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Phosphorus burial in the ocean over glacial-interglacial time scales

F. Tamburini[1,*] and K. B. Föllmi[2,]**

[1]Geological Institute, ERDW, ETH Zurich, Switzerland
[2]Institut de Géologie, Université de Neuchâtel, Switzerland
[*]now at: Plant Nutrition Institute, AGRL, ETH Zurich, Switzerland
[**]now at: Institut de Géologie et Paléontologie, IGP, Université de Lausanne, Switzerland

Correspondence to: F. Tamburini (federica.tamburini@ipw.agrl.ethz.ch)

Abstract

The role of nutrients and their impact on primary productivity and the fluctuations in atmospheric CO_2 over glacial-interglacial periods are intensely debated. One of the cornerstones is the role of phosphorus (P; in the form of phosphate). Suggestions as to the importance of P evolved from an earlier proposal that P actively participated in changing productivity rates and therefore climate change, to the most recent one that changes in the glacial ocean inventory of phosphorus were small and not influential if compared to other macronutrients, such as nitrogen. Using new data coming from a selection of ODP sites, we illustrate oceanic P sedimentary phases distribution and reactive P burial fluxes, and we show how P burial fluxes changed over the last glacial-interglacial period. Concentrations of reactive P are generally lower during glacial times, while mass accumulation rates (MAR) show higher variability. On a global scale, glacial burial fluxes of reactive P are lower than those during interglacial periods by 7–10%, because lack of burial of reactive P on the glacial reduced shelf was apparently not compensated by burial in other regions of the ocean. Using the calculated changes in P burial, we try to infer their possible impact on the phosphate inventory in the world oceans. Using a simple mathematical approach, we find that these changes alone could have increased the phosphate inventory of glacial ocean waters by 20–40% compared to interglacial stages. Variations in the C/P reactive ratios and in the P sedimentary phases distribution at the investigated sites seem to indicate that at the onset of interglacial stages, shallower sites experienced an increase in reactive P concentrations. This seems to point to P-richer waters at glacial terminations, supporting the shelf-nutrient hypothesis and giving phosphorus a role as a potential player in climate change.

1 Introduction

Phosphorus (P) is a limiting nutrient and this property closely couples it to the carbon (C) cycle. Therefore, past variations in its global budget (i.e., inputs from the continent vs. output to the sediments) have been invoked as a factor influencing climate change (Föllmi, 1996; Van Cappellen and Ingall, 1994). Due to the increased availability of sediment cores and development of accurate speciation techniques (Anderson and Delaney, 2000; Ruttenberg, 1992; Schenau et al., 2000), research on the P cycle in marine environments has intensified. It is widely accepted that degradation of organic matter and early diagenetic transformations in the sediments impact the P cycle, which may also be released to pore and bottom waters. The degree of diagenetic redistribution and regeneration of P is generally dictated by local and global processes, such as organic matter deposition to the sea floor, water column ventilation, oceanic circulation, and sedimentation rates. It is clear that long-term changes in these processes should be taken into account when studying the P cycle over glacial-interglacial times. An accurate compilation for modern sediments (Baturin, 1988) and, recently, other contributions have provided detailed estimates of P burial fluxes in the oceans (Delaney, 1998; Filippelli, 1997; Ruttenberg, 1993). However, few studies provided estimates of P burial flux during the last glacial period (Filippelli et al., 2007), and only few recent studies show local records of glacial P concentrations and mass accumulation rates (MAR) (Filippelli et al., 2007; Ganeshram et al., 2002; Schenau et al., 2005; Tamburini et al., 2003). As a consequence, all glacial reconstructions of phosphate concentration in the ocean have been generally based on the use of other proxies, such as δ^{13}C and Cd/Ca of benthic foraminifers (Boyle and Keigwn, 1987) and on their empirical relationships with phosphate concentration (Broecker and Peng, 1982). Here, using material coming from sediment cores distributed around the world oceans, we present sedimentary P concentrations, P MAR and C/P reactive ratio data covering the last glacial-interglacial period. After a description and discussion about the distribution and variations of P in the sediments, we use these data to provide a preliminary calculation on how the phos-

phate inventory of the ocean could have changed over glacial interglacial time scales. Because our knowledge of the marine P cycle is still not complete, we also consider these data of great importance, since they may be used to validate already existent geochemical models.

2 Materials and methods

Eight ODP (Ocean Drilling Program) Cores were chosen from different environmental and oceanographic settings (Fig. 1 and Table 1) and sampled. Depending on sediment accumulation rates, sampled intervals vary from 3 m (ODP Site 957) to 114 m (ODP Site 1144). To account for the post-depositional redistribution pattern, P was extracted using a modified SEDEX method (Filippelli and Delaney, 1996), which identifies four sedimentary phases containing P; i.e. three reactive phases (loosely and Fe-bound, authigenic, and organically-bound), and one unreactive detrital phase (for averages of the different P phases concentrations and reactive P MAR, see Table 1; for a compilation of all data, see Supplementary Material: http://www.biogeosciences-discuss. net/5/5133/2008/bgd-5-5133-2008-supplement.pdf). P concentrations in the different extracts were then quantified by colourimetry and by ICP-OES (Eaton et al., 1995). The original SEDEX method allows also the quantification of the loosely-bound or exchangeable P phase (generally P adsorbed on particle surfaces), which in the method used for this study is associated with Fe-bound P (Ruttenberg, 1992). This simplification was introduced because the targeted samples are ancient sediments, where concentrations of the exchangeable P phase are likely to be small and unimportant. In any case, the used analytical approach allows the distinction between bioavailable (reactive P) and unreactive phases, which is key to unraveling the relationship between the P and C cycles (Ruttenberg, 1992). Although the sedimentary phases recognized are operationally defined, the comparison of the P phases with other mineralogical and geochemical proxies indicates that the differentiation made during the extraction is reflecting a real distribution in the sediments (Tamburini, 2002). Organic carbon was quantified on about 100 mg of dried and ground sediment, with a Rock-Eval 6, and

using a standard whole rock pyrolysis method (Espitalié et al., 1986). The method consists of a pyrolysis step (temperature at 300°C for 3 min then gradual increase to 650°C) and an oxidation step (temperature gradient from 400°C to 850°C). Total organic carbon is obtained as the sum of four peaks: the S_1 peak (hydrocarbons released at
5 300°C, FID-curve), the S_2 peak (hydrocarbons produced between 300°C and 650°C), the S_3 peak (CO_2 from pyrolysis of organic carbon up to 400°C, IR-curve), and the S_4 peak (CO_2 released from residual organic carbon below ca. 550°C during the oxidation step). All analyses were performed at the Geological Institute of the University of Neuchâtel, Switzerland. Sediments were attributed to glacial and interglacial isotopic
10 stages characteristic of the last 150 000 years, based on the oxygen stable isotope stratigraphy available for each ODP Site, and P MAR were calculated (as the product of P concentration, dry bulk density, and sedimentation rates, see Table 1). Age models, sedimentation rates and dry bulk density data were obtained from the ODP literature, both ODP Preliminary Reports and Scientific Results volumes available for
15 each studied Leg (see Table 1 and Supplementary Material for the complete list of references). To estimate P burial fluxes (BF), we have used the average MAR and the areal extent of the oceanographic setting to which each site belongs (Tables 1 and 2). During glacials, the areal extent of the exposed shelf has been considered equal to 16×10^6 km^2 (Meybeck and Vörösmarty, 2005), while only 12.8×10^6 km^2, equal to
20 40% of the modern shelf, remained flooded. The estimated relative error for the BF is about 7–10% (calculated as the sum of the relative errors associated with each parameter included in the calculation of the burial fluxes). Because ODP does not target shallow water and shelf environments, we looked in the literature for reactive P data obtained using the SEDEX extraction. Data from Mississippi and Long Island Sound,
25 Indian margin and Japan Sea were averaged and the value was used to calculate burial fluxes for the shelf environment (Babu and Nath, 2005; Cha et al., 2005; Ruttenberg and Berner, 1993) (see Table 2). To account for P burial in hydrothermal regions, we use the value proposed by Wheat et al. (2003), who have evaluated the importance of hydrothermal processes in scavenging P. Based on several ODP and DSDP data,

they estimated a burial flux of P in these environments (e.g. mid-oceanic and volcanic ridges) of about 2.8×10^{10} mol/yr (Wheat et al., 2003). We have assumed this flux to be constant over glacial-interglacial periods. We are aware that our sites are not fully representative of world oceans, since several regions, such as the Southern Ocean or the Equatorial Pacific, important in terms of productivity and C cycle, are not included in this study. However, the calculated interglacial BF (33.2×10^{10} mol/yr) is in the range of independently published data and estimates between 28×10^{10} and 36×10^{10} mol/yr (Ruttenberg, 2005; Wallmann, 2003). For this reason, we consider that our data can be suitable for calculating glacial burial fluxes, which have been generally estimated only by indirect methods (Wallmann, 2003). The use of BF in our discussion avoids any overestimation of single location MAR. For instance, it is true that MAR are highest at the Peru and Oman upwelling sites (see Table 1 and Fig. 2), but BF in upwelling environments represent on average not more than 15% of the total BF.

3 Results

3.1 Phosphorus and carbon data

Authigenic P represents the most important sink of reactive P (considered the sum of loosely and Fe-bound, authigenic, and organically-bound), and averages between 36 and 78% of reactive P. In interglacial sediments, reactive P concentrations fall in a relatively narrow range, between 9.5 and 18 µmol/g (Fig. 2), while high reactive P concentrations are observed at the Peru and Oman margin sites (Table 1 and Fig. 2a). These two sites represent high productivity sites (total primary production higher than 500 mg C/m^2 per day), where phosphorite deposition also occurs. A similar distribution for P concentrations is observed in glacial sediments (Fig. 2), with authigenic P being the most important reactive phase. In most sites, we observe lower amounts of reactive P during glacial times, generally attributable to a decrease in loosely- and Fe-bound P, while authigenic P seems to increase. Concerning variations of the sedimentary P distribution, it is important to note that there is no evidence of a substantial diagenetic

imprint on the P records (for sedimentary P phases data, see Supplementary Material). In fact, even if at many sites organic P is characterized by a down-core decrease trend, the loss in organic P is not enough to explain the changes observed in the other reactive phases. In upwelling areas, loosely- and Fe-bound P show lower concentra-
5 tions at the beginning of interglacial stages, while in the other environments, this phase is always higher than during glacial stages (Fig. 3). Authigenic P is higher at glacial terminations and shows higher concentrations during interglacial stages in upwelling and rise environments. In slope and basin sites, this phase shows, on the other hand, lower values at glacial terminations and during interglacials. Organic P mimics organic
10 C behavior at each site, and shows low concentrations during glacial times. At the site level, it exhibits a range of patterns: no evident trend at basin sites, a variable behavior at rise sites (low concentrations at termination at Site 1143 and high at Site 1144), and generally higher concentrations during interglacials at upwelling sites. As expected, detrital P concentrations are higher during glacial periods. We have used organic C and P
15 data to calculate C/P organic and reactive molar ratios (Anderson and Delaney 2001; see Supplementary Material for all C/P ratios data). Because of analytical problems due to high contents of carbonate and detrital material, the C/P ratios for the two basin sites (ODP 860 and 907) are to be taken with caution, and only trends are discussed. C/P_{org} ratios approach the Redfield at the top cores at all but upwelling sites, where
20 higher values are observed. Both C/P_{org} and $C/P_{reactive}$ records generally show higher values during glacial times, peaking at glacial terminations and towards lower values during interglacial periods. Only at Sites 112 (upwelling) and 907 (basin) $C/P_{reactive}$ ratios increase towards interglacial periods. Dictated mainly by local sedimentation rates (Filippelli, 1997), reactive P MAR change from site to site, and show more substantial
25 variations than those shown by concentrations (Fig. 2). Interglacial and glacial reactive P BF show different behavior depending on the environment. P BF in slope and rise settings are higher during glacials, but this increase did not compensate for lower BF in shelf and basin environments, so, on a global average, P burial fluxes are lower for glacial intervals by about 8% (Table 2). The estimates of the oceanic replacement time

of phosphorus, calculated as the ratio between the oceanic phosphate inventory (equal to 3.2×10^{15} mol, see Delaney, 1998) and phosphorus burial fluxes, range between 9.6 and 10.4 kyr, for interglacial and glacial times, respectively. Lower P burial fluxes during glacial periods may be explained by changes in deep water characteristics (such
5 as oxygen content, alkalinity, organic matter delivery and burial in sediments) that may have enhanced regeneration of P and release from sediments (Colman and Holland, 2000), or by temporal changes in P flux from the continent to the ocean. Even though our data suggest a decrease in detrital P fluxes from the continent, there is no clear evidence to suggest that glacial reactive P input from the continents was lower during
10 glacial periods. In fact, global weathering rates and sedimentation rates are estimated to be similar if not higher during glacial than during interglacial times (Kump and Alley, 1994). On the other hand, changes in bottom water chemistry during glacials may have lowered P BF and, as a consequence, had an impact on the P cycle. Studies have suggested that during the last glacial maximum ocean waters were more stratified (Jaccard
15 et al., 2005; Paillard and Parrenin, 2004), bottom waters were more alkaline (Paillard and Parrenin, 2004; Sanyal et al., 1997), and oxygen concentrations in bottom waters and in surface sediments were locally lower than during interglacial periods (Sirocko et al., 2000). Changes in these parameters might have a significant impact on the marine P cycle, in that they define the paths of organic matter degradation, redox cycling
20 of Fe, formation of P-bearing minerals, and degree of remobilization of P during early diagenesis (Ingall and Jahnke, 1997; Jarvis et al., 1994). Additionally glacial lowering of sea level exposed large portions of the shelves (Meybeck and Vörösmarty, 2005), where extensive fluvial systems developed (Pelejero et al., 1999), and previously deposited organic and inorganic material might have been reworked and transported off-
25 shore, potentially delivering additional reactive P to the glacial ocean and increasing the oceanic phosphate inventory. These hypotheses are compatible with data and models; Cd/Ca measurements in benthic foraminifera suggest that glacial deep waters were enriched in PO_4^{3-} (Boyle and Keigwn, 1987), and results of numerical models suggest the presence of a less ventilated ocean, with deep waters being enriched in CO_2 and

PO_4^{3-} (Toggweiler, 1999; Wallmann, 2003). Our data show that loosely- and Fe-bound P phase presents low values during glacial periods, especially at deep environments. Studies of Heinrich events in the North Atlantic have, in fact, demonstrated how this phase is sensitive to changes in circulation and oxygen content of deep waters (Tam-
burini et al., 2002). High values are on the other hand observed at glacial terminations and during interglacial periods, possibly indicating a more ventilated deep ocean. In a recent study based on data from a single sediment core recovered in the Mexican upwelling area, Ganeshram et al. (2002) also inferred a glacial decline in phosphogen-esis in continental margins and upwelling areas, but dismissed the hypothesis of an
increased glacial oceanic phosphate inventory, which was assumed to be not higher than 10% of its interglacial value. They, in fact, suggested that the lack of P deposition in shallow sites was compensated by deposition in deeper areas. They concluded that glacial increases in productivity were most likely small because of the slow and not important increase in the oceanic phosphate inventory Ganeshram et al. (2002). Our
data indicate indeed a decrease in P burial fluxes during glacial periods, but they also show that during glacial times phosphorus burial fluxes were not higher in deep ocean sediments.

3.2 Modeling phosphate concentration changes

Using the estimated phosphorus burial fluxes and a simple mathematical approach, we try to understand if the differences in burial fluxes might have changed the global oceanic phosphate inventory over a full interglacial-glacial cycle (120 kyr). With this approach, we do not take into consideration and integrate other data, such as carbon fluxes, because some of the investigated ODP sites lack detailed and accurate infor-mation. In this respect, our approach has a limited scope compared to geochemical models that have been proposed in the last years, which try to understand how phos-phate inventory in the oceans has changed over geological time scales in response to changes in ventilation of ocean waters, productivity, and oxygen content (see Wallmann (2003)). Because we consider changes in burial fluxes, we first state that changes in

Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

the phosphate inventory of the ocean over time are controlled by the difference be-
tween input and output fluxes:

$$\frac{\delta I}{\delta t} = F_{\text{input}} - F_{\text{output}} \tag{1}$$

where I is the ocean phosphate inventory, F is flux and t is time. We calculate changes
in the oceanic phosphate inventory for a time interval of 120 kyr, which represents
a full interglacial-glacial cycle, using a time step of 2 kyr (see Fig. 3), with initial full
interglacial conditions set at $t=0$ kyr, full glacial at $t=100$ kyr, and final full interglacial
at $t=120$ kyr. Integrating Eq. (1), we obtain:

$$I(t) = I(0) + F_{\text{input}} - F_{\text{output}} \tag{2}$$

where $I(t)$ at $t=0$ equals to the interglacial phosphate inventory. Not knowing the ex-
act value of the oceanic phosphate inventory during the last interglacial (about 130 kyr
ago), and being at present in an interglacial period, we choose to set $I(0)$ to mod-
ern values (3.2×10^{15} mol, see Delaney, 1998). One major problem is given by the
determination and quantification of F_{input}, considered as the total input (dissolved and
particulate) of reactive P from the continent, during interglacial and glacial times. In-
vestigations on sediment traps and core top material have shown that water column
particulate P, with the exception of the detrital fraction, is actively involved in the bio-
geochemical cycling of P (Faul et al., 2005). Values for the pre-anthropogenic flux of
total dissolved and particulate P input from the continent to the ocean are set to be
about 41.2×10^{10} mol/yr (Wallmann, 2003 and references therein). Knowing from our
dataset the value for detrital P flux into the sediments (average of 5×10^{10} mol/yr), we
obtain an estimate for the flux of reactive P of approximately 36×10^{10} mol/yr. As inter-
glacial value, we also test the hypotheses that the input of reactive P corresponds to 1.
45% of total particulate and dissolved riverine P flux (30×10^{10} mol/yr), as suggested
by Ruttenberg (2003), and 2. to the interglacial F_{output} (33.2×10^{10} mol/yr). First, we
assume F_{input} constant in time. Then, we hypothesize that F_{input} may have changed
with time, as suggested by detrital phosphorus burial flux, which decreases of about

Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

19% during glacial times (Table 2). We assume that this decrease is proportional to the decrease of the total input of material from the continent to the ocean. In this scenario, we consider that F_{input} linearly changes with time (t) between the interglacial and the glacial values, this last equaling to the 81% of the input during interglacial time. We
5 obtain, thus,

$$F_{input} = a_{in} - b_{in}t \qquad (3)$$

where a_{in} is F_{input} during interglacial and b_{in} the rate of change (see Table 3 for all parameters). For the last 20 kyr of the glacial-interglacial cycle, corresponding to deglaciation, F_{input} changed linearly between glacial and interglacial values, following

10 $$F_{input} = a_{in} + b_{in}t \qquad (4)$$

   The last parameter intervening in the mathematical model is F_{output}. First, we have assumed that P burial fluxes change linearly between the average interglacial and glacial values, as we are interested in the average change of the oceanic phosphate inventory between the two end-members of an interglacial-glacial cycle. As discussed
15 previously, changes in P burial fluxes are considered linked to the gradual change in sea level and to the transport of P-bearing matter to deeper parts of the ocean, but in this simple model, changes in sea level are only implicitly considered. So, we consider that F_{output} is linearly dependent on time (t)

$$F_{output} = a_{out} - b_{out}t \qquad (5)$$

20 where a corresponds to average interglacial P BF (33.20×10^{10} mol/yr) and b represents the change in output flux over time ($\Delta t = 100$ kyr), and is equal to 2.54×10^5 mol/yr^2. The values of a_{output} and b_{output} are obtained by linear regression between the average interglacial (33.20×10^{10} mol/yr) and glacial burial fluxes (30.66×10^{10} mol/yr). For the last 20 kyr, a and b are obtained by linear regression
25 between the average glacial and interglacial burial flux values. Second, we have calculated 2 kyr averages of global P burial fluxes over the ocean, based of the data obtained

Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

from the eight studied sites. We have integrated in Eq. (2) the different estimates for F_{input} and F_{output} and obtained 12 distinct simulations. Some simulations predict final oceanic phosphate inventory, which are unrealistic. In most of these cases, F_{input} is not balanced by the burial flux of phosphorus in the sediments, leading to an unreasonably high oceanic phosphate inventory. In few simulations, where F_{input} is set to lowest values, glacial and interglacial oceanic phosphate inventory after 100 and 120 kyr, respectively, are unrealistically low. Beside the simplicity of the approach, we think that these results are also biased by our limited knowledge of reactive P F_{input}. Only three simulations show results for interglacial times (120 kyr after beginning of simulation) that are coherent with present oceanic phosphate inventory and also agree with already published geochemical models (Fig. 4; see Table 3 for model parameters). These suggest a general increase of the global ocean phosphate inventory from the beginning of the interglacial to the glacial period, which values ranging from 3.75 to 4.71×10^{15} mol at full glacial ($t=100$ kyr), while at the re-establishment of interglacial conditions values from 3.81 to 4.72×10^{15} mol are observed. The glacial increase ranges between 17 and 47% of the initial interglacial oceanic phosphate inventory. In all three simulations, though, the final oceanic phosphate inventory is not set again to present values. Wallmann (2003), using a coupled benthic-pelagic model, estimated the effect of changing sea level, primary productivity, and oxygen concentration in bottom waters on nitrate and phosphate ocean inventories. His work shows that ocean phosphate inventory increased of about 40% from interglacial to glacial values. However, once interglacial conditions were re-established, the phosphate inventory did not return to the original values, because the high phosphate accumulated in the ocean during the glacial period was not completely used before the beginning of the new interglacial period (Wallmann, 2003).

4   Discussion

Although aware of the pure speculative character of our mathematical approach, we will try to integrate its outcomes to the P and C data presented here, and forward hy-

F. Tamburini and
K. B. Föllmi

potheses on how P burial fluxes and phosphate concentrations in the ocean might have changed during a glacial-interglacial cycle. Averaged over the world ocean, all our data point to a decreased burial of P during glacial periods, as the lack of deposition on the shelves was not completely compensated by burial at deeper sites. Loosely-, Fe-bound

5   and organic P concentrations decrease on average, and C/P ratios present generally higher values. One possible cause for decreased phosphorus burial is the increased settling time of transported particulate P to the deep ocean. The longer settling time is the result of a reduction of the shelf area caused by the lowering of sea level from interglacial to glacial periods. As forwarded by Filippelli et al. (2007), the shift in the

10 sinking loci of P from high to low sedimentation rates areas, from shallow to deep sites, would have increased the time residence of P in the water column, favoring the degree of recycling. The C/P ratios are strongly indicative of this higher degree of recycling, since P present in organic matter is preferentially regenerated during degradation than C. If the available estimates of reactive P input from the continent during glacial times

15 are not completely biased, even the small recorded change in P burial during glacial times, might have helped in building up phosphate in the ocean waters, increasing the global oceanic phosphate inventory. The increase could have ranged between 17 and 40%, as indicated by our modeling and in agreement with previous studies. Although extremely speculative, this increase could have had an impact on marine productivity.

20 Ocean mixing could have provided phosphate rich waters to the the surface ocean at intermediate and low latitudes, while at high latitudes (i.e. Southern Ocean and Subarctic Pacific), because of stratification of the water column (Frank et al., 2000; Jaccard et al., 2005), phosphate most likely accumulated in deep waters, with little impact on the surface water phosphate pool. Interglacial P burial fluxes are higher, as supported

25 also by high reactive P concentrations almost at all sites. C/P ratios are lower on average probably confirming higher retention of P in the sediments (C/P reactive), or different degree of organic matter degradation (C/P organic). At glacial termination and onset of interglacial periods, the observed peaks in several reactive P phases concentrations, especially at shallow sites, might testify an increased P availability and

retention capacity of the sediments. Overall, the data and the depicted scenario are compatible with the shelf nutrient hypothesis (Filippelli et al., 2007). Higher input of micro- and macronutrients from the continent, and the locally increased phosphate inventory might have played a role in creating a more productive ocean, thus maintaining
5 low glacial CO_2 concentrations the atmosphere (Martin et al., 1991; Moore and Doney, 2007). At the onset of the next interglacial period, after sea level rose and the restarting of oceanic circulation, rich phosphate deep waters might have been brought up to the surface on the now re-flooded shelves. The larger extension of the shelves provided the conditions for the formation of P-rich sediments, as testified by the peaks of reactive
10 P at several sites and as seen at other upwelling and shelf locations at the beginning of interglacial periods (Ganeshram et al., 2002; Tamburini et al., 2003).

5   Conclusions

We have shown that P burial fluxes were on average only 8% lower during glacial periods. However, no matter how little the difference between interglacial and glacial
15 phosphorus burial fluxes is, and even though our knowledge of reactive P input from the continent is limited, ocean phosphate inventory could have changed on glacial-interglacial time scales. P concentrations in sediments remained homogeneous over time, but lower P BF suggest that delivery of material from the exposed shelves to the deeper ocean and changes in chemical characteristics of deep waters from inter-
20 glacial to glacial periods, as forwarded by other studies, likely promoted the release of P from particulate inorganic and organic material. This could have increased phosphate concentrations of glacial waters. Provided favorable oceanographic conditions, the increased phosphate ocean inventory during glacial times might have contributed to enhance primary productivity. At the termination of glacial times and onset of the
25 interglacial period, the re-submerged shelves could have provided locations for burial of phosphate-rich sediments, as testified by P and C/P data. Our sediment phosphorus data and the calculation of the possible changes in oceanic phosphate inventory based on these, support several reconstructions and geochemical model outcomes targeting

Printer-friendly Version

Interactive Discussion

changes in phosphate behavior and inventory between interglacial and glacial times (Broecker, 1982; Filippelli et al., 2007; Slomp and Van Cappellen, 2007; Wallmann, 2003). The oceanographic changes associated with the transition from an interglacial to a glacial time, and the postulated changes in oceanic phosphate inventory, suggest that the oceanic P cycle is indeed influenced by climate variability, even on short time scales. This is also supported by the most recent estimates of P residence time , which are in the range of Milankovitch and sub-Milankovitch scales (Ruttenberg, 2005). Even if confined to only 8 locations, the P geochemical data presented in this study provide a valuable database to use and integrate in geochemical modeling studies and for further understand the P cycle and its complex link to climate. Although more data, especially from shallow environments are needed, interglacial to glacial variations in the geochemistry of ocean P may be seen as a potential player in the complex carbon cycle-climate system.

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Phosphorus in the ocean

F. Tamburini and K. B. Föllmi

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures

|◄ | ►|
◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

Full Screen / Esc

Printer-friendly Version

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F. Tamburini and
K. B. Föllmi

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Table 1. Marine isotopic stages (MIS), average phosphorus phases concentrations (in µmol/g), mass accumulation rates (MAR, average and range of values; in µmol/cm^2/kyr) of reactive and detrital phosphorus from the studied ODP Sites (n is number of points on which the averages are based). ** Fe-bound P: as discussed in Material and Methods, phosphorus associated to this sedimentary phase represents both exchangeable and Fe-bound P, as they are defined in Ruttenberg (1992).

MIS	Fe-bound P	Authigenic P	Detrital P	Org. P	Reac. P MAR	P MAR range	Detr. P MAR	n
Site 108–658; Eastern Tropical Atlantic; mbsl (meters below sea level): 2263, continental rise[a]								
1	4.57	8.9	1.1	6.32	340.3	279–443	18.92	4
2	2.88	8.72	2.45	2.93	344.9	316–385	58.16	5
3	3.69	8.82	1.39	3.97	205.67	130–363	17.35	4
4	/	/	/	/	/	/	/	/
5	4.99	9.52	0.9	3.3	231.14	156–334	11.68	12
6	2.19	9.22	1.58	1.57	208.09	/	25.33	1
IG					247.88		15.98	
G					322.17		41.74	
Site 112–680; Peru Continental Margin; mbsl: 252.2, upwelling, Oxygen Minimum Zone[b]								
1	43.48	44.57	2.93	9.38	2469.06	1963–2583	74.25	5
2	75.66	37.87	3.67	2.8	1669.02	1291–1873	52.65	3
3	33.85	60.06	2.47	2.42	617.32	461–777	15.83	5
4	31.65	78.02	5.12	2.25	962.98	722–1368	44.05	3
5	18	52.43	2.04	2.8	521.19	199–1249	14.52	9
6	35.23	11.91	0.58	2.35	398.75	/	4.67	1
IG					1059.08		34.87	
G					1184.96		33.79	

[a] Ages and sedimentation rates (SR) from Sarnthein and Tiedemann (1989). Dry bulk density (DBD) from ODP Initial Reports Vol. 108 (Ruddiman et al., 1988).
[b] Ages and SR from Wefer et al. (1990). DBD from ODP Initial Reports Vol. 112 (Suess et al., 1988).

Table 1. Continued.

MIS	Fe-bound P	Authigenic P	Detrital P	Org. P	Reac. P MAR	P MAR range	Detr. P MAR	n
Site 117–724; Oman Margin; mbsl: 292.8, upwelling, Oxygen Minimum Zone[c]								
1	3.12	52.42	5.7	2.21	978.98	196–2142	96.63	4
2	1.73	24.09	6.53	1.28	459.3	366–542	110.67	3
3	1.65	28.65	7.26	0.92	356.3	251–439	82.86	9
4	1.73	31.27	6.28	0.74	381.34	151–719	70.98	4
5	1.79	34.42	5.87	0.72	146.04	43–218	23.21	7
IG					407.24		67.56	
G					414.75		90.83	
Site 128–798; Japan Sea; mbsl: 900.1, slope[d]								
1	3.33	3.98	1.11	6.18	143.79	76–265	11.83	7
2	2.88	5.88	1.24	5.06	110.2	98–131	9.89	5
3	2.66	5.17	0.97	4.06	69.77	49–90	5.69	4
4	5.56	7.18	1.38	4.72	180.52	101–258	14.27	7
5	6.44	4.98	0.96	4.77	106.15	46–198	6.29	12
6	2.82	5.09	1.25	3.53	58.84	43–74	6.43	2
IG					111.28		7.94	
G					138.02		10.19	
Site 130–806; Ontong-Java Plateau; mbsl: 2520.7, above present lysocline, basin[e]								
1	8.26	5.12	0.41	1.08	57.96	37–76	1.64	3
2	3.11	6.7	0.49	0.91	40.56	38–46	1.85	3
3	5.24	6.57	0.46	1.04	42.41	33–61	1.52	6
4	3.46	7.52	0.44	1	27.68	26–30	1.02	3
5	3.73	6.43	0.62	0.68	33.84	18–58	1.94	11
6	1.96	5.38	0.42	0.65	143.2	/	7.53	1
IG					40.03		1.7	
G					31.29		3.47	

[c] Ages and SR from Zahn and Pedersen (1991). DBD from ODP Initial Reports Vol. 117 (Prell et al., 1989).
[d] Ages and SR calculated by graphic correlation of the grey scale provided in Föllmi et al. (1992) and of the age model provided in Tada et al. (1999). DBD from ODP Initial Reports Vol. 128 (Ingle et al., 1989).
[e] Ages and SR from Bickert et al. (1993). DBD from ODP Initial Reports Vol. 130 (Kroenke et al., 1991).

Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 1. Continued.

MIS	Fe-bound P	Authigenic P	Detrital P	Org. P	Reac. P MAR	P MAR range	Detr. P MAR	n
Site 151–907; North Atlantic Gateways; mbsl: 1800.8, low productivity, basin[f]								
1	5.82	4.18	1.75	3.35	17.62	17–18	2.31	2
2	4.75	7.72	3.16	2.63	22.42	18–27	4.69	2
3	5.9	9.54	2.75	2.32	24.76	20–29	3.83	5
4	6.6	10.24	3.33	2.58	28.12	25–31	4.82	2
5	6.91	8.73	3.03	2.46	26.05	22–29	4.36	8
6	6.13	11.27	2.15	2.16	27.07	26–29	2.98	2
IG					24.49		3.5	
G					25.87		4.16	
Site 184–1143; South China Sea; mbsl: 2772, carbonate platform, continental rise[g]								
1	5.52	4.18	0.69	3.46	31.82	30–33	1.67	5
2	3.86	2.76	1.05	3.51	41.86	25–49	4.34	5
3	3.31	2.42	0.93	3.6	44.82	37–63	4.47	21
4	2.1	4.09	1.37	2.96	41.01	29–52	6.14	9
5	3.51	7.19	1.05	2.54	45.13	13–107	3.58	16
6	3.42	3.73	0.63	2.31	15.45	/	1.03	1
IG					43.39		3.24	
G					39.59		3.84	
Site 184–1144; South China Sea; mbsl: 2037, sediment drift, continental rise[h]								
1	7.06	6.93	2.24	2.99	772.98	447–1341	101.97	7
2	3.58	6.67	2.49	2.79	1648.21	1195–1947	314.73	9
3	4.85	6.67	3.4	2.67	1495.13	804–3781	358.24	22
4	6.76	7.29	3.18	2.57	1539.53	975–2377	294.57	9
5	7.08	6.94	2.69	2.46	1195.68	240–1833	195.17	13
6	6.95	6.57	1.83	2.19	794.87	/	92.59	1
IG					1282.09		218.46	
G					1551.82		233.96	

[f] Ages and SR from McManus et al. (1996). DBD from ODP Initial Reports Vol. 151 (Myhre et al., 1995).

[g] Ages and SR from Tian et al. (2002). DBD from ODP Initial Reports Vol. 184 (Wang et al., 2000).

[h] Ages and SR from Bühring et al. (2000). DBD from ODP Initial Reports Vol. 184 (Wang et al., 2000).

Table 2. Global estimates of reactive phosphorus burial fluxes (P BF; in 10^{10} mol/yr). The areal extent of the exposed shelf during full glacial conditions is considered equal to $16 \times 10^6 \, km^2$ (Meybeck and Vörösmarty, 2005). The calculated global burial fluxes are then used in the modeling, the outcomes of which are presented in Fig. 4; see text and Table 3 for details.

Oceanographic settings	Areal extent	Interglacial IP BF	Glacial P BF	Δ G-IG	Interglacial detrital BF	Glacial detrital BF
Shelf[a]	28.9 (IG) 12.8 (G)	8.66	3.8	−56%	2.89	1.28
Slope (Site 798)	26.35	2.92	3.37	15%	0.23	0.27
Rise (Sites 658, 1143, 1144)	19.1	10.01	12.18	22%	1.51	1.78
Basin (Sites 806, 907)	151.3	4.84	4.23	−12%	0.39	0.58
Upwelling (Sites 680, 724)	5.42)	3.97	4.28	8%	0.28	0.38
Hydrothermal[b]		2.8	2.8	0%		
TOTAL		33.20	30.66	−8%	5.3	4.29

[a] Burial fluxes for the shelf environment are calculated using an average reactive P concentration of 12 μmol/gr, average sed. rates of 250 mm/kyr, and average DBD of 1 gr/cm^3.
[b] Hydrothermal phosphorus input from (Wheat et al., 2003).

Table 3. Parameters used in modeling, see Fig. 4 for outcomes.

Simulation	Time (kyr)	F_{input} (10^{10} mol/yr)	F_{output} (10^{10} mol/yr)	Phosphate inventory (10^{15} mol)	Increase[a]
1	0	36.00	33.20	3.2	
	100	29.00	30.66	3.75	17%
	120	36.00	33.20	3.86	21%
2	0	33.20	33.20	3.2	
	100	33.20	30.66	4.45	40%
	120	33.20	33.20	4.7	47%
3	0	30.00	2 kyr aver.	3.2	
	100	24.10	2 kyr aver.	4.56	43%
	120	30.00	2 kyr aver.	3.86	21%

[a] Burial fluxes for the shelf environment are calculated using an average reactive P concentration of 12 µmol/gr, average sed. rates of 250 mm/kyr, and average DBD of 1 gr/cm^3. b: Hydrothermal phosphorus input from (Wheat et al., 2003).
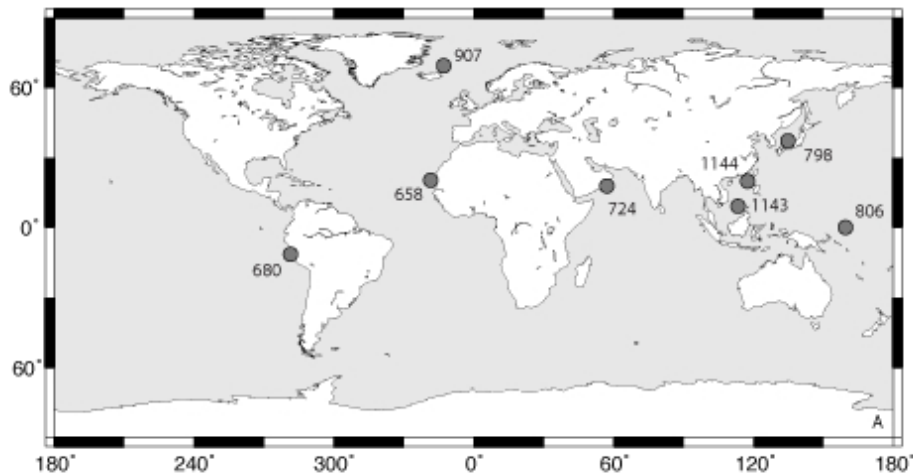
Fig. 1. World map with the investigated ODP Sites locations (see Table 1 for more information).

Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

Full Screen / Esc

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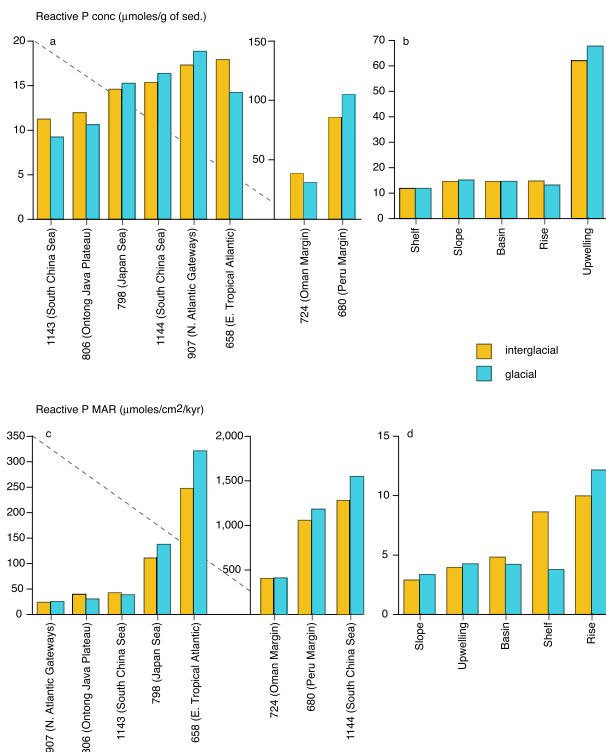
Interactive Discussion

Fig. 2. Reactive P concentrations (**a** and **b**) and MAR averages (**c** and **d**) for interglacial (dark bars) and glacial (light bars) periods. Vertical scales change for ODP Sites 680 and 724 (**a** and **c**). Concentrations of reactive P range between 9 and 105 µmol/g of sediment. Highest concentrations are observed in sediments from the Peru margin (ODP Site 680). Reactive P MAR range between 25 and 1500 µmol/cm^2/kyr, and the highest values are recorded in the northern part of the South China Sea (ODP Site 1144). Average concentrations and P MAR for the different oceanographic settings are reported in panels (**b**) and (**d**).
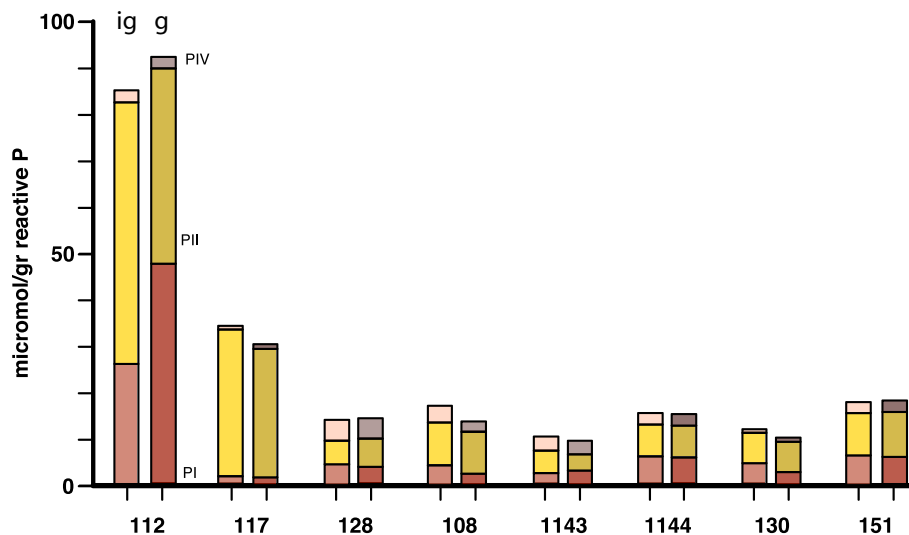
Fig. 3. Interglacial (lighter) and glacial (darker) reactive P concentrations at the studied ODP Sites. PI=loosely- and Fe-bound P; PII=authigenic P; and PIV=organic P.

Phosphorus in the ocean

F. Tamburini and
K. B. Föllmi

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄◄ | ►►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
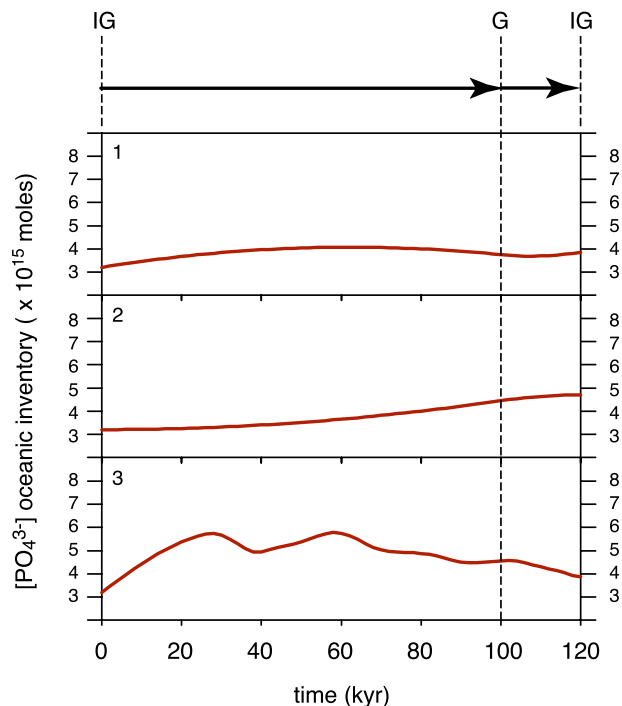

Fig. 4. Results of the interglacial-glacial simulations. The three parameters considered in the model are the initial oceanic phosphate inventory (set at interglacial values), F_{input} and F_{output} (see text for equations and discussion). 1: F_{input} and F_{output} are calculated by linear regression between interglacial and glacial average values. 2: F_{input} is considered constant over the investigated period, while F_{output} changes linearly. 3: F_{input} is calculated by linear regression between interglacial and glacial estimates of reactive P input from the continent, while F_{output} is represented by 2 kyr averages of oceanic P burial fluxes (see Table 3 for the complete list of the used parameters). $t(0\,kyr)$=initial interglacial condition, IG; $t(100\,kyr)$=glacial condition, G; $t(120\,kyr)$=final interglacial conditions.