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EVALUATING THE RADIOCARBON RESERVOIR EFFECT IN LAKE KUTUBU, PAPUA NEW GUINEA

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ABSTRACT. We examined the radiocarbon (^{14}C) reservoir effect in Lake Kutubu using tephrochronology and terrestrial plant material to deliver a precise age-depth profile and sedimentation rates for this lake. Based on the presence of two tephra horizons (Tibito and Olgaboli), we found a reservoir age offset in sediments of between 1490 and 2280 ^{14}C yr using the sediment ages derived from the lead-210 (^{210}Pb) dating method. The live submerged biological samples collected exhibited a higher reservoir age offset than the sediment. This is most likely a result of delayed transport of "bomb" ^{14}C from the atmosphere to aquatic and sedimentary system. The ^{14}C reservoir effect increased with distance from the lake inlet and also decreased with depth. Dissolution of ^{14}C-depleted carbon from surrounding limestone and direct in-wash of old soil or vegetation remnants from the catchment are the most likely causes of the ^{14}C reservoir effect. Based on limestone areas mapped in Papua New Guinea, we indicate lakes which may be subject to a significant ^{14}C reservoir effect. The results of this study demonstrate the magnitude of the ^{14}C reservoir effect in lakes and provide insights to the correct interpretation of past environmental and archaeological events in PNG.

KEYWORDS: ^{14}C, limestone, old carbon, plant, sediment.

INTRODUCTION

High-quality chronologies associated with sediment records have the potential to elucidate decadal to century-scale paleoenvironmental changes and have a pivotal role in providing the chronological context for many fields of environmental sciences and archaeology (Santos et al. 2007; Grimm et al. 2009). Sedimentary records retrieved from lakes, in particular, hold invaluable information for assessing past ecosystems and climates (Last and Smol 2002; Yang et al. 2008; Schneider et al. 2016). Nevertheless, constructing age-depth models based solely on bulk sediments can be highly problematic (Deevey 1954; Broecker and Walton 1959; Hall and Henderson 2001; Hendy and Hall 2006; Grimm et al. 2009; Soulet 2015), and even targeted organic debris such as seeds, leaves, and pollen can be subject to contamination from older and/or younger carbon (Olsson 1986; Wohlfarth et al. 1998; Fletcher et al. 2017). Low pollen concentrations, poor preservation and potential for reworking may preclude pollen separates being used as an alternative dating option (cf. bulk sediments) (Thorley 1981; Harle et al. 1999).

Here we highlight the occurrence of old carbon within sediments of Lake Kutubu in Papua New Guinea (PNG), a phenomenon known as "radiocarbon (^{14}C) reservoir effect." We discuss

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methods to overcome this issue and provide guidance for future sedimentary studies from this and other lakes within the limestone-dominated Papuan Fold Belt that require chronological definition and evaluation.

The reservoir effect occurs because the ordinary procedure of ^{14}C dating assumes that the carbon, before entering the material under investigation, has achieved isotopic exchange equilibrium with the CO_2 of the air (Deevey 1954). This assumption may not be valid in some cases. For example, hard-waters derived from old limestone that does not contain any ^{14}C may cause the reservoir effect in materials formed in such water. Colloidal $CaCO_3$ may provide a reservoir of carbon of low ^{14}C activity even in water that is in equilibrium with the atmosphere (Ohle 1952; Deevey 1954). Decomposition of old organic material (e.g. peat) may also supply old carbon/bicarbonate to lakes (Bengtsson and Törneman 2004).

Dating sediment from archaeological sites in limestone catchments in PNG has proven to be problematic. ^{14}C ages on calcite flowstones associated with archaeological and palaeontological material excavated from Nombe Rockshelter in the highlands of PNG (Gillieson and Mountain 1983) yielded results estimated to be at least 2000–5000 yr too old due to the reservoir effect. A recent highland wide assessment of archaeozoological records has shown that dating uncertainties, most likely introduced by a reservoir effect, hinder detailed temporal analysis of faunal assemblages (Sutton et al. 2009).

In the freshwater lakes within limestone catchments of PNG, the impact of the reservoir effect on ^{14}C dates has received only limited attention in the literature. In a PhD thesis (Garrett-Jones 1979) a sediment record from Lake Yanamugi, a small lake in the Markham Valley west of Lae, contained evidence of a volcanic ash of known age (Tibito Tephra, at that time thought to date ~1700 AD) that had ^{14}C dates ranging from 380 to 1730 yr older than they should have been when calibrated against Tibito Tephra. Oldfield (1977), who has worked on lakes in PNG, also notes that the error introduced by the photosynthesis of aquatic organisms in hard-water lakes is well documented (e.g. Deevey et al. 1954). This phenomenon of "apparent age" is of major importance in evaluating ^{14}C age determinations based on material wholly or partly subaqueous in origin. Oldfield (1977) reports that attempts to correct for this "hard-water" effect have mainly been assessing the difference between real and expected ^{14}C activity for contemporary surface sediment, and then applying this correction factor to each date down a profile. But although this method provides a conformable sequence of dates, the assumptions upon which it rests have never been fully justified nor have independent age determinations on ^{14}C dates "corrected" in this way confirmed their accuracy.

The present study tackles Oldfield's last point and, using techniques vastly improved since 1977, examines independent age determinations as a means of applying corrections to dates affected by old carbon. We now have a much better understanding of the utility of multiple dated tephra layers that provide isochronous horizons wherever they might be in PNG. Using tephrochronology we are able to validate ^{14}C ages and allow for the correction of age offsets caused by the reservoir effect.

Here we examine two cores from Lake Kutubu in the Southern Highlands of PNG (Figure 1) to provide a precise age-depth profile and sedimentation rates for the lake. Based on the presence of two widespread thin (~1 cm) distal tephra layers (Olgaboli and Tibito), which have been ^{14}C dated at many coastal and highland localities and in different sedimentary settings (Blong et al. 1982; Haberle 1998; Coulter et al. 2009; Blong et al. 2017b; Schneider et al. 2017), we are able to compare these previously acquired ^{14}C ages with those obtained from Lake Kutubu where old

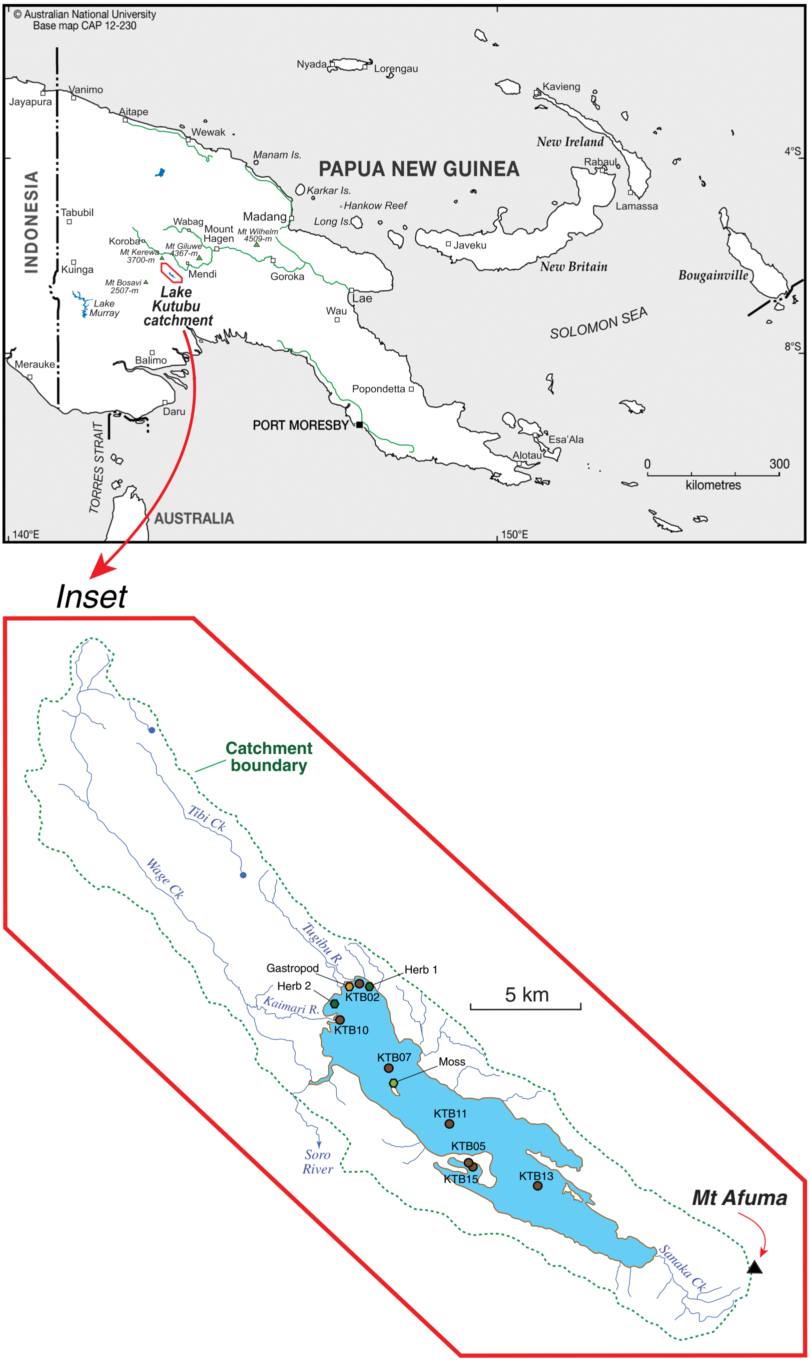

Figure 1 Map of Papua New Guinea and adjacent offshore islands. The red inset denotes the location of Lake Kutubu within the highlands. The inset map of Lake Kutubu shows the location of sediment cores and other samples used in this study. Note that the largest inlets and the outlet are confined in the northwest portion of the lake. Blockage of the catchment and formation of Lake Kutubu has been attributed to Plio-Pleistocene volcanism associated with Mt Afuma at the southeast end of Lake Kutubu (Bayly et al. 1970). (Please see online version for color figures.)

carbon appears to have influenced the ^{14}C ages obtained. Finally, based on geological maps, we indicate those PNG lakes most likely to be affected by the ^{14}C reservoir effect to alert researchers to the likely need to correct ^{14}C dates from these lakes.

METHODS

Location

Lake Kutubu (latitude 06°25.79′S, longitude 143°20.22′E, altitude 808 m; Figure 1) in the Southern Highlands Province of PNG has been recognized as one of the most pristine freshwater lakes in the whole of the Asia-Pacific region (D'cruz 2008). Lake Kutubu is approximately 19 km long and 4 km wide and is flanked by high hills along its length. The lake is fed by a series of small creeks and subterranean rivers that flow through and within the surrounding limestone and karst ranges (Osborne and Totome 1992; WWF 2008). The lake has an estimated area of about 50 km^2 and a volume of 1.825 km^3 (D'cruz 2008). Approximately 20 km^2 of wet land reed-beds and swamp forest is found on the lake shore. Establishing definite boundaries to

Figure 2 (A) Southwest margin of Lake Kutubu looking towards Mt Afuma ridge; (B) Local dugout canoe on Lake Kutubu with remains of non-biting midges (Chironomids) on the surface of the lake and limestone geology exposed in the background; (C) Aerial view of the central area of Lake Kutubu. Photos from Simon Haberle 1992.

the catchment is difficult because of the complex karst terrain, but the total catchment area is about 260 km^2, which is covered mainly by tropical rainforest (D'cruz 2008) (Figure 2).

Physical and Chemical Characteristics

The climate in the Lake Kutubu catchment is warm and wet, with little variation throughout the year. The mean annual temperature is 23°C, with a minimum recorded temperature of 5°C, and maximum of 39°C. The mean annual rainfall is 4500 mm, with rain being recorded throughout the year (D'cruz 2008).

Lake Kutubu is an oligomictic lake with a maximum depth of ~60 m and is characterized by distinct thermal stratification with irregular periods of mixing (Osborne 2012). Mixing events may be triggered when abnormally cold and stormy weather occurs, resulting in mixing of the deoxygenated layers (hypolimnion) with the oxygenated layers (epilimnion) (Osborne and Totome 1992).

Two mixing events have been reported for Lake Kutubu. There is anecdotal evidence of mixing in 1960 (Osborne and Totome 1992). A second mixing event in 1990 was studied in detail by Osborne and Totome (1992). This latter event, driven by cool temperatures and strong southeast winds, caused vertical mixing and overturning in the lake towards the middle of September 1990 (Osborne and Totome 1992).

During the second mixing event in Lake Kutubu, anoxic water bubbles were released to the surface at the southeastern end of the lake and the lake water became cloudy with iron particles. At the other end of the lake, dissolved oxygen was delivered to depth (Osborne 2012). Prolonged stratification and irregular periods of mixing have pronounced effects on the spatial distribution of elements in the water column and the sediments (Osborne and Totome 1992). This process is likely to add to the potential for old carbon to be incorporated into bulk sediment samples derived from the lake deposits.

The lake was formed when the southeastern end of the valley was blocked off by lava flow from Mount Afuma (Bayly et al. 1970) in the late Pliocene to Pleistocene (Brown and Robinson 1982). The upland areas of the catchment are a dissected terrain of Darai Limestone (Upper Oligocene to Middle Miocene) with minor interbedded mudstones and sandstones (Brown and Robinson 1982). The limestone surface is dominated by numerous sinkholes. Along its length the lake is flanked by karst landforms; it was the first subterranean karst wetland type to be added to the Ramsar Classification System by Resolution VI.5 (Ramsar site no. 961). Karst (cave) wetland systems are connected to underground rivers and act as recharge areas when the surrounding water table is low, and as discharge areas when it is high.

Lake Kutubu has high chemical concentrations relative to other lakes in PNG (Bayly et al. 1970), with total cations ranging from 1.02–1.99 meq/L, total anions from 1.48–2.11 meq/L and conductivities from 110–182 μS/cm. Because it receives water from limestone areas, its water has a high alkalinity of 48–94 mg/L $CaCO_3$ (Chambers 1987), with a pH of 8.1 (Osborne and Totome 1992).

The waters of Lake Kutubu are usually clear (Secchi disc depths of 7.0–8.3; Osborne and Totome 1992). This high water clarity is due to the usually low nutrient status of the epilimnetic waters, which results from the low phosphorus and nitrogen loadings from the predominantly limestone and sparsely populated catchment area (Osborne and Totome 1992).

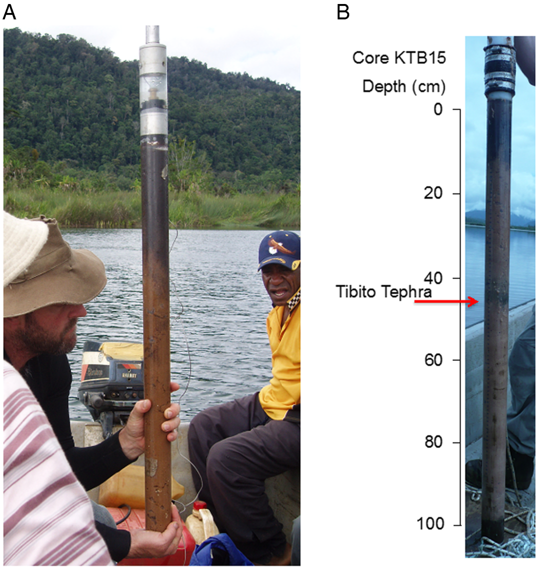

Figure 3 Coring the mud-water interface, Lake Kutubu, PNG. (A) Core collected on the shallow margins (<2 m water depth) of central-southern shore of Lake Kutubu showing carbonate sediments (lighter sediments). (B) Core KTB15 showing the universal coring device retrieving ~95 cm of sediment. The arrow shows Tibito Tephra at 45 cm depth in the core. Photos from Simon Haberle 2007.

Sediment Core Collection

In this study, multiple mud-water interface sediment cores were retrieved from Lake Kutubu to assess paleo-environmental baselines. Fieldwork was conducted between 2007 and 2009 with the logistical support of the World Wildlife Fund (PNG) and Oil Search Ltd. (PNG). Sediment cores were obtained from seven sites: KTB-02, KTB-05, KTB-07, KTB-10, KTB-11, KTB-13 and KTB-15 (Figure 1 inset and Figure 3). These sites were chosen on the basis of their positions relative to river outlet and inlets. Cores of ca. 1 m length were extracted using a universal coring system with polycarbonate tubes (Aquatic Research Instruments). After retrieval, cores were subsampled at 1 cm intervals and placed in airtight zip-lock plastic bags for transport from the field to the Australian National University. Sediment samples were then stored at 4°C at the Archaeology and Natural History cool-store quarantine facilities at the Australian National University.

The cores extend along a northwest to southeast transect (core KTB-02, -10, -07, -11, -05, -15 and -13 respectively; Figure 1). The sedimentation rate of each core is related to its position relative to river inlets and outlet in the northwest end of Lake Kutubu. Cores KTB-2 and KTB-10 are close to the Tugibi and Kaimari inlet rivers, respectively. Both rivers have small deltas, and high rates of sediment influx occur in their vicinity. For this reason, they could be precisely dated using ^{210}Pb because of high deposition rates and young sediments. In contrast, cores KTB-07, -05, -15, -11 and -13 retrieved in the central and eastern portions of the lake, and well away from present-day points of river inflow, contain the most complete sedimentary records with two macroscopic (~1 cm) tephra beds (Tibito and Olgaboli) showing clearly visible upper and lower contacts (Figure 3).

Although well-positioned in relation to sedimentation rates to offer ^{14}C dates, cores KTB-05 and -15 were not used in this study to calculate the reservoir effect because of the predominance of *Chara* spp. growing at the coring site. *Chara* spp. is a multicellular green algae that resembles land plants because of stem-like and leaf-like structures (Berg et al. 1999). They are found in fresh water, particularly in limestone areas where they grow submerged, attached to the muddy bottom. They prefer less oxygenated and hard water and are covered with calcium carbonate deposits (Berg et al. 1999).

Cores KTB-05 and -15, in the littoral zone where *Chara* spp. grows in abundance, are subject to the addition of organic matter derived from floating vegetative mats at the lake surface and present a more complex dating environment than those in the centre of the lake. Accordingly, only cores KTB-07, -11 and -13 were used for calculations of the reservoir effect because these are the most reliable in relation to the continuity of the sediment profiles.

In addition to the sediment cores, three plant samples were collected, two herbaceous plants (Cyperaceae) unidentified sedges (Herb 1 and Herb 2) growing at the edge of the lake (emergent), and one moss sample (Moss) growing above the waterline of the lake on an exposed rock outcrop (Figure 1). The moss sample is not expected to have received any source of carbon from Lake Kutubu. One gastropod snail shell (*Melanoides sp.*), still alive at the time of core collection, was retrieved from top of core KTB-2 to check for the reservoir effect on modern shells.

Lead-210 Dating

Lead-210 (^{210}Pb) samples were processed at the Australian Nuclear Science and Technology Organisation (ANSTO), following methods described by Harrison et al. (2003). The alpha spectrometry method, which requires only 2 g of sample, was chosen. Each dried sediment sample was spiked with polonium-209 (^{209}Po) and barium-133 (^{133}Ba) tracers. Each sample was then leached with hot concentrated acids to release polonium and radium. Polonium was autoplated onto silver disks after adding the reducing agent hydroxylammonium chloride. Radium and barium were isolated by co-precipitation and collected as colloidal microprecipitates of barium sulphate on fine membrane filter papers. The activities of ^{210}Po on the silver disks and ^{226}Ra on the membrane filters were determined by alpha spectrometry. Each membrane filter was also counted by gamma spectrometry to measure the ^{133}Ba tracer activity. Chemical yield recoveries of ^{210}Po and ^{226}Ra were calculated using the recoveries of ^{209}Po and ^{133}Ba tracers, respectively. Unsupported ^{210}Pb activities between 4 and 17 cm depth exhibit a decreasing trend with depth. The top 3 cm is more disturbed than the lower part of the core, which may be caused by disturbance at the sediment/water interface or a more rapid sedimentation rate. Using unsupported ^{210}Pb data between 3 and 17 cm depth for the Constant Initial Concentration (CIC) model, and 0–17 cm depth for the Constant Rate of Supply (CRS) model (Appleby and Oldfield. 1978) sediment ages and mass accumulation rates were calculated. The CIC model is suitable for dating only sediment cores with a monotonic unsupported ^{210}Pb profile against depth, and thus calculates only a single mass accumulation rate over the whole core. The CRS model is the preferred model for dating sediment cores with varying mass accumulation rates, or non-monotonic unsupported ^{210}Pb profiles. Lead-210 results from Lake Kutubu are presented in Table 1.

Caesium Dating

Caesium analyses were carried out at ANSTO. Between 1 and 2 g of dried and ground whole sediment were analyzed for caesium-137 (^{137}Cs) activity by gamma-ray spectrometry using a

Table 1 Summary of ^{210}Pb age data collected from Lake Kutubu sediment core KTB-07. The CIC mass accumulation rate was calculated to be 0.059 ± 0.003 gcm^{-1}yr^{-1}.

ANSTO ID	Core	Depth (cm)	Total ^{210}Pb (Bq/kg)	Supported ^{210}Pb (Bq/kg)	Unsupported ^{210}Pb (Bq/kg)	CIC ages (year)	Age (years AD)
P868	***KTB 07***	0–1	1007 ± 63	103 ± 8	904 ± 64	2 ± 2	2007
P869	***KTB 07***	2–3	964 ± 45	149 ± 12	815 ± 47	12 ± 2	1995
P870	***KTB 07***	4–5	1150 ± 65	79 ± 6	1071 ± 65	21 ± 3	1986
P871	***KTB 07***	6–7	875 ± 42	129 ± 10	746 ± 43	33 ± 3	1974
P872	***KTB 07***	8–9	610 ± 25	172 ± 14	438 ± 29	48 ± 4	1959
P873	***KTB 07***	10–11	463 ± 19	147 ± 11	316 ± 23	63 ± 4	1944
P874	***KTB 07***	12–13	415 ± 18	196 ± 15	219 ± 24	79 ± 5	1928
Q160	***KTB 07***	16–17	164 ± 7	118 ± 9	46 ± 12	119 ± 7	1888

High Purity Germanium (HPGe) detector. The detector system energy calibration was carried out using a National Institute of Standards and Technology (NIST) traceable multi-nuclide standard source and the detector system efficiency calibration was determined using IAEA reference materials including RGU-1, RGTh-1, RGK-1 and Soil-6. ^{137}Cs was analyzed to validate the ^{210}Pb chronology. Fallout from the atmospheric testing of nuclear weapons, such as ^{137}Cs, accumulates in sediment materials and can be used to validate ^{210}Pb chronologies. The horizon with the maximum activity of ^{137}Cs should indicate the year 1963. ^{137}Cs results from Lake Kutubu are presented in Table 2.

Radiocarbon Dating

Samples were analyzed for ^{14}C using bulk sediment. Ages were obtained by accelerator mass spectrometry at DirectAMS (Washington, USA), NOSAMS Woods Hole Oceanographic Institution (Massachusetts, USA) and at the Research School of Earth Sciences at the Australian National University. The dried, crushed samples were pre-treated with HCl–NaOH–HCl to remove carbonates, humic and fulvic acids and washed with milli-Q, and the insoluble residue was freeze-dried for AMS analysis (Fallon et al. 2010). All samples were normalized to the AMS δ^{13}C and background subtracted using ^{14}C free coal.

^{14}C ages were calibrated using the northern hemisphere terrestrial curve IntCal13 ^{14}C calibration data set (Reimer et al. 2013) in the Bayesian age depth modelling R programme Bacon (Blaauw and Christen 2011). We have opted to use the Northern Hemisphere curves as PNG lies very close to the January inter-tropical convergence zone, which is probably strongly influenced by northern air masses (McKee et al. 2015; Blong et al. 2017b). We also consider that the Southern Hemisphere calibration curve SHCal13 (Hogg et al. 2013) is less relevant to our age conversions because this calibration curve is dominated by tree ring chronologies from New Zealand that are more strongly influenced by upwelling in the Southern Ocean (McCormac et al. 2004).

Reservoir Effect Calculation

The presence of two closely spaced tephra beds within Lake Kutubu sediments provided an ideal opportunity to calculate the reservoir effect in this large oligomictic lake system. These tephras, Tibito and Olgaboli, have been widely recognized and dated in a variety of depositional settings throughout the PNG highlands (Blong 1982; Haberle 1998; Blong et al. 2017b; Schneider et al. 2017). Both tephra beds can be distinguished by their major and trace element glass shard compositions and therefore represent important inter-regional chronostratigraphic marker horizons (Schneider et al. 2017).

To correct ^{14}C dates derived from bulk-sediment that may have been contaminated with old carbon, we calculated the reservoir offset by associating the ^{14}C age with ^{210}Pb ages and mean ages for tephra beds extracted from the same depth interval. The mineralogical and geochemical details of these tephras were previously studied to determine their sources and refine their chronology based on past geological and palaeoenvironmental events (Haberle 1998; Coulter et al. 2009; Schneider et al. 2017; Blong et al. 2016, 2017a, 2017b). Tibito tephra dates ca. 305–270 cal BP (2σ) (Blong et al. 2017b) and Olgaboli ca. 1180–980 cal BP (2σ) (Schneider et al. 2017).

In this study, the ^{14}C age of each tephra bed was determined from bulk sediment samples collected immediately above and below its upper and lower contact (Figure 3), supported by the overall age/depth profile (Dugmore et al. 2006). One outlier was detected in core KTB07 and this is removed from the age-depth model and reservoir age offset calculations.

Three cores were used for tephra pairing (see sediment core collection above for more details): KTB-07, -11 and -13. In core KTB-07 it was possible to retrieve only Tibito tephra as the sedimentation rate was too high because of its close proximity to the lake inlets (Figure 1). The high sedimentation rate of core KTB-07 meant it could be dated using only ^{210}Pb as this method is only able to date a horizon less than approximately 200 yr old.

The uncorrected ^{14}C age for each tephra was calculated using the midpoint between the two bracket ages for each tephra (Table 3). Dating the sediment above and below a tephra is important to accurately date an ash fall. In addition, given that the ages of these tephras are known, they can be used as a control to calculate the off-set age of the sediments.

In dating both above and below the tephra layers, we note that, because Lake Kutubu sediments are mainly of an anoxic nature, biological bioturbation is unlikely to have occurred in the layers dated in this study. We also note that the preservation of tephra layers only ~1 cm thick suggests minimal disturbance by either physical or biological agencies.

For Tibito tephra in cores KTB07 and KTB11, only one ^{14}C date adjacent to the tephra was available. In these two cases, the single ^{14}C age is used, immediately below the tephra deposit in KTB07 and immediately above the tephra deposit in KTB11.

In addition to the bulk-sediment method, living biological samples were collected in surface layers of sediment (Herb 1, Herb 2, Moss and Gastropod) and used to calculate the reservoir age offset. The Moss sample, collected above the waterline of the lake in an exposed rock outcrop, was used as a control. Terrestrial plants take up CO_2 directly from the atmosphere and their carbon-isotopic composition is expected to have been in equilibrium with contemporary atmospheric CO_2 concentration and thus not prone to ^{14}C age offsets.

All reservoir offset ages were calculated using the R package ResAge (Soulet 2015) run in the free open-source statistical software *R* (R Development Core Team 2017). The calibration curve used to calculate the reservoir age offsets was IntCal13 (Reimer et al. 2013).

The age-depth models in this study were built using the R package Clam (Blaauw 2010).

Table 2 ^{137}Cs activities from Lake Kutubu sediment core KTB-07, showing the maximum activity between 7 and 9 cm depth, validating the ages calculated from the CIC ^{210}Pb chronology.

ANSTO ID	Depth (cm)	^{137}Cs (Bq/kg)
Q442	3–4	3.19 ± 0.37
Q443	4–5	1.73 ± 0.83
Q444	5–6	5.76 ± 0.51
Q445	7–8	9.57 ± 0.5
Q446	8–9	9.94 ± 0.88
Q447	9–10	4.13 ± 0.39
Q448	11–12	0.39 ± 0.24
Q449	13–14	<1.04

RESULTS AND DISCUSSION

Dating Results and Reservoir Age Offsets

^{210}Pb dating results for core sediments collected in Lake Kutubu are shown in Table 1. As the ^{137}Cs results support the constant initial concentration (CIC) model (Table 2), only CIC ages were considered to calculate the reservoir effect in Lake Kutubu. As in many other limestone-sediment records, the ^{210}Pb dates in core KTB07 do not correlate with ^{14}C dates.

^{14}C calibrated ages for sediments and modern plant and shell samples are presented in Table 3. The stratigraphic distribution of ^{14}C dates in Lake Kutubu sediments did not correspond to the known tephra ages. This age offset, the reservoir effect, is demonstrated in Figure 4.

Calculated reservoir values (in ^{14}C yr) and associated uncertainties (1σ) are shown in Table 4. The reservoir mean value for core samples varied between 1490 and 2280 ^{14}C yr.

The moss sample, which was growing above the waterline of the lake on an exposed rock outcrop, did not display a ^{14}C reservoir effect as it receives rainwater in equilibrium with atmospheric CO_2. The submerged biological samples had a reservoir value between 2220 ± 37 and 2270 ± 34 ^{14}C yr (Table 5). This result agrees with a previous study showing that submerged aquatic plants and animals living on sediments of lakes with ^{14}C reservoir effect metabolize the ^{14}C-depleted C from the environment, becoming markedly depleted in ^{14}C compared to organisms utilising atmospheric CO_2 as their source (Deevey and Stuiver 1964).

The modern submerged biological samples presented a much higher offset reservoir age (2220–2270, $\bar{x} = 2250 \pm 41$ ^{14}C yr) than those calculated in sediments for the period before AD 1950 (1490–2280, $\bar{x} = 1900 \pm 286$ ^{14}C). Delayed transport of bomb ^{14}C from the atmosphere to aquatic and sedimentary systems is almost certainly the reason for the higher R values for recent time (Srdoč 1986).

Effect of Depth and Distance From Lake's Inlet on the Reservoir Effect

Core depth and distance from the inlet both had an effect on the freshwater reservoir effect in sediments of Lake Kutubu. The freshwater reservoir effect increased with distance from the inlet, with core KTB07 having the lowest reservoir offset (1490–1820 ^{14}C yr), KTB11 having an intermediary offset (1530–2200 ^{14}C yr) and KTB13 having the highest offset (1800–2360 ^{14}C yr) (Figure 5). This is explained by the hydrodynamics of Lake Kutubu where only one small part

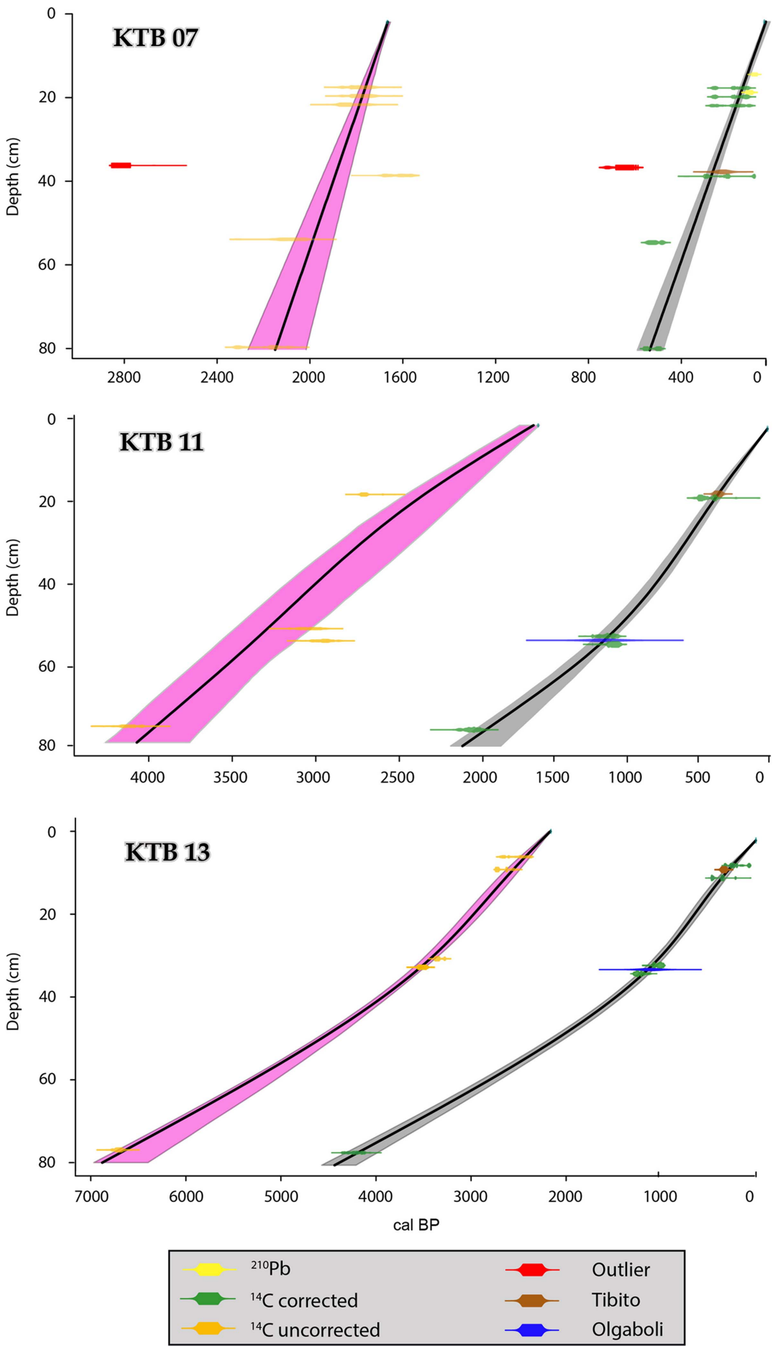

Figure 4 Age-depth model of cores KTB 07 (linear regression), KTB11 and KTB13 (smooth spline). In grey is the age-depth model corrected and in pink is the age-depth model not corrected for the ^{14}C ages reservoir effect.

Table 3 Radiocarbon sample and age data collected from Lake Kutubu sediment cores.

											Calendar age BP			Calendar age BP			Calendar age BP			Calendar age BP		
Sample	Laboratory	Lab ID	Core ID	Depth (cm)	$\delta^{13}C$	±	$F^{14}C$	±	^{14}C age	±	from	to	% prob	from	to	% prob	from	to	% prob	from	to	% prob
Sediment	SANU	49106	***KTB 07***	15–16	-30.5	0.3	0.7941	0.0022	1852	27	1864	1841	9.7	1837	1718	85.1						
Sediment	SANU	49107	***KTB 07***	17–18	-31.3	0.3	0.7950	0.0022	1843	28	1863	1842	5.9	1831	1711	88.9						
Sediment	SANU	49109	***KTB 07***	19–20	-30.0	0.3	0.7913	0.0022	1881	28	1734	1882	95.4									
Sediment	SANU	49110	***KTB 07***	33–34	-30.3	0.3	0.7200	0.0028	2639	36	2819	2843	5.8	2799	2726	89.1						
Sediment	SANU	49111	***KTB 07***	35–36	-30.4	0.3	0.8078	0.0024	1715	29	1641	1698	35.5	1639	1558	59.3						
Sediment	DirectAMS	OZN142	***KTB 07***	50–51	-31.9	0.1	0.7711	0.0030	2090	35	2151	1986	93.5	1979	1970	0.8	1960	1951	1.0			
Sediment	DirectAMS	OZN143	***KTB 07***	74–75	-32.0	0.1	0.7650	0.0022	2150	25	2303	2242	29.5	2179	2167	2.4	2163	2052	63.1			
Sediment	SANU	49113	***KTB 11***	17–18	-31.4	0.3	0.7230	0.0023	2606	30	2917	2771	95.4									
Sediment	SANU	49114	***KTB 11***	50–51	-31.6	0.3	0.6950	0.0021	2923	29	3160	2975	95.4									
Sediment	SANU	49115	***KTB 11***	53–54	-33.1	0.3	0.6999	0.0021	2867	29	3073	2880	95.4									
Sediment	DirectAMS	OZN145	***KTB 11***	74–75	-34.7	0.1	0.6233	0.0024	3795	35	4346	4334	0.7	4295	4082	93.4	4029	4010	1.3			
Sediment	SANU	45414	***KTB 13***	6–7	-40.6	0.3	0.7376	0.0019	2445	26	2700	2631	26.3	2618	2585	9.8	2575	2563	1.4	2541	2359	57.9
Sediment	SANU	45413	***KTB 13***	9–10	-40.3	0.3	0.7281	0.0016	2549	23	2750	2697	68.0	2634	2616	8.1	2590	2536	15.4	2530	2505	4
Sediment	SANU	45416	***KTB 13***	30–31	-39.5	0.3	0.6781	0.0015	3120	23	3468	3379	95.4									
Sediment	SANU	45412	***KTB 13***	32–33	-38.1	0.3	0.6645	0.0015	3284	23	3564	3457	95.4									
Sediment	DirectAMS	OZN146	***KTB 13***	75–76	-34.3	0.2	0.4806	0.0024	5885	40	6825	6820	0.3	6797	6630	94.1	6584	6569	1.0			
Sediment	SANU	45421	***KTB 15***	0–2	-25.5	0.3	0.4783	0.0023	5924	43	6880	6871	1.1	6860	6658	94.3						
Sediment	SANU	51820	***KTB 15***	30–31	1.1	0.3	0.7373	0.0023	2448	30	2702	2631	26.1	2619	2560	14.5	2543	2360	54.8			
Sediment	SANU	51819	***KTB 15***	70–71	0.1	0.3	0.6999	0.0022	2866	30	3074	2878	95.4									
Sediment	SANU	51818	***KTB 15***	538–539	3.5	0.3	0.4004	0.0017	7353	38	8306	8240	14.3	8219	8034	81.1						
Herb 1	SANU	45224	***Edge of lake***	Surface		-29.4	0.3	0.7853	0.0021	1941	31	1970	1960	1.6	1951	1821	93.8					
Herb 2	SANU	45225	***Edge of lake***	Surface		-22.9	0.3	0.7906	0.0019	1887	33	1895	1727	95.4								
Moss	SANU	45409	***Edge of lake***	Surface		-26.3	0.3	1.0424	0.0022	modern												
Gastropod	SANU	51821	***KTB 02***	Surface		-5.0	0.3	0.7827	0.0024	1968	30	1993	1865	95.4								

SANU: Radiocarbon Laboratory at the Australian National University. Canberra, ACT. Australia.
DirectAMS: DirectAMS commercial laboratory. Seattle, Washington, USA.

Table 4 Reservoir calculation (R) of sediments from Lake Kutubu, PNG.

Tephras/ Pb	Core	Depth (cm)	^{14}C age (mean ± 1σ; yr BP)	Tephra/Pb (yr BP)	Reservoir min (95% CI)	Reservoir max (95% CI)	Reservoir (mean ± 1σ; ^{14}C yr)
210Pb	KTB 07	12–13 (CIC)		19 ± 5			
	KTB 07	15–16 (CIC)	1852 ± 27	46 ± 7	1705	1824	1764 ± 31
	KTB 07	16–17 (CIC)		59 ± 7			
Tibito	KTB 07	34–35	1715 ± 29	289 ± 10	1490	1616	1494 ± 67
	KTB 07	35–36	1715 ± 29				
Tibito	KTB 11	17–18	2337 ± 30				
	KTB 11	18–19	2337 ± 30	289 ± 10	2000	2200	2100 ± 65
Olgaboli	KTB 11	50–51	2923 ± 29				
	KTB 11	51–52	2895 ± 29	1080 ± 100	1538	1874	1748 ± 80
	KTB 11	53–54	2867 ± 29				
Tibito	KTB 13	6–7	2445 ± 26				
	KTB 13	7–8	2497 ± 25	289 ± 10	2205	2358	2282 ± 63
	KTB 13	9–10	2549 ± 23				
Olgaboli	KTB 13	30–31	3120 ± 23				
	KTB 13	31–32	3202 ± 23	1080 ± 100	1800	2280	2040 ± 77
	KTB 13	32–33	3284 ± 23				

of the lake (northwest side, Figure 1) is reliably flushed and the water leaving (through the northwest side as well) is largely epilimnetic (Osborne 2012). Creek water probably enters the lake at depth depositing dissolved and suspended materials directly into the hypolimnion (Osborne and Totome 1992) and most likely brings outside carbon source to the northwest side of the lake.

The reservoir age offset decreased with increasing depth for the three cores (Figure 5). Unfortunately, the number of data points used for the reservoir calculation is too limited to allow statistical analyses quantifying the effect of depth and distance from lake's inlet on the reservoir effect in this lake.

The distance and depth effects shown in Figure 5 are difficult to interpret. Statistical analyses of the results do not allow definite conclusions, mainly because there are several factors causing the reservoir effect. Figure 5, however, does show a clear increase in reservoir age with distance northwest to southeast, and with core depth, which is indicative of age. There is also river input at depth (hypolimnion), and increased gas bubbling and iron precipitation in the southeast in response to strong cold southeast winds as probable initiator of overturning circulation. Figure 5 fits these circumstances, but speculation about these factors is premature and would require more data to be collected.

Changes and environmental events have been reported elsewhere as influencing the reservoir effect and causing a within-depth difference in reservoir years (Blaauw et al. 2011). In Lake Kutubu, enhanced sedimentation may have resulted from an intensification of agricultural activity in the Kutubu catchment around the time of the fall of Tibito Tephra, possibly as a result of the arrival of the South American sweet potato in the PNG highlands (Bayliss-Smith et al. 2017). However, while such changes occurred elsewhere in the highlands, we have no evidence that they occurred in the Kutubu catchment. In any case, human disturbance in the catchment seems to have been very minor until the last few decades (Schneider et al. 2015).

Table 5 Reservoir calculation (R) of plants and gastropod from Lake Kutubu, PNG.

Aquatic	^{14}C age (mean ± 1σ; yr BP)	F^{14}C (1σ) terrestrial moss	^{14}C age (yr BP) terrestrial moss	Reservoir (mean ± 1σ)
Herb 1	1941 ± 31	1.0424 ± 0.0022	−334 ± 17	2275 ± 35
Herb 2	1887 ± 33	1.0424 ± 0.0022	−334 ± 17	2221 ± 37
Gastropod	1968 ± 30	1.0424 ± 0.0022	−334 ± 17	2302 ± 34

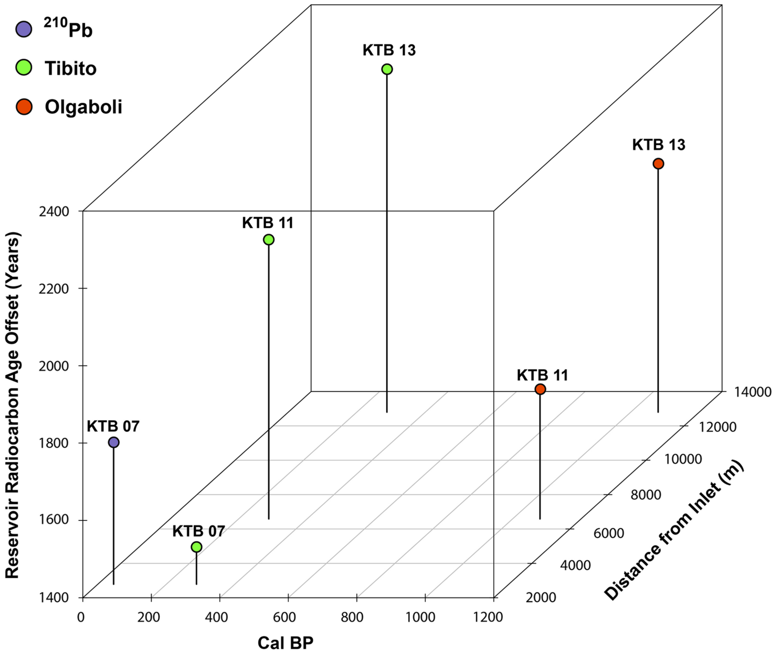

Figure 5 3D scatterplot of the reservoir effect showing the effect of distance from the lake inlets (m) and depth of sampling points indicated by ages (cal BP).

These results demonstrate that it is not advisable to use one core to extrapolate dating results for the entire lake. It is necessary to validate ^{14}C dates with other proxies at different sites and depths to understand the hydrodynamic and environmental effects on sediments of lakes that are suspected of a freshwater reservoir effect. That other factors may change reservoir ages over time is important, and suggests that the primary ^{14}C data are much more precise, and should be used rather than the averages.

Source of Old ^{14}C in Lake Kutubu

There are multiple possibilities for the contamination of ^{14}C-dated bulk-organic matter with carbon older than the time of deposition (Bjork and Wohlfarth 2001; Blaauw et al. 2011): (i) groundwater input containing ^{14}C-depleted CO_2 (e.g., from leaching old soils or volcanic activity), (ii) dissolution of ^{14}C-depleted carbon from surrounding carbonate-rich bedrock or old lake deposits (e.g., the limestone occurring locally), (iii) emanation of ^{14}C-depleted biogenic gas (CO_2 or CH_4) from organic bottom deposits followed by CH_4 oxidation at the oxycline, (iv) direct in-wash of old soil or vegetation remnants from the catchment, or (v) reworking of organic material from older lake-sediment horizons through bioturbation or a resuspension event.

Process (i) is known to happen in Lake Kutubu as the nature of the limestone substrate in this area allows groundwater to travel to the lake. The lake is an exposure of the regional groundwater table, and the surrounding limestone is highly porous. D'cruz (2008) mentions seasonal changes of water level of over a meter which are probably, at least in part, a consequence of fluctuations in the regional groundwater table. There is no information on the location of groundwater drainage divides. This underground water connection was demonstrated in a previous study that detected barium (Ba) within Lake Kutubu sediments from oil field drilling activities located outside of Lake Kutubu's surface catchment (Schneider et al. 2016). Barium concentrations in the northwest side of the lake followed a clear temporal connection with the start of mining exploration drilling activities outside of Kutubu catchment (Schneider et al. 2016). No other sources of anthropogenic inputs of Ba to the lake are known.

Process (ii) is most likely to happen in Lake Kutubu, as it is in a karst area consisting of limestone plateaus and broad ridges separated by narrow corridors formed in clastic sedimentary rocks. Process (ii) is further supported by the mean annual rainfall of 4500 mm in this lake. When rain permeates through limestone, dissolved (soil) CO_2 forms carbonic acid (H_2CO_3) that can dissolve carbonates. As it infiltrates cracks or crevices it dissolves the bedrock through a complex chemical reaction that results in the influx of old carbon to the lake (Ford and Williams 2007).

Process (iii), although it is not certain how significantly, occurs in Lake Kutubu during mixing events. Gas emissions are quite strong at the southeastern end of the lake. The lake water releases bubbles with gases likely to be a combination of CO_2, CH_4 and H_2S (P. Osborne, personal communication, 4 January 2018). We could speculate that climate dependent overturning circulation influences the reservoir age distribution in Lake Kutubu, but with only 2 overturning events recorded it is not possible to assess this.

Process (iv), as for process (ii), is supported by the lake geology and high precipitation. The limestone catchment promotes the uptake of old carbon by plants and the high precipitation supports the washing of old soil and vegetation remnants from the catchment to the lake. In addition, turbidite flows from the edge of lake slopes can also cause reservoir effects by washing old carbon into lakes (Blaauw et al. 2011; Karlin and Abella 1996), but no evidence has been yet recorded for this in Lake Kutubu. It is, however, a common process reported in large lakes (Blaauw et al. 2011) and is likely to happen in Lake Kutubu because of its steep basin and rapid inputs of sediments at the northwestern end, as shown by the small deltas.

Process (v) is believed to have contributed to the reservoir effect of Lake Kutubu at some level, but in an inverse role. As an oligomictic lake, Kutubu has a distinct stable thermal stratification where water at the bottom of the lake is prevented from mixing thoroughly with the atmosphere (Osborne and Totome 1992). The residence time of these stratified waters, therefore, may also amplify the ^{14}C reservoir effect in Lake Kutubu, as described by Hall and Henderson (2001). This process could be one of the causes of the depth effect on the reservoir effect.

Although we have not conducted chemical analyses of the lake water, a previous study has shown that alkalinity and hardness in waters of Lake Kutubu are a result of ^{14}C-depleted limestone in the catchment area (Osborne and Totome 1992), thus supporting processes (ii) and (iv) as main contributors to the recorded ^{14}C reservoir effect.

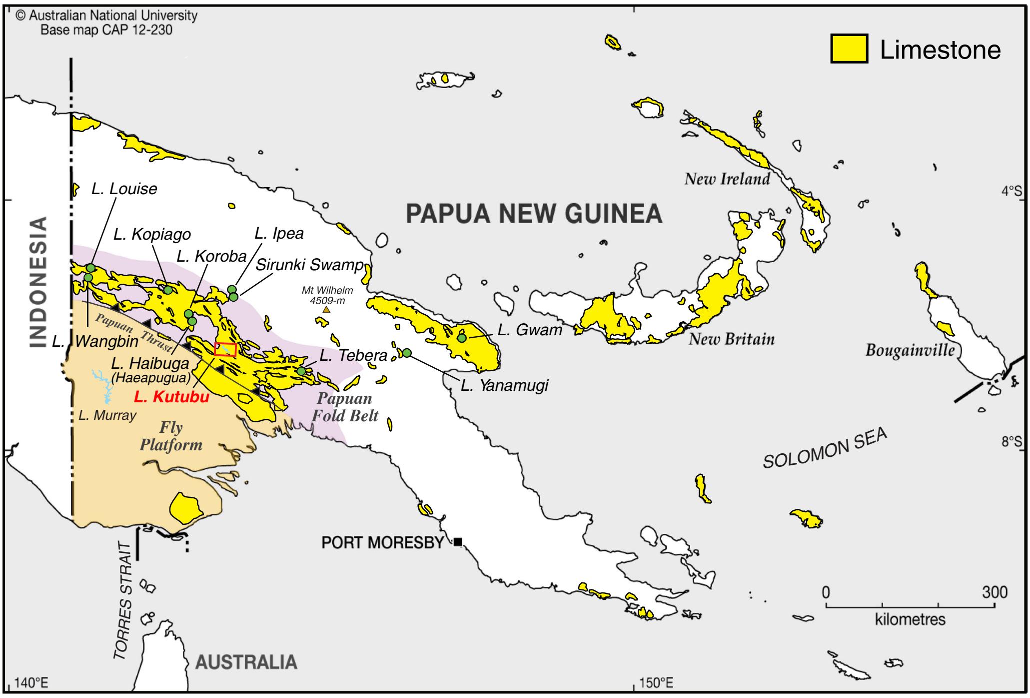

Figure 6 Limestone areas in Papua New Guinea, and the freshwater lakes on the mainland that are known to or may have a ^{14}C reservoir effect. The Papuan Fold Belt (purple), Fly Platform (yellow), and the intervening Papuan Thrust are indicated (from Williamson and Hancock 2005), as are areas of PNG with outcropping limestone in brown, modified from D'Addario (1975).

Implications for Lake Sequences Elsewhere in PNG

Limited attention has been given to the issue of freshwater reservoir effects in the carbonate catchment lake sediments of PNG. This is worrisome given the geological context of the island, which is favourable to the ^{14}C reservoir effect.

The main island of PNG has been formed by long-continued interaction between the Australian Plate in the southwest, and the Pacific Plate in the northeast. The ^{14}C reservoir effect in PNG is, therefore, mainly a result of the high occurrence of limestone areas in this country (Figure 6).

The highest occurrence of limestone is in the Papuan Fold Belt (Figure 6). This area was inundated in the early Miocene and shallow-water shelf limestone was deposited throughout the early and middle Miocene (Dow 1977). Table 6 presents a list of lakes within limestone bedrock areas, or lakes within drainage areas containing limestone (Figure 6, Supplementary Material). We consider them to require further ^{14}C reservoir studies in order to produce a reliable age-depth model to support palaeoecological research.

The ^{14}C reservoir effect in PNG is not a problem confined to limestone-rich catchments. Other rock units, especially in the Papuan Fold Belt, have calcareous and/or carbonaceous components (i.e. Bain et al. 1975) where $CaCO_3$-rich groundwaters are circulating freely. The Papuan Fold Belt has outcropping coal and shales with petroleum potential that may generate a ^{14}C

Table 6 List of freshwater lakes in Papua New Guinea suspected or known to have a reservoir effect in sediments (based on Figure 6 map).

Lake	Coordinates		Altitude (meter a.s.l.)	Reservoir effect (years)	Reference
Yanamugi Lake	6°24.730′S	146°16.236′E	217	1636	Garrett-Jones 1979
Lake Gwam	6°19′17.72″S	147°6′53.52″E	3517	2200	M. Prentice and G. Hope pers. comm
Sirunki	5°27′22.22″S	143°32′14.79″E	2500	Unknown	Flenley 1967; Walker and Flenley 1979
Lake Ipea	5°25′16.47″S	143°31′42.99″E	2500	Unknown	Flenley 1967; Walker 1972; Oldfield 1980
Lake Koroba	5°43′24.42″S	142°45′43.48″E	1500	Not detected	Williams et al. 1972; Haberle 1998
Lake Haibuga (Haeapugua)	5°49′7.89″S	142°49′9.09″E	1500	Not detected	Williams et al. 1972; Haberle 1998
Lake Louise	4°57′52.94″S	141°19′31.91″E	1600	Unknown	Chambers 1987
Lake Kopiago	5°24′19.1″S	142°29′3.32″E	1300	Unknown	Vyverman and Cronberg 1993
Lake Tereba	6°45′25.15″S	144°35′49.47″E	600	Unknown	Chambers 1987; Petr 1985
Lake Wangbin	5°14.670′S	141°15.299′E	1211	Unknown	Chambers 1987

reservoir effect (Brown and Robinson 1982). Old organic deposits, like peat bogs, can also cause a reservoir effect (Kilian et al. 2000).

Lake Kutubu Reservoir Age Offset and Worldwide Comparisons

When comparing Lake Kutubu to freshwater lakes elsewhere in the world, its reservoir age offset is similar to other limestone lakes, higher than limestone-free areas, and lower than lakes of glacial influence. In the Tibetan Plateau, for example, the reservoir effect in lakes was reported to be mostly in the range from 1000 to 3000 yr (Mischke et al. 2013). Permian limestones, Permian to Neogene clastic rocks and Quaternary unconsolidated sediments dominate the catchment (Wang and Yang 2004).

Queechy Lake in New York, a small lake lying on the Stockbridge marble (Paleozoic limestone) has shown a reservoir offset age of 2200 yr. The lake lacks inlets and receives its water by seepage through the limestone and the overlying glacial drift (Deevey 1954).

In the carbonate rocks (limestone and dolomite) and carbonaceous rocks (lignite, coal, and shale) of central North America, reservoir age offsets have been reported from 0 to 8000 yr, with an offset age of 500–2000 yr commonly found. In this region, much of this old carbonaceous material is in glacial drift that is redeposited in lake sediments as silt-sized particles (Grim et al. 2009).

Langvatnet Lake in Nordenskild Land in Svalbard, a Norwegian archipelago between mainland Norway and the North Pole, has shown ^{14}C reservoir offset ages of 1000 and 3000 yr when modern aquatic and terrestrial vegetation dates were paired. The reservoir effect stems from the lakes' bedrock, which is dominated by the Carboniferous Billefjord group of sedimentary rocks, including abundant limestone and dolomite as well as coal seams (Snyder et al. 1994).

A 1500 year ^{14}C offset age was reported for Lake Qinghai, the largest inland water body in China. The reservoir effect in this lake also comes from its watershed which is partly underlain by late-Paleozoic limestone (Yu et al. 2007).

Lakes without major influence from limestone usually have a lower offset age than lakes receiving waters from a limestone catchment. For example, Lake Chala in Africa has a reservoir offset age of 200 yr, which is caused by a variable contribution of old terrestrial organic matter eroded from soils, and controlled mainly by changes in vegetation cover within the crater basin (Blaaw et al. 2011).

Glacial lakes have the highest variability in the reservoir age offset error, because they have more environmental variables playing a role in the distribution of ^{14}C within lakes. In Lake Trowbridge, in Antarctic, a ^{14}C offset of ca. 18,000 yr has been reported. This large reservoir effect is believed to result from the direct input of old CO_2 from glacial meltwater (Hall and Henderson 2001). For lakes in the dry valleys of Antarctica, ^{14}C offset reservoir ages have been reported between 2700 yr and 20,000 yr. The ^{14}C reservoir effect in these lakes is primarily derived from subsurface melt of adjacent glaciers (Hendy and Hall 2006).

Future Directions for Studies in Lake Kutubu

In future studies in Lake Kutubu, we recommend: (1) application of tephrostratigraphy to complement ^{14}C dating, (2) application of the Fletcher et al. 2017 approach to dating pollen samples in carbonate rich lake environments, (3) the use of terrestrial macrofossils, (4) the collection and dating of extant snail shells and terrestrial plants from lake shore or islands at the same time as sediments are collected, (5) avoid collection of cores in the littoral zone where there is high frequency of *Chara* spp. and (6) avoid collection of cores close to any stream inlet.

Although only two tephras were recorded in our 1 m cores from Lake Kutubu, at least two tephras are expected to be found in deeper samples. Based on the available information on tephras occurring in Kuk Swamp, 130 km north east of Lake Kutubu (Blong et al. 2017a), at least two tephras may occur below Olgaboli in Lake Kutubu: Kim (3980–3630 cal yr BP) and Ep (18,480–14,920 cal yr BP).

The collection of cores far from any inlet to the lake is recommended to avoid the influence of additional carbon outside the local catchment. If lake cores need to be collected close to the inlet, dating the top samples using both ^{210}Pb and ^{14}C dating methods is recommended to determine the reservoir effect as a function of depth in the core.

CONCLUSION

The distribution of calcareous rocks in Papua New Guinea favours a ^{14}C reservoir effect in freshwater lakes. This study has produced a reliable ^{14}C-based chronology for the 4500-cal BP sediment record of Lake Kutubu, allowing future studies in this lake as key archives of past climate and environmental change in PNG. The reservoir age offset has been estimated through paired ^{14}C dates and ^{210}Pb and tephras.

Differences in reservoir age offset varied around the lake and within depth, demonstrating the need to verify the ^{14}C reservoir effect in all parts of the lake where sediments are used to produce historical studies. We recommend consideration of the inputs of external sources of carbon in lakes via inlets, which can require more proxies for refining the correction of the ^{14}C reservoir effect.

The use of tephras and terrestrial plants to correct for the ^{14}C reservoir effect has been successfully applied in this study, demonstrating the potential of tephra as chronomarkers of PNG palaeoecological events. Other lakes in PNG are likely to require corrections for ^{14}C reservoir effect: Yanamugi Lake, Lake Gwam, Sirunki, Lake Ipea, Lake Koroba, Lake Haibuga, Lake Louise, Lake Kopiago, and Lake Tereba.

The widespread distribution of calcareous and/or carbonaceous bedrock lithologies in PNG, coupled with the overall lack of recognition that ^{14}C reservoir effects may have had an influencing effect on previously published chronologies suggests that care is required in interpreting previously published age data and in the acquisition of future data. A multimethod approach similar to that adopted in this study is recommended.

The disentangling the reservoir effect in freshwater lakes of PNG is of high importance for future acquisition of long sediment cores and corresponding palaeoclimatic records from such lake systems will inevitably lead to advances in our overall understanding of the tempo and magnitude of tropical climate variability in the western Pacific realm. To our knowledge, this is the first study to address the ^{14}C reservoir effect in a freshwater lake in PNG.

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SUPPLEMENTARY MATERIAL

To view supplementary material for this article, please visit https://doi.org/10.1017/RDC.2018.49

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