J., Björck, S., Bonsdorff, E., Carstensen, J., Destouni, G., Gustafsson, B. G., Hietanen, S., Kortekaas, M., Kuosa, H., Markus
- 523 Meier, H. E., Müller-Karulis, B., Nordberg, K., Norkko, A., Nürnberg, G., Pitkänen, H., Rabalais, N. N., Rosenberg, R., Savchuk, O. P.,
- Slomp, C. P., Voss, M., Wulff, F., and Zillén, L.: Hypoxia-Related Processes in the Baltic Sea, Environmental Science & Technology, 43,
 3412–3420, doi:10.1021/es802762a, http://pubs.acs.org/doi/abs/10.1021/es802762a, 2009.
- 526 Dortch, Q.: The interaction between ammonium and nitrate uptake in phytoplankton, Marine Ecology Progress Series, 61, 183–201,
 527 doi:10.3354/meps061183, 1990.
- 528 Droop, M.: Some thoughts on nutrient limitation in algae, Journal of Phycology, 9, 264–272, doi:10.1111/j.1529-8817.1973.tb04092.x, 1973.
- 529 Eilola, K., Meier, H. E. M., and Almroth, E.: On the dynamics of oxygen, phosphorus and cyanobacteria in the Baltic Sea; A model study,
- 530 Journal of Marine Systems, 75, 163–184, doi:10.1016/j.jmarsys.2008.08.009, http://dx.doi.org/10.1016/j.jmarsys.2008.08.009, 2009.
- Eilola, K., Gustafsson, B. G., Kuznetsov, I., Meier, H. E. M., Neumann, T., and Savchuk, O. P.: Evaluation of biogeochemical cycles in an ensemble of three state-of-the-art numerical models of the Baltic Sea, Journal of Marine Systems, 88, 267–284, doi:10.1016/j.jmarsys.2011.05.004, http://dx.doi.org/10.1016/j.jmarsys.2011.05.004, 2011.
- Eilola, K., Mårtensson, S., and Meier, H. E. M.: Modeling the impact of reduced sea ice cover in future climate on the Baltic Sea biogeo chemistry, Geophysical Research Letters, 40, 149–154, doi:10.1029/2012GL054375, 2013.
- Eilola, K., Almroth-Rosell, E., and Meier, H. E. M.: Impact of saltwater inflows on phosphorus cycling and eutrophication in the Baltic Sea:
 a 3D model study, Tellus A, http://dx.doi.org/10.3402/tellusa.v66.23985, 2014.
- Flynn, K. J.: Ecological modelling in a sea of variable stoichiometry: Dysfunctionality and the legacy of Redfield and Monod, Progress in
 Oceanography, 84, 52–65, doi:10.1016/j.pocean.2009.09.006, http://dx.doi.org/10.1016/j.pocean.2009.09.006, 2010.
- 540 Graham, L. P.: Modeling runoff to the Baltic Sea, Ambio, 28, 328–334, 1999.
- 541 Granéli, E., Wallström, K., Larsson, U., Granéli, W., and Elmgren, R.: Nutrient limitation of primary production in the Baltic Sea Area,
 542 Ambio, 19, 1990.
- 543 Grinsted, a., Moore, J. C., and Jevrejeva, S.: Application of the cross wavelet transform and wavelet coherence to geophysical time series,
- 544 Nonlinear Processes in Geophysics, 11, 561–566, doi:10.5194/npg-11-561-2004, http://www.nonlin-processes-geophys.net/11/561/2004/,
- 545 2004.

- 546 Gustafsson, B. G., Schenk, F., Blenckner, T., Eilola, K., Meier, H. E. M., Müller-Karulis, B., Neumann, T., Ruoho-Airola, T., Savchuk, O. P.,
- and Zorita, E.: Reconstructing the development of baltic sea eutrophication 1850-2006, Ambio, 41, 534–548, doi:10.1007/s13280-012-
- 548 0318-x, 2012.
- Hansson, D., Eriksson, C., Omstedt, A., and Chen, D.: Reconstruction of river runoff to the Baltic Sea, AD 1500-1995, International Journal
 of Climatology, 31, 696–703, doi:10.1002/joc.2097, 2011.
- 551 HELCOM: Approaches and methods for eutrophication target setting in the Baltic Sea region., Balt. Sea Env. Proc. No. 1, 2012., 2012.
- 552 Jackett, D. R., McDougall, T. J., Feistel, R., Wright, D. G., and Griffies, S. M.: Algorithms for density, potential temperature, con-
- servative temperature, and the freezing temperature of seawater, Journal of Atmospheric and Oceanic Technology, 23, 1709–1728,
 doi:10.1175/JTECH1946.1, 2006.
- Jakobsen, H. H. and Markager, S.: Carbon-to-chlorophyll ratio for phytoplankton in temperate coastal waters: Seasonal patterns and rela tionship to nutrients, Limnol. Oceanogr., 61, 1853–1868, doi:10.1002/lno.10338, 2016.
- Kahru, M., Elmgren, R., and Savchuk, O. P.: Changing seasonality of the Baltic Sea, Biogeosciences, 13, 1009–1018, doi:10.5194/bg-131009-2016, 2016.
- Lau, K. and Weng, H.: Climate signal detection using wavelet transform: How to make a time series sing, Bulletin of the American Meteo rological Society, 76, 2391–2402, doi:10.1175/1520-0477(1995)076<2391:csduwt>2.0.co;2, 1995.
- Meier, H. E. M. and Kauker, F.: Modeling decadal variability of the Baltic Sea : 2 . Role of freshwater inflow and large-scale atmospheric
 circulation for salinity, Journal of Geofphysical Research, 108, 1–16, doi:10.1029/2003JC001799, 2003.
- Meier, H. E. M., Döscher, R., and Faxén, T.: A multiprocessor coupled ice- ocean model for the Baltic Sea: application to the salt inflow.,
 Journal of geophysical research, 108, doi:10.1029/2000JC000521, 2003.
- 565 Meier, H. E. M., Andersson, H. C., Arheimer, B., Blenckner, T., Chubarenko, B., Donnelly, C., Eilola, K., Gustafsson, B. G., Hansson, A.,
- 566 Havenhand, J., Höglund, A., Kuznetsov, I., MacKenzie, B. R., Müller-Karulis, B., Neumann, T., Niiranen, S., Piwowarczyk, J., Raudsepp,
- 567 U., Reckermann, M., Ruoho-Airola, T., Savchuk, O. P., Schenk, F., Schimanke, S., Väli, G., Weslawski, J.-M., and Zorita, E.: Comparing
- reconstructed past variations and future projections of the Baltic Sea ecosystem—first results from multi-model ensemble simulations,
- 569 Environmental Research Letters, 7, 034 005, doi:10.1088/1748-9326/7/3/034005, 2012.
- Meier, H. E. M., Höglund, A., Eilola, K., and Almroth-Rosell, E.: Impact of accelerated future global mean sea level rise on hypoxia in the
 Baltic Sea, Climate Dynamics, pp. 1–10, doi:10.1007/s00382-016-3333-y, 2016.
- 572 Parker, R. A.: Dynamic models for ammonium inhibition of nitrate uptake by phytoplankton, Ecological Modelling, 66, 113–120,
 573 doi:10.1016/0304-3800(93)90042-Q, 1993.
- Redfield, A. C.: The biological control of chemical factors in the environment, American Scientist, 46, 205–221, doi:10.5194/bg-11-15992014, 1958.
- Ruoho-Airola, T., Eilola, K., Savchuk, O. P., Parviainen, M., and Tarvainen, V.: Atmospheric nutrient input to the baltic sea from 1850 to
 2006: A reconstruction from modeling results and historical data, Ambio, 41, 549–557, doi:10.1007/s13280-012-0319-9, 2012.
- 578 Savchuk, O. P.: Large-Scale Dynamics of Hypoxia in the Baltic Sea, in: Chemical structure of pelagic redox interfaces: Observation and
- 579 modeling, Hdb Env Chem, edited by Yakushev, E. V., pp. 137–160, Springer-Verlag, Berlin Heidelberg, doi:10.1007/698_2010_53, 2010.
- 580 Savchuk, O. P., Wulff, F., Hille, S., Humborg, C., and Pollehne, F.: The Baltic Sea a century ago a reconstruction from model simulations,
- verified by observations, Journal of Marine Systems, 74, 485–494, doi:10.1016/j.jmarsys.2008.03.008, http://linkinghub.elsevier.com/
- 582 retrieve/pii/S0924796308000572, 2008.

- Savchuk, O. P., Gustafsson, B. G., Rodríguez, M., Sokolov, A. V., and Wulff, F. V.: External nutrient loads to the Baltic Sea , 1970-2006,
 2012.
- Schernewski, G. and Neumann, T.: The trophic state of the Baltic Sea a century ago: a model simulation study, Journal of marine systems,
 53, 109–124, doi:https://doi.org/10.1016/j.jmarsys.2004.03.007, 2004.
- Schimanke, S. and Meier, H.: Decadal to centennial variability of salinity in the Baltic Sea, Journal of Climate, pp. JCLI–D–15–0443.1,
 doi:10.1175/JCLI-D-15-0443.1, http://journals.ametsoc.org/doi/10.1175/JCLI-D-15-0443.1, 2016.
- Siegel, H., Gerth, M., and Tschersich, G.: Sea surface temperature development of the Baltic Sea in the period, Oceanologia, 48, 119–131,
 2006.
- Tamminen, T. and Andersen, T.: Seasonal phytoplankton nutrient limitation patterns as revealed by bioassays over Baltic Sea gradients of
 salinity and eutrophication, Marine Ecology Progress Series, 340, 121–138, doi:10.3354/meps340121, 2007.
- 593 Torrence, C. and Compo, G. P.: A practical guide to wavelet analysis, Bull. Amer. Meteor. Soc., pp. 61–78, 1998.
- 594 Tyrrell, T.: The relative influences of nitrogen and phosphorus on oceanic primary production, Nature, 400, 525–531, 1999.
- 595 Vahtera, E., Conley, D. J., Gustafsson, B. G., Kuosa, H., Pitkanen, H., Savchuk, O. P., Tamminen, T., Viitasalo, M., Wasmund, N., and Wulff,
- 596 F.: Internal Ecosystem Feedbacks Enhance Nitrogen-fixing Cyanobacteria., Ambio, 36, 186–193, 2007.
- 597 Winder, M. and Cloern, J. E.: The annual cycles of phytoplankton biomass., Philosophical transactions of the Royal Society of London.
- Series B, Biological sciences, 365, 3215–26, doi:10.1098/rstb.2010.0125, http://rstb.royalsocietypublishing.org/content/365/1555/3215,
 2010.



Figure 1. Study area. The grey scale represents depth in m.



Figure 2. The top panel shows riverine <u>phosphate loads DIN</u> (blue) and <u>mixed layer concentration of phosphate (red)</u> and the loads. The bottom panel shows riverine mixed layer DIN (blue) and <u>mixed layer DIN</u> phosphate (red).

Modelled basin integrated chlorophyll compared to observations. (a) shows observations (blue) and model results (red) for the period 1990-2009 together with model results for the period 1880-1999 (yellow). The lower three panels shows the wavelet spectra for (b) observations, (c) model results for 1990-2009 and (d) model results for the period 1880-1899. The y-axis shows the periodicity and the colors represent the wavelet power. The black curves in the wavelet figures represent the 95% confidence



Figure 3. Wavelet coherence between riverine phosphate and mixed layer phosphate concentration <u>Time-series of phytoplankton biomass</u> (topa) together with the corresponding wavelet power spectrum (b) and riverine DIN and surface DIN concentration global wavelet spectrum (bottomc). More yellow means more power. The arrows indicates black curves in (b) represent the phase lag. When pointing 95% confidence level relative to red noise. The white areas in (b) represent the right cone of influence in which the two time-series results are in phase impacted by edge-effects and when pointing in the opposite direction anti-phaseare therefore not shown. The right panels show different lines in represent the integrated coherence for the whole period global spectrum 1880-2009 (blue)and before, 1880-1899 (green)and after, 1990-2009 (red)1950...

level relative to an AR1 spectrum. (c), (f) and (g) show the corresponding global power spectra together with the AR1 spectrum

(red). The white areas in the wavelet figures represents the cone of influence in which the results are impacted by edge-effects
 and are therefore not shown.

607 Time-series of anoxic volume (top), below mixed layer concentrations of phosphate (blue) and DIN (nitrate + ammonium,

608 red) (middle) and nitrate (blue) and ammonium (red)(bottom).

609 Time-series of mixed layer phosphate (blue) and DIN (nitrate + ammonium, red) concentration (middle) and nitrate (blue)

610 and ammonium (red) concentration (bottom).

611 Wavelet coherence between mixed layer NUTLIM and diatoms (top), flagellates (middle) and cyanobacteria (bottom).



Figure 4. Wavelet coherence between mixed layer salinity and phosphate The month of maximum concentration (top) of diatoms, flagellates and mixed layer salinity and nitrate concentration (bottom) cyanobacteria as well as their sum. The right panels show the integrated coherence spectrum.

Figure 5. Wavelet coherence between riverine phosphate and diatoms_Time-series of anoxic volume (top), flagellates_below mixed layer concentrations of DIN (nitrate + ammonium, blue) and phosphate (red) (middle) and eyanobacteria_nitrate (blue) and ammonium (red)(bottom). The right panels show the integrated spectrum.

Figure 6. Wavelet coherence between riverine DIN and diatoms Time-series of nutrient limitation in the mixed layer (top) , flagellates and below (middlebottom) for diatoms (blue) and eyanobacteria flagellates (bottomred). The right panels thicker lines in the top panel show the integrated spectrum5yr moving average.

Figure 7. Nitrogen or phosphate limitation as function of time in the mixed layer (upper panels) and in the deep water (lower panels) of diatoms (left panels) and flagellates (right panels). Note that simultaneous N and P limitation is not possible although the size of the rings in the figures gives this appearence.

Time-series of nutrient limitation in the mixed layer (top) and below (bottom) for diatoms (blue), flagellates (red) and nitrogen fixation (yellow). The thicker lines in the top panel show the 5yr moving average.

Figure 8. Wavelet coherence between mixed layer phosphate concentration and diatoms (top), flagellates (middle) and cyanobacteria (bottom). More yellow means more coherence. The arrows indicate the phase lag. When pointing to the right the two time-series are in phase and when pointing in the opposite direction anti-phase. The right panels show the coherence averaged over the whole period (blue) and before (green) and after (red) 1950.

Figure 9. Wavelet coherence between mixed layer DIN concentration and diatoms (top), flagellates (middle) and cyanobacteria (bottom).

Figure 10. Wavelet coherence between deep water NUTLIM and diatoms (top), flagellates (middle)and eyanobacteria (bottom)

Figure 11. The month of maximum NUTLIM for diatoms (left) and flagellates (right) in the mixed layer (top) and below (bottom).

Figure 12. The month of maximum-Wavelet coherence between riverine phosphate and mixed layer phosphate concentration of diatoms, flagellates (top) and eyanobacteria as well as their sumriverine DIN and mixed layer DIN concentration (bottom). The arrows indicates the phase lag. When pointing to the right the two time-series are in phase and when pointing in the opposite direction anti-phase. The right panels show the averaged coherence for the whole period (blue) and before (green) and after (red) 1950.

Figure 13. Wavelet coherence between mixed layer salinity and phosphate concentration (top) and mixed layer salinity and nitrate concentration (bottom). The right panels show the averaged coherence spectrum.

Figure 14. 2-yr moving average of mixed layer temperature (top) and mixed layer depth (bottom).

Figure 15. Wavelet coherence between mixed layer temperature and diatoms (top), flagellates (middle) and cyanobacteria (bottom).

Figure 16. Wavelet coherence between mixed layer depth and diatoms (top), flagellates (middle) and cyanobacteria (bottom).