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1 **Abrupt onset of carbonate deposition in Lake Kivu**
2 **during the 1960s: response to recent environmental**
3 **changes**
4

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1 **Abstract**

2 This study interprets the recent history of Lake Kivu, a tropical lake from the East African Rift Valley. The
3 current gross sedimentation was characterized from a moored sediment trap array deployed over two years. The
4 past net sedimentation was investigated with three short cores from two different basins. Diatom assemblages
5 from cores are interpreted as reflecting changes in mixing depth, surface salinity and Si:P ratio. The sediment
6 cores reveal an abrupt change ~40 years ago, when carbonate precipitation started. Since the 1960s, deep-water
7 methane concentrations, nutrient fluxes and soil mineral inputs have increased considerably and diatom
8 assemblages have altered. These modifications probably resulted from a combination of three factors, commonly
9 disturbing lake systems: the introduction of a non-native fish species, eutrophication and climatically-related
10 hydrological change, inducing greater upwelling. Catchment population increase accompanied by changes in
11 land use probably had a longer term impact, as the flux of soil minerals due to more intense erosion increased
12 earlier in the last century. The contemporary sediment trap data indicate seasonal variability, governed by
13 phytoplankton blooms during the annual mixing in the dry season, similar to Lakes Malawi and Tanganyika. The
14 ratio of settling fluxes to net sediment accumulation rates implies mineralization rates of 80-90% at the
15 sediment-water interface.

16

17

18 *Keywords: East Africa, nutrients, net and gross sedimentation, carbonates, diatoms,*
19 *ecological change.*

20

1 Introduction

2 Situated in East Africa and influenced by two active volcanoes, Lake Kivu has experienced
3 dramatic changes in its history (Degens et al. 1972). From 10,000-7,000 yr BP, Kivu was
4 shallower, 300 m below present level, and transformed into a deep lake around 6,000 yr BP.
5 About 5,000 yr BP carbonate deposition ceased and diatoms were completely modified in a
6 short time. These dramatic changes were attributed to volcanism and hydrothermal activities
7 (Haberyan and Hecky 1987). In modern Lake Kivu, dissolved gases (Schmid et al. 2005) in
8 the deep water are estimated at $60 \text{ km}^3 \text{ CH}_4$ and $300 \text{ km}^3 \text{ CO}_2$ (at $0 \text{ }^\circ\text{C}$ and 1 atm). Recent
9 studies indicate an increase in methane concentrations by 15% during the last 30 years
10 (Schmid et al. 2004), which might result from environmental modifications. First, gross
11 sedimentation may have increased due to the fast-growing population, intensified land use and
12 erosion on the steep catchment slopes. Second, the introduction of *Limnothrissa miodon* in
13 1960 altered the food web by eliminating the *Daphnia* communities (Dumont 1986). Third,
14 intensified rainfalls may have increased the subaquatic inflows, which delivered more
15 nutrients to the surface layer. Therefore, understanding the recent sedimentary history of Lake
16 Kivu is essential to characterize trends and changes of nutrient accumulation and to explain
17 why methane has increased.

18 In this study, net sedimentation was quantified with three short sediment cores from
19 Lake Kivu. Gross sedimentation was determined with material collected monthly in sediment
20 traps. We analyzed total organic carbon (TOC), total inorganic carbon (TIC), total nitrogen
21 (TN), total phosphorus (TP), biogenic silica (BSi), major and trace minerals, and diatoms.
22 This study addresses the following research questions: How did nutrient and carbon
23 accumulation change? How large is the seasonal variation in the gross sedimentation? Which
24 fraction of organic matter (OM) is degraded in the water column and at the water-sediment
25 interface?

1 The cores analysis provided evidence for an onset of calcite accumulation about half a
2 century ago. We consider possible triggers of this change in sedimentation regime, including
3 food web alterations after the fish introduction, land-use changes, and hydrological variability
4 modifying the upwelling rates.

5 **Study site**

6 Lake Kivu is located in the East African Rift Valley, between the Republic of Rwanda and the
7 Democratic Republic of the Congo. At an elevation of 1463 m, it has an area of 2370 km², a
8 volume of 580 km³ and a maximum depth of 485 m (Schmid et al. 2004; Tietze 1978)). The
9 lake (Fig. 1) consists of a major and four smaller basins (Botz et al. 1988). Subaquatic springs
10 enter below 180 m depth, with an estimated flow of 1.3 km³ yr⁻¹ (Schmid et al. 2005). In the
11 catchment (5097 km²), more than 127 rivers enter the lake (2.4 km³ yr⁻¹) and the Ruzizi River
12 (3.6 km³ yr⁻¹) is its outflow (Muvundja et al. in press). Precipitation (3.3 km³ yr⁻¹) is nearly
13 equal to the lake-surface evaporation (3.4 km³ yr⁻¹). The water inputs and outputs (7 km³ yr⁻¹)
14 are thus in equilibrium (Muvundja et al. in press). The oxycline varied seasonally from 30 m
15 in the stratified rainy season to 60 m in the dry windy season (June-August).

16 **Methods**

17 **Sites, coring and sediment trap sampling**

18 Gravity sediment cores (Fig. 1) were taken in May 2006 at Kibuye (2°02.886'S,
19 29°18.307'E, 190 m depth) and Ishungu (2°16.077'S, 28°59.374'E, 175 m depth) and in May
20 2007 at Gisenyi (1°46.383'S, 29°15.763'E, 155 m depth). Due to high gas concentrations,
21 undisturbed cores could only be taken above 200 m depth. The three cores were transported to
22 the Eawag laboratories in Switzerland, where they were sectioned at 0.5 cm intervals.
23 Samples were frozen and lyophilized. Dried samples were ground and homogenized.
24 Unfortunately, the Gisenyi core lost its uppermost section, as assessed by the ¹⁰Be absence in
25 the first layer.

1 One sediment trap mooring was set in Ishungu Basin in May 2006 (Fig 1). Sediment
2 traps consisting of two plastic cylinders (diameter 9.2 cm, length 100 cm) were placed at 4
3 different depths (50, 90, 130 and 172 m). Trap material was collected monthly with overlying
4 water into 250 ml bottles. Frozen samples were transported to Switzerland. Lyophilized
5 samples were weighed, ground and homogenized. During freeze-drying, salts contained in the
6 lake water contaminated our sediment samples. The water contribution was estimated by
7 assuming that the measured Na^+ stemmed only from the water phase. The other water
8 components (Ca, Mg, K, Na, Cl, TIC, TN, TP, Sr and Mn) were corrected based on their
9 molar ratios with Na^+ at the given depth (Pasche et al. 2009). TIC was corrected with the
10 molar ratio between Na^+ and alkalinity (HCO_3^-) and then with a CO_2 emission factor
11 accounting for CO_2 loss via carbonate precipitation by bivalent cations.

12 *Dating*

13 The three cores were dated using ^{210}Pb and ^{137}Cs activities measured by γ -counting in a well-
14 type Ge-Li borehole detector. As sedimentation rates showed two distinct slopes (Fig. 2), a
15 constant rate of supply was applied (Appleby and Oldfield 1978). The supported ^{210}Pb was
16 calculated as the averaged background activity below 20 cm, where ^{210}Pb remained constant.
17 This supported ^{210}Pb varied from 60 Bq kg^{-1} for Gisenyi to 133 Bq kg^{-1} for Ishungu. In
18 Kibuye, supported ^{210}Pb was calculated from ^{226}Ra activities, equalling 104 Bq kg^{-1} . The ^{210}Pb
19 supply averaging 330 $\text{Bq m}^{-2} \text{yr}^{-1}$ represented a high atmospheric flux probably explained by
20 the lake's remoteness from oceanic influence and the abundant rainfall (Appleby et al. 2001).
21 ^{210}Pb can only be used to date cores over the last 150 years. Sedimentation rates before 1850
22 were set to the second unit's average. ^{137}Cs activity peaks could not be detected.

23 *Analytical methods*

24 The total carbon (TC) was measured using a combustion CNS elemental analyser
25 (VARIO Co and EuroVector Co). TIC was analyzed as CO_2 by coulometry (UIC

1 Coulometrics) after acidification with 3M HCl. The difference between TC and TIC equalled
2 TOC. For TN and TP, sediment material was digested for 2 hours at 121 °C with
3 peroxydisulfate solution and then analyzed photometrically using a flow-injection analyzer
4 (FIA, Procon AG). For quantitative mineral analysis, major and trace elements were measured
5 using X-fluorescence (UniQuant). The sum of carbonates and organic content was determined
6 by loss on ignition at 1100 °C for 2 hours.

7 Biogenic silica was measured using the single-step wet-alkaline leach method
8 (Ohlendorf and Sturm 2008). Freeze-dried material was transferred to a Teflon crucible and
9 treated with 10 ml 1M NaOH. After digestion for 3 hrs at 90 °C, the centrifuged aliquot was
10 treated with 0.075M HNO₃ and BSi was measured within 24 h by inductively-coupled
11 plasma, optical-emission spectroscopy (ICP-OES/Spectro-Ciros). Simultaneous determination
12 of aluminum and sodium allows to correct (1:2 for Al: Si) for Si derived from silicate
13 minerals dissolution (Eggiman et al. 1980).

14 Samples were prepared for diatom analysis following the water bath method of Renberg
15 (1990). Diatom concentrations were estimated by adding a known number of inert
16 microspheres (Battarbee and Kneen 1982) and slides were mounted in Naphrax. At least 300
17 frustules per sample were counted in transects under oil-immersion phase-contrast light
18 microscopy (Leica DMRE microscope) at x1000 magnification. General (Germain 1981;
19 Krammer and Lange-Bertalot 1986; 1988; 1991a; 1991b) and regional floras (Cocquyt 1998;
20 Gasse 1986) were consulted to identify to species level. Preservation was assessed using the F
21 index (Ryves et al. 2001), the fraction of valves without dissolution signs to the total valves
22 (Ryves et al. 2006; Ryves et al. 2001).

23 The diatom data were divided into assemblage zones using optimal sum of squares
24 partitioning (Birks and Gordon 1985) by the program ZONE (version 1.2; (Juggins 2002), and
25 detrended correspondence analysis used to highlight the main underlying patterns of variation

1 (ter Braak 1995). Ordinations were implemented using CANOCO 4.54 (ter Braak and
2 Šmilauer 2002).

3 Quantitative diatom conductivity reconstructions were inferred using the European
4 Diatom Database website (<http://craticula.ncl.ac.uk/Eddi/jsp/>). Modern analogue matching of
5 full samples with conductivity dataset showed that the closest analogues were from the East
6 and North African subset, which was applied to fossil diatom assemblages using a weighted-
7 averaging model with inverse deshrinking ($WA_{inv}: r_{jack}^2 = 0.758$, RMSEP = 0.44 log units).
8 Goodness-of-fit is assessed by considering the minimum dissimilarity coefficient between the
9 fossil sample and training set (Juggins (2001). All quantitative reconstructions using diatom
10 data were carried out using the ERNIE software package within EDDI.

11 **Results**

12 **Contemporary gross sedimentation**

13 Material collected in sediment traps varied annually with maximum fluxes during annual
14 mixing between June and August. In 2006, TOC, TN, TP and BSi fluxes were maximal
15 between August and October (Fig. 7). In 2007, nutrients had their highest fluxes from June to
16 August. OM peak was higher in 2007 than in 2006. These maxima occur during the dry
17 season, when stronger winds lower the oxycline depth below 50 m. TIC was characterized by
18 peaks in June, November 2006, and October 2007.

19 Similar fluxes were observed for the three traps in the anoxic water but fluxes were
20 reduced at 50 m. The average sedimentation rates, TP, TN, TOC and TIC fluxes were similar
21 between 90 and 172 m, but smaller at 50 m (Table 2). Molar ratios between nutrients at 50 m
22 were always higher than below 90 m. The trap at 50 m was exposed to oxygen during annual
23 mixing, which might have favoured degradation and direct feeding during exposure.

24 A mass balance for sediment traps was established for its different fractions (Fig. 8).
25 $CaCO_3$ was determined as the average between TIC and CaO measurements. OM was

1 calculated as the sum of TOC, TN, TP, as well as O and H using the Redfield ratio. Secondly,
2 OM was determined from the loss on ignition minus the loss of CaCO₃ as CO₂. BSi and total
3 minerals were measured directly. BSi was measured only at 90 m and reported for the other
4 depths of the same sampling date. The total of these four components was compared to the
5 sedimentation rate, which agreed within 2%. The main constituents were minerals with a
6 slight minimum from mid-April to mid-June. Biomass and BSi had maxima from mid-June to
7 mid-October, TIC was more variable. OM fluxes remained constant throughout the water
8 column, whereas fluxes of soil minerals and CaCO₃ were smaller at 50 m. Colder and heavily
9 loaded stream water probably stratify below 50 m.

10 Diatom (Fig. 9) were analysed on monthly collections at 90 m from May 2006 to May
11 2007. All samples are dominated by *Nitzschia lancettula*, with seasonal appearances of *N.*
12 *bacata*, *N. amphibia*, *N. graciliformis* and *Synedra ulna*. There are low but persistent
13 occurrences of periphytic taxa, notably *Amphora pediculus* and *Encyonema muelleri*, but no
14 *Chaetoceros* cysts or *Urosolenia* spp. (Sarmiento et al. 2007) were seen. Testate amoebae
15 remains are always present with TA:D <0.1, except for June 2006 (0.25). In this sample, low
16 abundances of *N. amphibia* and *S. ulna* and significant amounts of pennate taxa suggest
17 substantial transport of littoral sediments to the trap. Diatom preservation is excellent. Diatom
18 concentrations are highest from June to August 2006 and April to May 2007.

19 **Past sediment accumulation**

20 *Sedimentation rates and visual description*

21 For sediment cores, the logarithmic ²¹⁰Pb activities revealed two slopes separated by an
22 interval, where white laminae appeared (Fig. 2). The shallower slope averaged 0.33 cm yr⁻¹,
23 whereas the deeper slope was 0.18 cm yr⁻¹. This extreme shift is caused by a sudden
24 carbonates accumulation, diluting ²¹⁰Pb activity. For the Gisenyi core, this pattern was

1 reproduced assuming a loss of the first 5.5 cm (Fig. 2). The constant rate of supply model
2 indicated that sedimentation rates increased by three fold (Table 1, Fig. 3).

3 An abrupt change from non-laminated dark to white laminae was visible in the upper
4 cores. This white layer corresponded, within dating uncertainties, to a transition in the early
5 1960s.

6 *Main components concentrations*

7 Concentration profiles of C, N, P (Fig. 3), BSi and minerals (Fig. 4) reflected changes
8 between these two layers. TIC increased abruptly at their transition. TOC and TN were less
9 concentrated in the white layer, while TP remained constant. N: P ratio was lower in the upper
10 layer. In the earlier record (< 1850), two OM peaks separated by a TIC peak were visually
11 observed in the Kibuye and Gisenyi cores (Fig 3). For the Kibuye core, BSi concentrations
12 have decreased since 1960. Elements linked to the carbonate cycle (Ca, Sr, Ba) showed a
13 clear increase in the white layer, while indicators for soil minerals (Si, Al, Ti) showed a
14 gradual increase from 30 cm depth upwards. Mg, K and Mn remained constant, while Fe and
15 S slightly increased.

16 *Main component fluxes*

17 Fluxes, derived from sedimentation rate and concentrations, increased in most constituents
18 since the 1960s (Fig. 3). To quantify the changes, the averaged flux in the carbonate layer was
19 compared to the averaged flux before 1960 (Table 1). TIC accumulation was 11 times higher
20 in the last 40 years, TP fluxes more than doubled, while TN increased by 60%. TOC fluxes
21 were less consistent; recent fluxes increased by 100%, 40% and decreased by 7% in the
22 Ishungu, Kibuye and Gisenyi cores, respectively. Total lithogenic fraction has increased by
23 3.5 times in the last 40 years (Fig 4). Carbonate forming elements increased by 14, 26 and 20
24 times for Ca, Sr and Ba, respectively. The Si, Al and Ti fluxes were augmented by factors of

1 3.2, 4.4, and 4, respectively. A mass balance for the Kibuye core was established for its
2 different fractions (Fig. 5).

3 *Diatom analyses*

4 Diatom analyses of the Kibuye core recorded 83 species (Fig 6). Five diatom assemblage
5 zones were identified (Kiv-1-5). The deeper layer (zone Kiv-1) is dominated by long, thin
6 *Nitzschia* species (*N. marginata*, *bacata*, *graciliformis*), with important contributions from
7 periphytic taxa (*Amphora pediculus*, *Encyonema muelleri* and *Rhopalodia gibba*). The testate
8 amoeba to diatom ratio (TA:D) is low. F index is very good with high BSi fluxes. Diatom-
9 inferred (DI) conductivity is around $1250 \mu\text{S cm}^{-1}$.

10 Zone Kiv-2 consists of Kiv-2a and Kiv-2b. The two subzones contain similar species
11 assemblages dominated by *Nitzschia* spp. At 25.5 cm, *Cyclostephanos dubius* and
12 *Stephanodiscus parvus* appear for the only time in the record, with a peak in *Chaetoceros*
13 *muelleri* (7%), the reappearance of periphytic taxa, and an increased TA:D. Preservation is
14 good, but reaches its lowest value at 25.5 cm, while diatom concentrations remain low
15 throughout. DI-conductivity is generally $< 2000 \mu\text{S cm}^{-1}$, but rises to $4000 \mu\text{S cm}^{-1}$ at 25.5
16 cm.

17 Kiv-3 is again dominated by *Nitzschia* taxa (*N. fonticola*, *confinis*, *bacata* and
18 *amphibia*) with *Amphora pediculus*, *Synedra ulna* and *Encyonema muelleri*. DI-conductivity
19 is $600 \mu\text{S cm}^{-1}$ while diatom preservation and concentrations are both high. High BSi fluxes
20 occur towards the top, coincident with the appearance of *Synedra ulna*.

21 Zone Kiv-4 is dominated by *Nitzschia fonticola*, with *N. confinis*, *N. amphibian* and *N.*
22 *inconspicua* all important, and *Chaetoceros muelleri* reappear. Preservation is good but
23 diatom concentrations are variable, and TA:D increases. DI-conductivity rises with a peak at
24 $4000 \mu\text{S cm}^{-1}$, while BSi fluxes fall from high values.

25 The most recent zone Kiv-5 is dominated by *Nitzschia* species and *Chaetoceros*
26 *muelleri*, with *Amphora pediculus*, *Encyonema muelleri* and *Synedra ulna* significant. The

1 TA:D reaches its maximum in the core, with diatoms well preserved but in low abundance.
2 DI-conductivity fluctuates, but surface sediment values agree well with current measurements
3 (Sarmiento et al. 2006).

4 **Discussion**

5 *Degradation at the sediment-water interface*

6 The constant fluxes from 90 to 172 m demonstrate a minor degradation within the anoxic
7 water column. In contrast, fluxes at 50 m are constantly lower, probably due to higher
8 microbial activities and oxic conditions during annual mixing. Lake Malawi traps (Pilska
9 2004) also demonstrated maximum fluxes during the dry and windy season, when upwelling
10 induces high algal production. BSi fluxes were similarly high during mixing and disappeared
11 in the rainy season. Carbonate fluxes were both characterized by precipitation events, but
12 were higher in Lake Kivu.

13 The fluxes comparison between sediment traps and cores (Table 3) approximate the
14 degradation at the sediment-water interface. Gross sedimentation is the average flux between
15 90 and 172 m over two years. Net sedimentation is the average of the carbonate layer for the
16 Ishungu core. These comparisons indicate that OM is the most degraded; 75 % for TN, 76 %
17 for TP and 83 % for TOC. Similar degradation ratios were reported for temperate lakes
18 (Bloesch et al. 1977; Lehmann et al. 2002). BSi (20%) is also partly re-dissolved.
19 Surprisingly, TIC gross sedimentation represents only 40% of the net sedimentation. As
20 carbonate sedimentation is characterized by short spikes (Fig. 7), our traps measurements
21 were not representative.

22 Comparison with internal loading of nutrients (Pasche et al. 2009) suggests, however,
23 that gross sedimentation is underestimated by 50 to 70 %. Indeed, primary production varies
24 significantly between years, and *in situ* measurements of chlorophyll a (J.-P. Descy,
25 unpublished data, 2009) suggest that primary production in 2006 and 2007 was lower than the

1 average for 2002 to 2005 (Sarmiento et al. 2006). Further, degradation within the traps
2 certainly occurred due to the long exposure time without poisoning. In steady state,
3 degradation at the sediment-water interface should be similar to internal loading (Table 3),
4 allowing the correction of gross sedimentation. The degradation within traps was estimated as
5 48% for TP, 68% for TN and 59 % for BSi. BSi degradation may rather reflect the
6 significantly lower primary production during the sampling years. Indeed, preservation in
7 Kivu's sediments is excellent (Haberyan and Hecky 1987), as expected in an anoxic
8 hypolimnion with low pH and high dissolved silica (Ryves et al. 2006). The high TP and TN
9 degradation is probably explained by the long exposure time and the high water temperature.
10 This long-recognized problem is often solved by poisoning traps. Degradation within traps
11 was found acceptable for temperate lakes with one to two weeks exposure time (Mudroch and
12 MacKnight 1991). In Lake Baikal, N, P and C fluxes were also corrected by a factor of 1.8 for
13 the degradation within traps (Müller et al. 2005). In tropical lakes, this decomposition is
14 certainly higher due to warmer water.

15 *Diatom ecology and paleoecology*

16 The sediments in Lake Kivu are dominated by *Nitzschia* species, specific to tropical or
17 African regions (Sarmiento et al. 2006), and since 1970s, *Chaetoceros muelleri* has become
18 significant. Similarly, *Chaetoceros* and *Nitzschia* have been present for the last 6000 years
19 (Haberyan and Hecky 1987). The present-day phytoplankton of Kivu is similar to Victoria
20 prior to its human-induced nutrient changes (Sarmiento et al. 2006). The *Nitzschia* group of
21 Kivu are good competitors at high Si: P ratio and are always dominant; centrics
22 (*Stephanodiscus*) that compete best at lower Si: P are rare (Sarmiento et al. 2006). However,
23 surveys (Hecky and Kling 1987; Sarmiento et al. 2007) and trap material from Ishungu (Fig.
24 9) did not record *Chaetoceros*, despite its importance in the upper Kibuye core (Fig. 6).

25 Several changes in the diatom assemblages were observed in the recent sediment record
26 from Lake Kivu. Their interpretation is complicated by the unusual water chemistry and

1 mixing regimes affecting Si:P ratio (Kilham et al. 1986). In Lake Kivu, shallow mixing
2 maintains high Si:P ratios in the epilimnion, while deeper mixing entrains more P and salts
3 lowering surface Si:P ratios and increasing salinity. Changes in DI-conductivity should
4 therefore be viewed with caution, as certain key taxa may be driven more by Si:P ratio than
5 salinity. The first major change occurs before 1850 with the disappearance of *Nitzschia* cf.
6 *marginata*, the reduction in *N. bacata* and the replacement by *Nitzschia fonticola* agg. and *N.*
7 *confinis* agg. The dominance of *Nitzschia* species in the earlier record are indicative of
8 increased lake stability with reduced mixing regime (Stager et al. 1997). *Nitzschia bacata* has
9 been shown to form near-surface populations under daily stratification (Sarmiento et al. 2006;
10 Stager et al. 2003; Stager et al. 2005).

11 Below ~30 cm, significant occurrence of benthic *Rhopalodia* (Haberyan and Hecky
12 1987) and *Chaetoceros* might indicate a lower lake level and closer near-shore habitats, with
13 deeper mixing, and reduced Si:P ratio. Above ~29 cm, declines in *Rhopalodia* and
14 *Chaetoceros* and *Nitzschia marginata* blooms suggest higher water level, more stable
15 stratification and shallower mixing, with high Si:P ratio, and may reflect the widespread
16 return to wetter conditions across East Africa during the 1800s (Stager et al. 2005, 2009;
17 Bessems et al. 2008). The replacement with *N. fonticola* agg. (~22 cm) may indicate
18 continuing high lake level, stable stratification and shallow mixing under N limitation.
19 Continued dominance of *N. fonticola* until nowadays suggest persisting conditions, with the
20 appearance of *Synedra ulna* at ~13 cm suggesting maximum Si:P ratio in the record and the
21 re-appearance of *Chaetoceros* at around 9 cm (1960s).

22 At ~25 cm, a short-lived perturbation appeared in the diatom record characterized by
23 low Si:P taxa (*Cyclostephanos dubius* and *Stephanodiscus parvus*), increased TA:D ratio,
24 high proportion of littoral diatoms, minor peaks in the saline taxa *Thalassiosira rudolfi* and
25 *Chaetoceros* spp. This is paralleled by increased TOC, TN and TP in the cores (Fig 3). This
26 combination tempts us to speculate that a sudden, short-lived deep water mixing event took

1 place, bringing saline water, with low Si:P, and that also disrupted near-shore sediments
2 providing a littoral signal to the core site.

3 A second major change in the diatom stratigraphy occurs c. 1970, coincident with the
4 onset of carbonates accumulation (Fig 3). This recent period is dominated by the planktonic,
5 freshwater *Nitzschia fonticola* and *N. lancettula*. Stager and colleagues (2005) report the latter
6 taxon in cores from Lake Victoria at times of higher lake levels when the lake is deep and
7 very fresh. The large increase in testate amoebae scales throughout this section is indicative of
8 increased catchment in-wash as recorded elsewhere in East Africa over this period (Mills
9 2009). The largest increase in the sedimentation rate occurs during this period, illustrating the
10 importance of in-wash from the catchment and marginal lake areas.

11 Between 1970 and 2006, there is also a substantial peak in *Chaetoceros*, probably
12 indicating higher salinity or changes in Si:P. *Chaetoceros* was found only in more saline lakes
13 (Gasse et al. 1995, Fritz et al. 1993) or coastal waters, and *Chaetoceros muelleri* has a high
14 conductivity optimum within the EDDI training set. Their dominance in mid-Holocene
15 sediments from Kivu implied surface salinity of 3 ppt (Haberyan and Hecky 1987). However,
16 salinity profiles from the 1970s are similar from today (Tietze 1978), and high lake level
17 argue against a significant salinity increase, especially at the inferred $4000 \mu\text{S cm}^{-1}$. However,
18 in the Baltic, increases *Chaetoceros* are often indicative of anthropogenic nutrient enrichment
19 (Stachura-Suchoples et al. 1998). We propose that *Chaetoceros* reflects slightly increased
20 salinity, but is rather an indicator of low Si:P, similarly to *Stephanodiscus/Cyclostephanos* in
21 fresh conditions. Lowering surface Si:P ratios may be induced by recent nutrient inputs from
22 the catchment, and increased upwelling of saline, low Si:P deep waters.

23 *Major changes in sedimentation during the last 40 years*

24 The most sudden change is the onset of carbonate precipitation in the early 1960s. Calcite was
25 nearly absent in the sediment before and its accumulation suddenly increased by 15 fold. In
26 the last 40 years, fluxes slightly increased for TOC (10%) and TN (60%) but more than

1 doubled for TP (Table 2). Total mineral accumulation increased by a factor of 3.5 and positive
2 trends were observed for soils tracers or elements co-precipitating with carbonates.

3 The sudden onset of carbonates is probably induced by higher primary productivity. In
4 Lake Kivu, the water above 60 m is oversaturated related to calcite and the saturated deep-
5 water prevents any re-dissolution (Pasche et al. 2009). Even if the water is oversaturated,
6 nucleation is necessary to start the precipitation process, often linked to phytoplankton
7 activity (Dittrich and Obst 2004; Stabel 1986). Carbonates in older sediment (Botz et al.
8 1988) were identified as primary precipitates (aragonite, monohydrocalcite) and diagenetic
9 products (siderite, calcite, and dolomite). Aragonites ($\delta^{13}\text{C} = 4$ to 6 ‰) precipitated (Botz et
10 al. 1988) in isotopic equilibrium with the lake water bicarbonate ($\delta^{13}\text{C} = 4.8 \text{ ‰}$). In
11 comparison, recent carbonates are isotopically heavier ($\delta^{13}\text{C} = 6.1$ to 7 ‰), which might
12 indicate more intense primary production.

13 Three major hypotheses could explain the abrupt changes around 1960: food web
14 modifications, increased external nutrient inputs, or more intense upwelling via subaquatic
15 sources.

16 *Top-down effects due to the introduction of *Limnothrissa miodon**

17 Changes could be caused by an increased top-down control after the introduction of
18 the first pelagic and planktivorous fish, *Limnothrissa miodon*. This non-native fish could exert
19 more pressure on the larger zooplankton, which would then loosen control over the
20 phytoplankton. Increased primary production has often triggered carbonate precipitation
21 (Hodell et al. 1998; Teranes and McKenzie 1999). In the epilimnion, the recycling and export
22 of nutrient could change due to the shift in zooplankton communities.

23 *Limnothrissa miodon* was introduced in Lake Kivu in 1958-1960. This new predator
24 eliminated the efficient grazer, *Daphnia curvirostris* (Dumont 1986), which was replaced by
25 less efficient cyclopoid copepods (Isumbisho et al. 2006). This reduced efficiency might have

1 favored high phytoplankton biomass (Sarmiento et al. 2006). Increased primary production is,
2 however, difficult to demonstrate, as growth depends on nutrient availability. Our sediment
3 core analyses rather suggest a change in nutrient recycling within the epilimnion than
4 enhanced export production. This recycling might result from the shift in zooplankton
5 dominance: *Daphnia* favours nutrient recycling (Bossard and Uehlinger 1993; Darchambeau
6 et al. 2005), while copepods export P through fecal pellets. Reduced pressure on smaller
7 pelagic diatom may explain increases in *Nitzschia lancettula* and *N. inconspicua* since the
8 1950s (Haberyan 1985). These food web changes may have occurred rapidly, which further
9 explain the carbonates onset around the time of *Limnothrissa*'s introduction.

10 *Bottom-up effects due to increased external nutrient inputs*

11 The increased erosion due to deforestation over the last decades has increased sediment
12 load to the lake, which would also explain the increase in periphytic diatoms and testate
13 amoebae washout, and abundant nucleation sites for carbonate precipitation. P sedimentation
14 could have been amplified via the flux of P-rich soil particles. Increased nutrient loading,
15 accentuated by direct inputs from the fast-growing population, could accelerate lake
16 productivity, alter Si:P ratio in upper waters, and affect both pH and the super-saturation of
17 CaCO_3 .

18 In Lake Kivu catchment, the population has steeply increased to 2.1 million over the
19 last decades (Muvundja et al. in press). Enhanced anthropogenic activities and the lack of
20 sewage treatment will have increased the external nutrient inputs to Lake Kivu. An increase in
21 nutrients would induce enhanced primary production, supported by the appearance in the last
22 40 years of *Chaetoceros muelleri*, linked to nutrient enrichment in saline systems (Stachura-
23 Suchoples et al. 1998). Unfortunately, no historic data are available for comparison. But
24 recent analyses (Muvundja et al. in press) demonstrated that external nutrient inputs
25 contribute only a minor part (15% bio-P, 35% DIN, 45% SRSi) to the total loading,

1 dominated by upwelling (Pasche et al. 2009). Even if the external nutrient loading has
2 increased, this anthropogenic influence may be small for the nutrient budget to the epilimnion.

3 Allochthonous material reaching Lake Kivu might have been enhanced by soil erosion
4 in the deforested catchment area. The Kibuye core reveals a three-fold increase of soil
5 minerals during the last 40 years. The larger TP fluxes might be explained by the extremely
6 high TP loads in rivers (Muvundja et al. in press). Similar catchment disturbance was also
7 reported for Lake Tanganyika, where sedimentation rates in deltas dramatically increased in
8 the early 1960s (McKee et al. 2005), due to extraordinary rainfalls in 1961-62. TOC fluxes
9 also increased in mid-20th century, driven by allochthonous OM and increased nearshore
10 productivity (Palacios-Fest et al. 2005). In Lake Malawi, anthropogenic impacts may have
11 increased external nutrient loading by 50% (Hecky et al. 2003) .

12 Population increase and the catchment deforestation has led to growing external nutrient
13 inputs, although these sources contributed a small fraction of the internal loading (Muvundja
14 et al. in press). In contrast, erosion was clearly responsible for the increase of adsorbed P and
15 soil minerals in near-shore sediment cores.

16 *Hydrological changes*

17 Hydrological changes in East Africa have been documented for several lakes (Nicholson and
18 Yin 2001; Ryner et al. 2008). In Lake Kivu, subaquatic springs contribute about 20% of total
19 water inputs and drive a slow upwelling (Schmid et al. 2005). An increase in the subaquatic
20 discharge could deliver more nutrients and Ca^{2+} in the epilimnion by upwelling, inducing
21 higher primary production, lower Si:P ratios, CaCO_3 precipitation and higher salinity.

22 The African Great lakes are sensitive to climatic changes and the water budgets of
23 Lakes Victoria and Tanganyika could reconstruct past climate. In the 20th century, these lakes
24 rose rapidly after the extreme rainfalls from 1961-1964 followed by high water level until
25 1990 (Nicholson 1999; Nicholson and Yin 2001). The water level of Lake Kivu was 0.44 m
26 higher during 1965-1993 compared to 1945-1960. Annual rainfall from meteorological

1 stations in Lake Kivu catchment increased by about 100 mm between 1932/61 and 1962/90
2 (Rwanda Meteorological Service), representing 20 % of the whole catchment discharge.
3 Supposing enhanced subaquatic discharge, more salts and Ca^{2+} would be delivered to the
4 surface due to a higher upwelling. These changes could induce higher primary production,
5 carbonate precipitation and a shift toward diatoms typical of lower Si:P, saline conditions. In
6 addition, a strengthening of the main chemocline has been observed since 1972, probably
7 resulting from increased discharge from subaquatic springs (Pasche et al. submitted).

8 The rise in lake level between 1960 and 1993 resulted from an increase in rainfall,
9 ultimately feeding the subaquatic sources. We propose that higher subaquatic discharges
10 enhanced the upwelling of nutrients and salts to the epilimnion, leading to higher primary
11 production and a shift towards saline diatoms. Together with more dynamic algal blooms
12 induced by the top-down mechanism and further enhanced by increased catchment P loading,
13 this could have triggered the intense CaCO_3 precipitation.

14 **Conclusions**

15 Gross sedimentation had a seasonal maximum coinciding with the primary production
16 peak in the dry season. In permanently stratified lakes, the seasonal deepening of the mixed
17 layer releases nutrients from their nutrient-rich deep waters. During the anoxic descent, OM
18 degradation was minor. On the contrary, mineralization at the sediment water interface
19 released 75 to 80% of the nutrients and organic carbon, and 18% of biogenic silica, back into
20 the water column.

21 Sediment cores revealed an onset of CaCO_3 accumulation 40 years ago, accompanied
22 by nutrients increases, and a shift towards more saline diatoms with lower Si:P requirements.
23 These changes probably resulted from ecosystem modifications, occurring around 1960. First,
24 the introduced *Limnothrissa Miodon* reduced the zooplankton communities, allowing
25 increased phytoplankton dynamics. Secondly, the stronger upwelling fed by enhanced
26 subaquatic springs delivered more nutrients to the surface water, sustaining a higher primary

1 production. Thirdly, the increasing population in the catchment might have induced
2 eutrophication. In conclusion, these environmental changes might have potentially triggered
3 the carbonate precipitation; however we are not able to determine which of these mechanisms
4 or which combination of mechanisms is the actual dominant cause.

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Tables

Table 1 Averaged fluxes of sediment components for the three sediment cores and molar ratio between TOC, TN and TP. The first layer contained the white laminae (white layer), which were absent from second unit (brown layer)

Core name	Sed. Rate	TIC	TOC	TN	TP	C:N	C:P	N:P
Depth range cm	$\text{g m}^{-2}\text{yr}^{-1}$	fluxes in $\text{g m}^{-2}\text{yr}^{-1}$				molar ratio		
Ishungu 0 - 10	226	5.92	7.2	0.83	0.136	15	164	11
Kibuye 0 - 8.5	212	8.41	12.8	0.60	0.108	25	300	12
Gisenyi 0 - 3.5	351	21.12	16.6	1.00	0.180	19	238	12
Avr. white layer	263	11.81	12.2	0.81	0.141	20	234	12
Ishungu 10 - 31.5	76	0.93	3.4	0.41	0.045	10	144	14
Kibuye 8.5 - 39	65	0.53	9.0	0.33	0.037	32	627	20
Gisenyi 3.5 - 43	125	1.74	17.8	0.73	0.094	28	538	19
Avr. brown layer	89	1.06	10.1	0.49	0.059	23	436	17
ratio white/brown	3.0	11	1.2	1.6	2.4	0.85	0.54	0.67

Table 2 Fluxes of nutrients, inorganic carbon and BSi fluxes averaged over two years at four depths (50, 90, 130 and 172 m), representing 17 samples per depth from Ishungu basin sediment traps. Molar ratios C_{org}: N, C_{org}: P and N: P are also presented. Values not measured are represented by a hyphen

Depth	Sed. rate	TIC	TOC	TP	TN	BSi	C:N	C:P	N:P
m	g m ⁻² yr ⁻¹					molar ratio			
50	347	1.4	38	0.33	2.9	-	16	305	20
90	422	1.9	40	0.47	3.2	6.1	13	205	16
130	462	2.4	41	0.51	3.1	-	12	177	15
172	496	2.3	43	0.55	3.1	-	12	177	15

Table 3 Degradation at the sediment / water interface determined by the comparison between the annual averaged flux in the sediment trap from 90 m to 172 m and the average of the white layer from the Ishungu core, taken at the same location. This degradation is compared with internal loading of nutrients (Pasche et al. 2009). For TOC, internal loading was the doubled flux of methane, to take into account the organic degradation in CH₄ and CO₂. Gross sedimentation was recalculated to balance the internal loading. These calculations allowed the determination of degradation within the sediment traps during exposure time

	Sed. rate	TOC	TP	TN	BSi
	g m ⁻² yr ⁻¹				
Gross sedimentation (avr. 90 to 172 m)	460	41	0.51	3.1	6.1
Net sedimentation (Ishungu core)	223	7	0.12	0.8	5.0
Degradation at the sediment water interface	237	34	0.38	2.4	1.1
Internal loading			0.86	8.9	10.0
Gross sedimentation required to balance internal loading			0.98	9.7	15.0
Degradation within sediment traps			0.48	6.5	8.9

Figures

Modify all the figure because traps before cores

Fig. 1 Pasche
add bathymetry, name of the basin and Ruzizi.

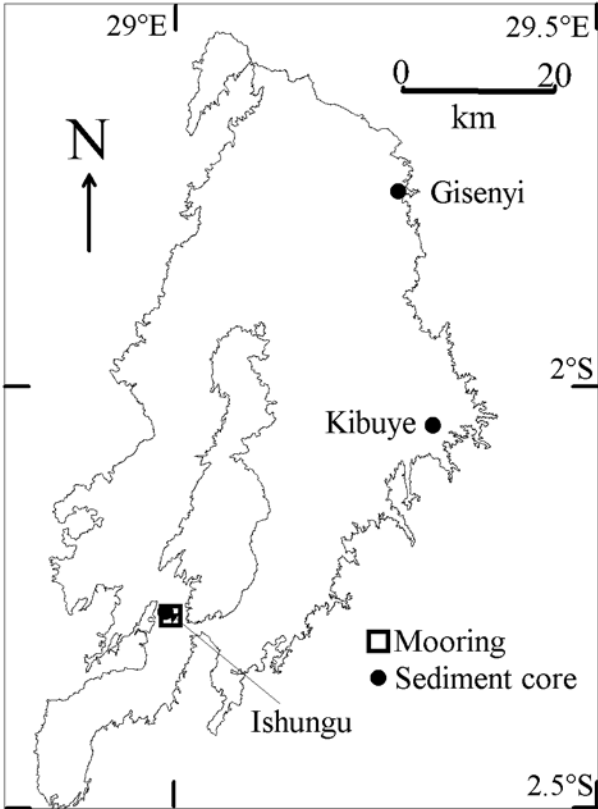


Fig. 2 Pasche

The age model needs to be shown as applied to each core, the activity vs. depth graph is of less interest. add switching depth to the y axis and showing two panels beside each other.

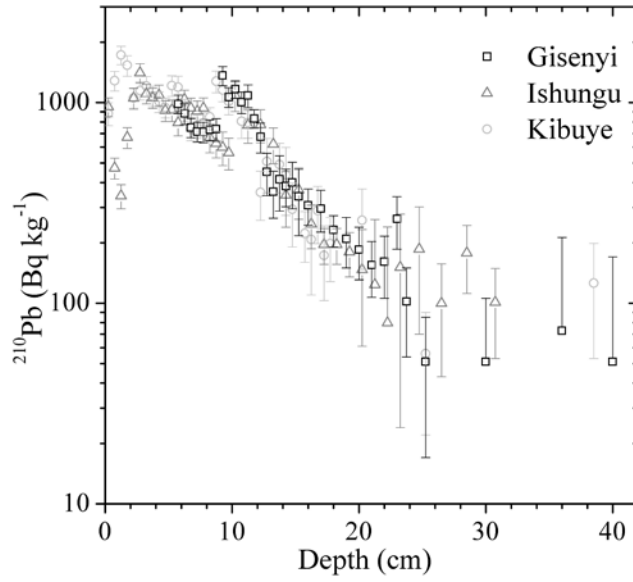


Fig. 3 Pasche

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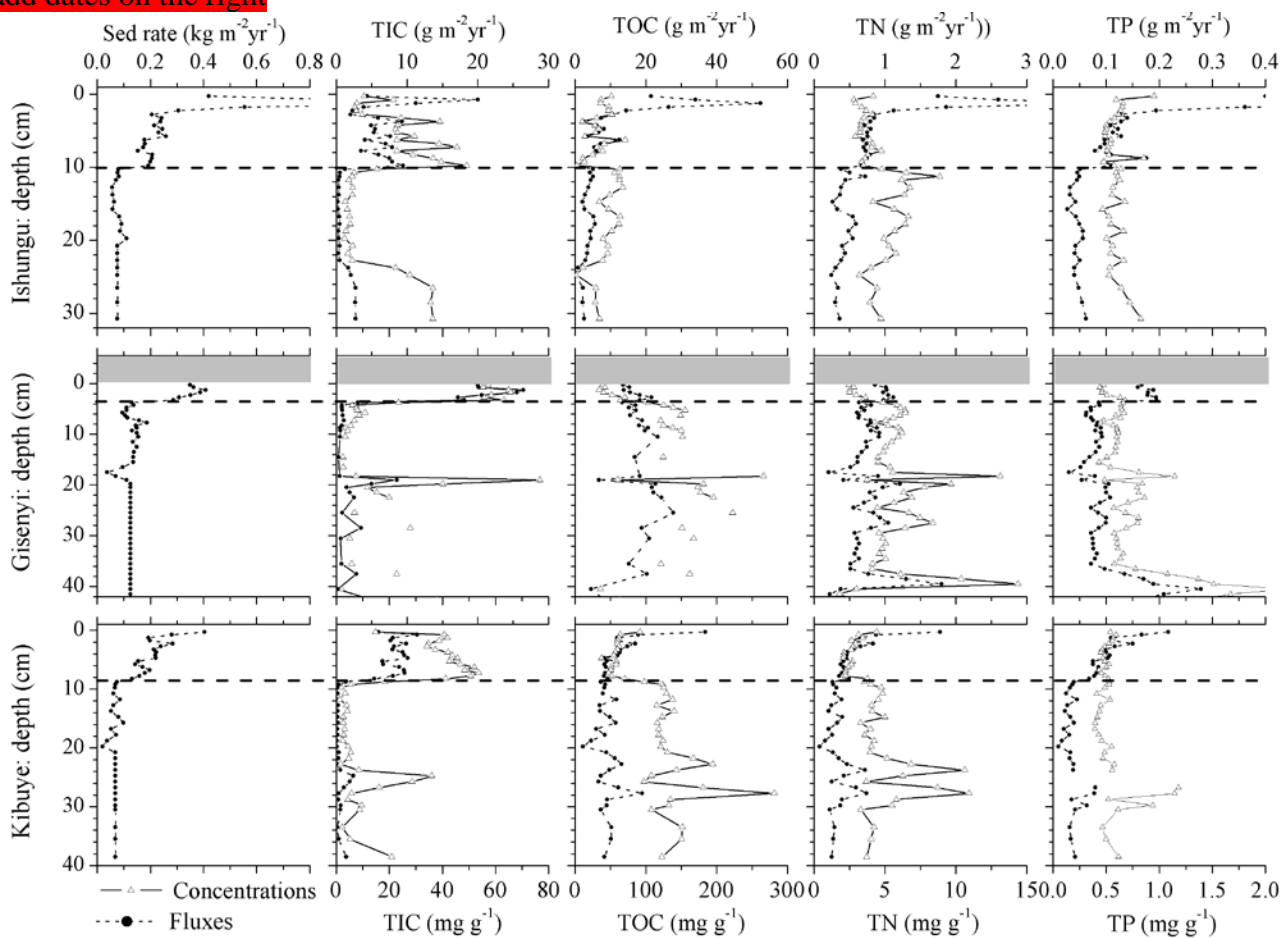


Fig. 4 Pasche
add dates on the right

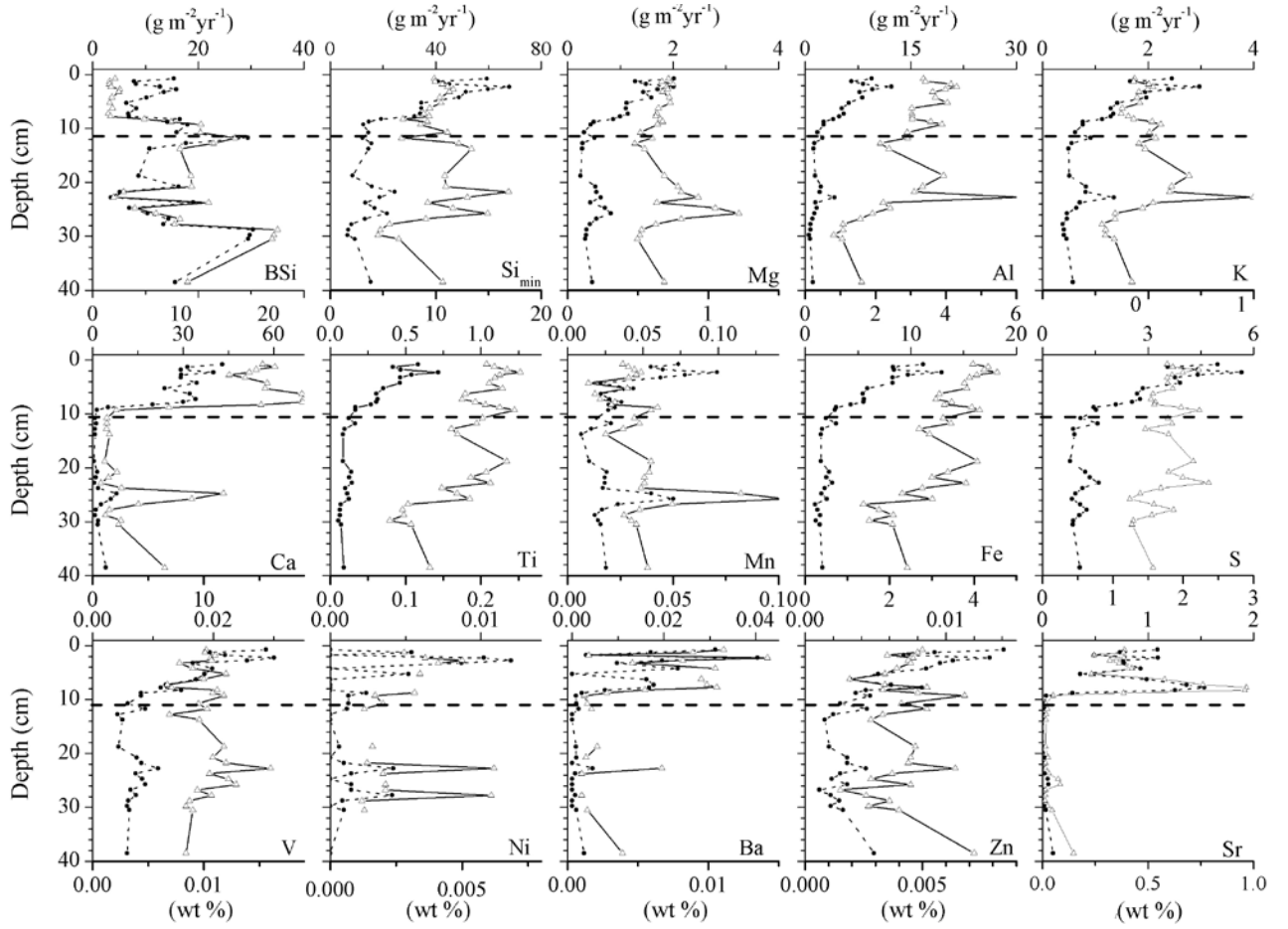
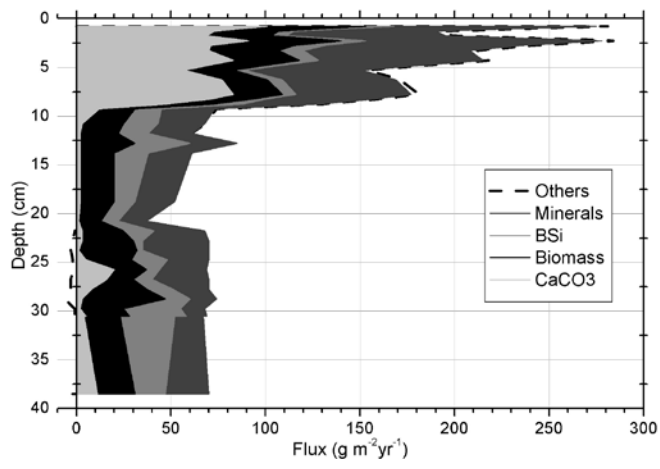


Fig. 5 Pasche



Improve legend, use squared in legend or color

Fig.6 Pasche

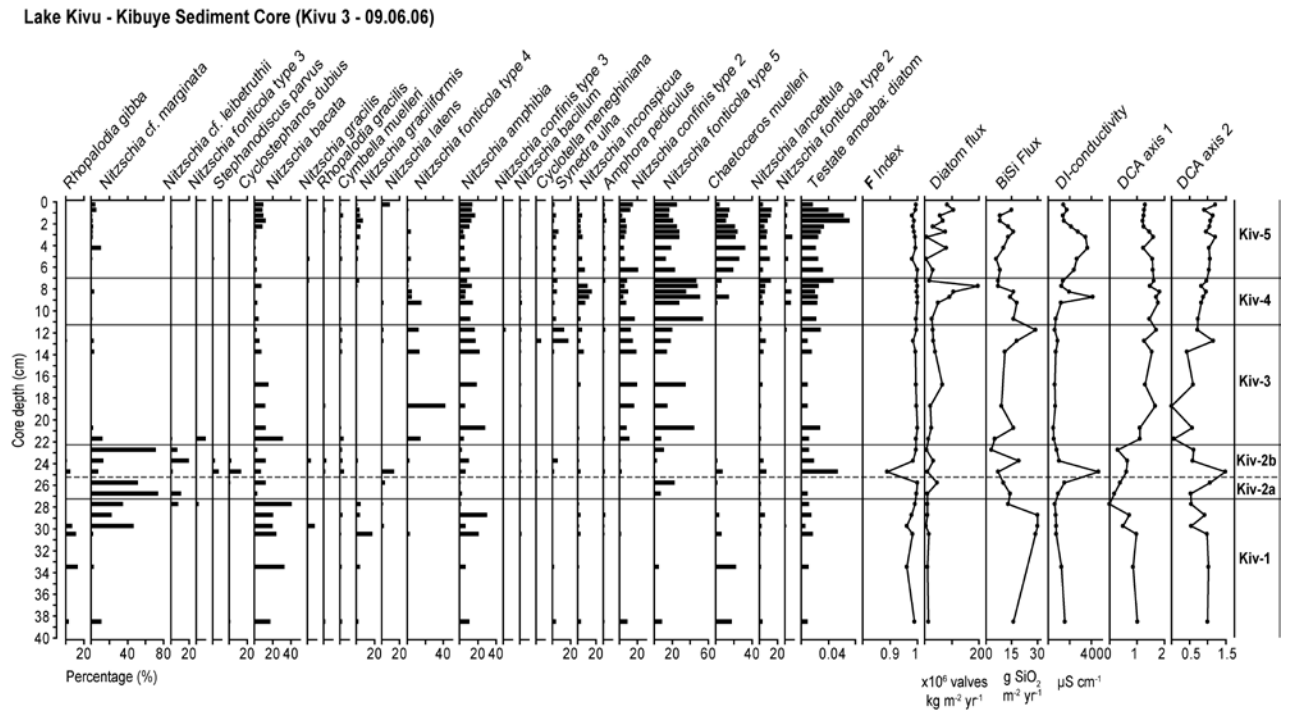


Fig. 7 Pasche

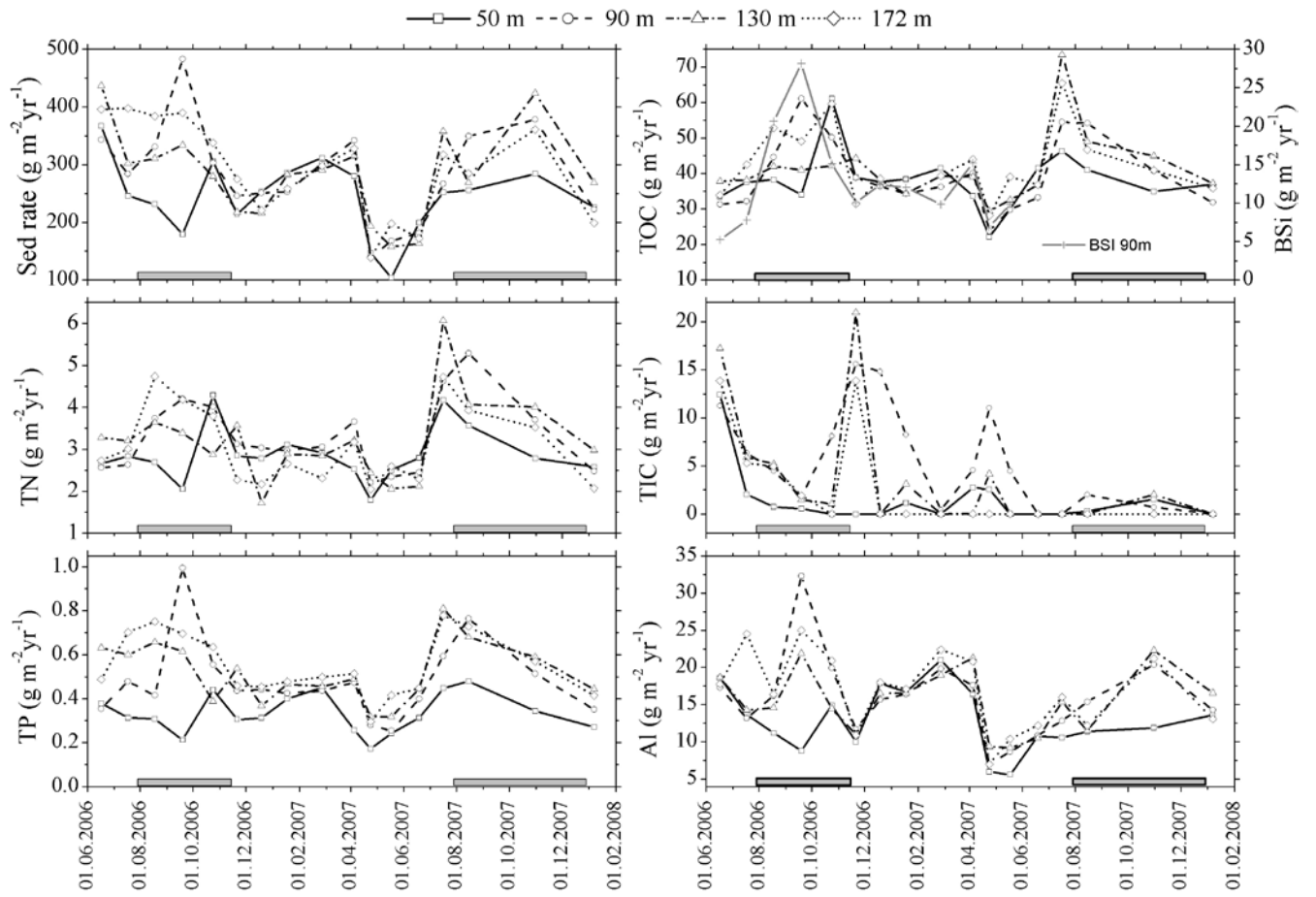


Fig. 8 Pasche

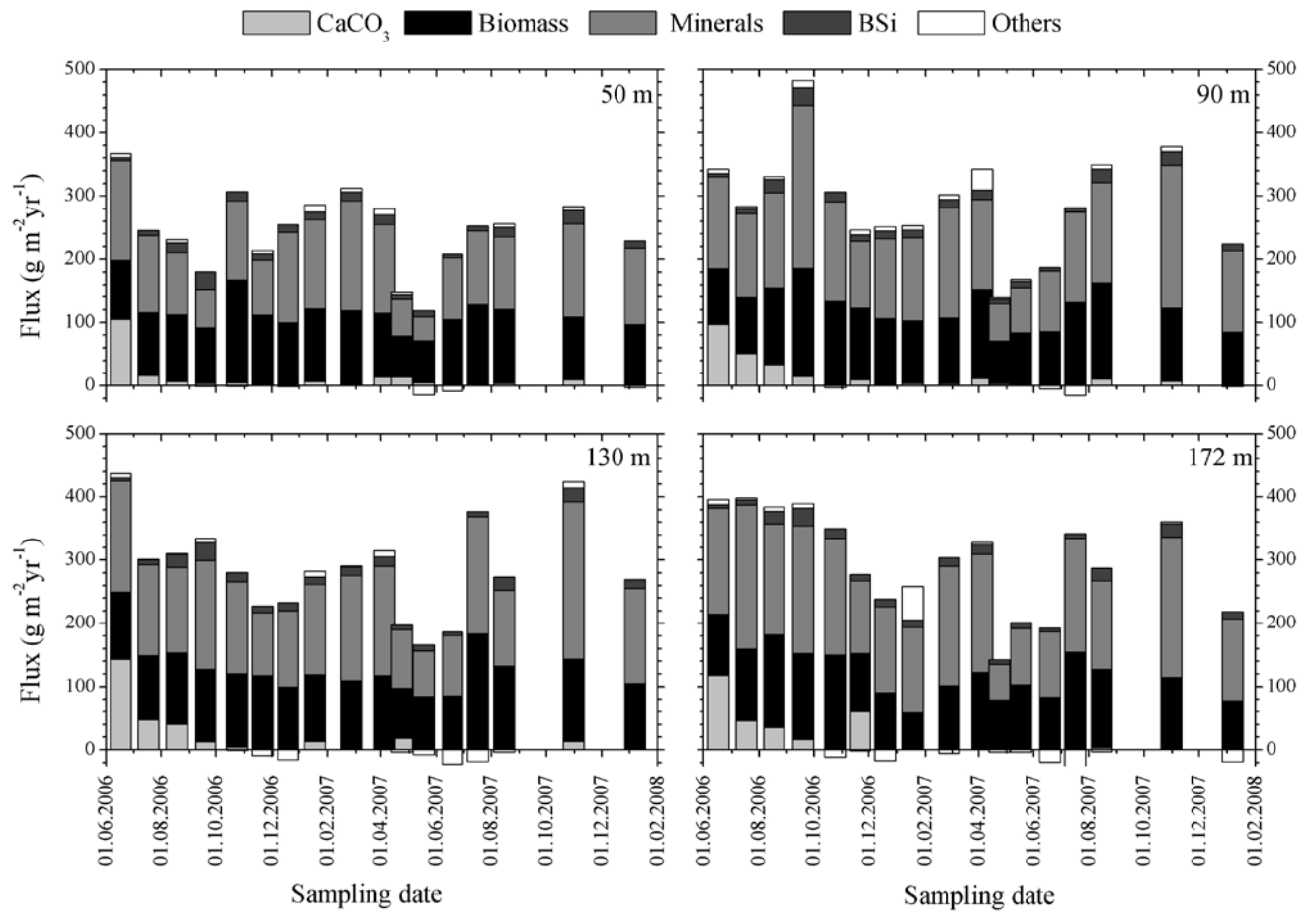


Fig. 9 Pasche

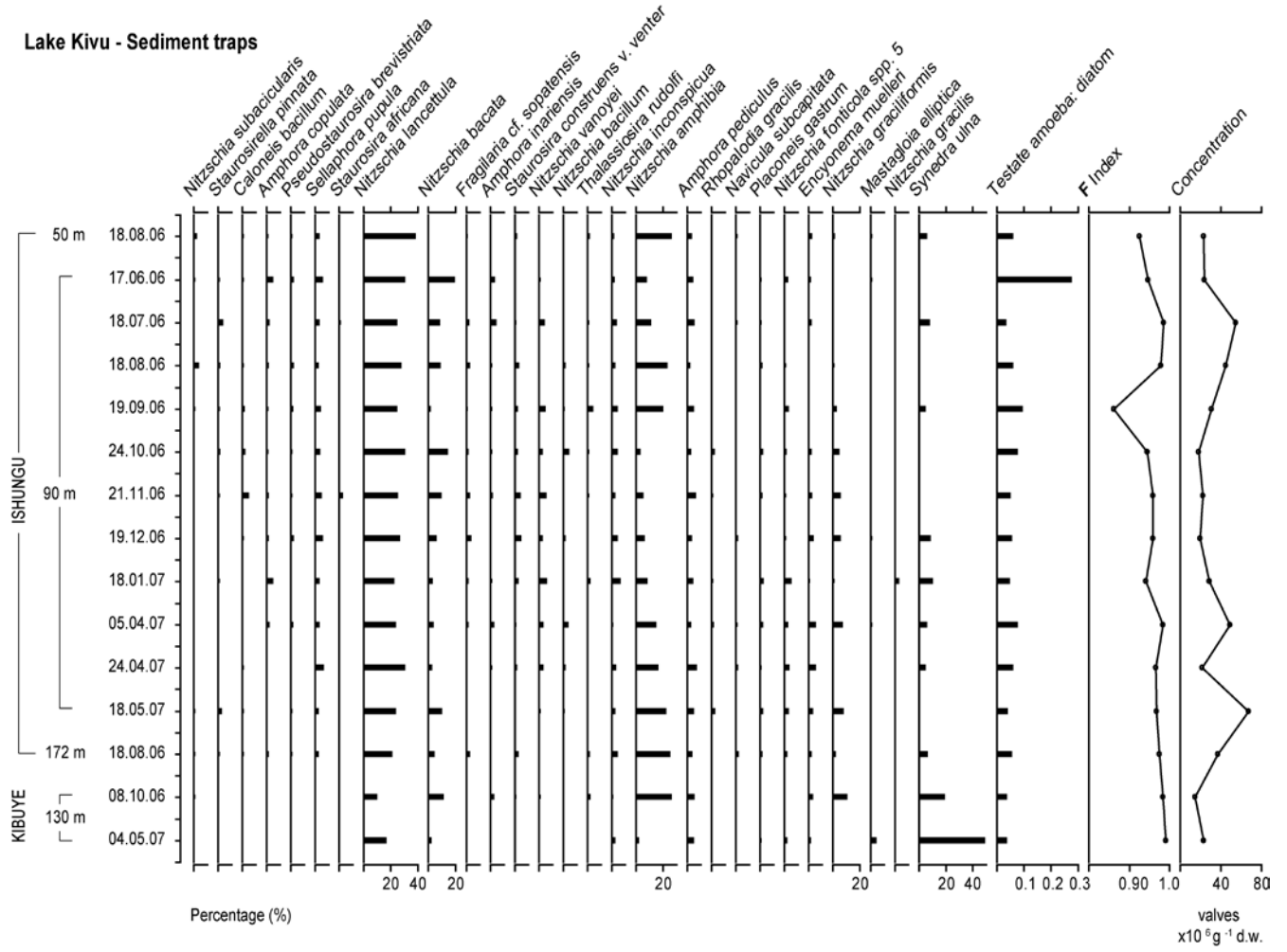


Fig. 10 Pasche

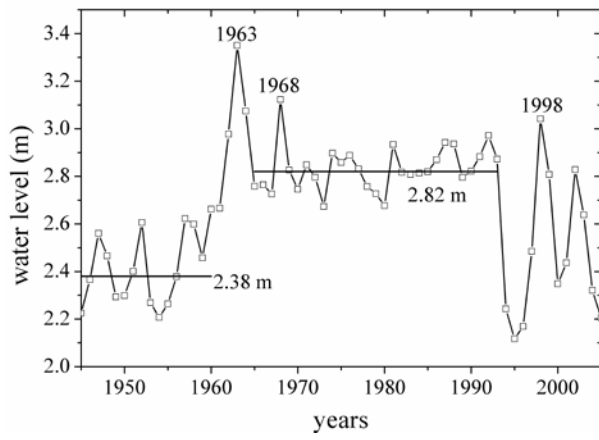


Figure captions

Fig. 1 Map of Lake Kivu showing the location of the sediment cores (circles) and the sediment traps (rectangle). The Kibuye sediment core (2°02.886'S, 29°18.307'E, depth of 190 m) and the Gisenyi sediment core (1°46.383'S, 29°15.763'E, depth of 155 m) were taken in the deep basin. In the Ishungu basin, the mooring with the sediment traps (02°16.255'S, 28°59.780'E) was fixed close to the coring site (2°16.077'S, 28°59.374'E, depth of 175 m)

Fig. 2 ²¹⁰Pb activities in three different sediment cores measured in Bq kg⁻¹. Error bars represent accuracy of each measurement. The data from the Gisenyi sediment core were shifted by 5.5 cm, assuming the upper part of the sediment core was lost during sampling

Fig. 3 Concentrations (continuous lines with triangles, mg g⁻¹), sedimentation rates (circle red, kg m⁻²yr⁻¹) and fluxes of nutrients (dashed line with circle, g m⁻²yr⁻¹) for the three different sediment cores. The dashed lines represent the lower limit of the layer containing white laminae (10 cm for Ishungu, 3.5 cm for Gisenyi and 8.5 cm for Kibuye sediment core)

Fig. 4 Concentrations (continuous lines with triangles) and fluxes of different elements (dashed lines with circles, g m⁻²yr⁻¹) measured by X-ray fluorescence for Kibuye sediment core. The dashed lines represent the lower limit of the layer containing white laminae (8.5 cm)

Fig. 5 Composition of Kibuye sediment core divided between carbonates, biomass, biogenic silica and minerals. The fraction “others” correspond to the rest in order to agree with the sedimentation rate

Fig. 6 Diatom stratigraphy from Lake Kivu. All species > 2% in any one sample are displayed. The diatom taxa have been ordered according to their weighted average abundance in the core (ascending) and are split into assemblage zones Kiv-1-5. The ratio of testate amoebae scales is displayed alongside the diatom preservation index (**F** index), diatom concentrations and the axis 1 and axis 2 sample scores from Detrended Correspondance Analysis (DCA). Reconstructed conductivities are based on the combined African dataset for salinity in EDDI

Fig. 7 Sedimentation rates, fluxes of nutrients (TOC, TP, TN) and carbonates measured in the four sediment traps (50, 90, 130 and 172 m) on Ishungu mooring. Biogenic Si at 90 m is illustrated in the TOC graph (black crosses), and Al at 90 m is represented in TIC graph (black crosses). The grey bars represent periods of annual mixing where the oxycline was below 50 m

Fig. 8 Composition of sediment traps at 50 m, 90 m, 130 m and 172 m on Ishungu mooring divided between carbonates (ligh grey), biomass (black), biogenic silica (grey) and minerals (dark grey). The fraction “others” (white) correspond to the rest in order to agree with the sedimentation rate

Fig. 9 Diatom assemblages of Lake Kivu sediment traps. All species > 2% in any one sample are displayed. The diatom taxa have been ordered according to their weighted average abundance in the core (ascending). The ratio of testate amoebae scales is displayed alongside the diatom preservation index (**F** index), diatom concentrations and the axis 1 and axis 2 sample scores from Detrended Correspondance Analysis (DCA).

Fig. 10 Water level measured at Ruzizi I hydropower station. The average from 1945 to 1960 and 1965 to 1993 are represented as a horizontal black line. Extreme peaks in water level were recorded in 1963, 1968 and 1993