Lacustrine sedimentological and geochemical records for the last 170 years of climate and environmental changes in southeastern China

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Reconstruction of modern climate and environmental changes in east Asia using inland natural climate archives can provide valuable insights on decadal–multidecadal climate and environmental patterns that are probably related to both natural and anthropogenic forcing. Here we investigated an 89-cm-long sediment core (TH1) from Tian Lake, southeastern China, for sedimentological, physical and geochemical parameters in order to understand climate and environmental changes for the latest two centuries. 

\(^{137}\text{Cs}\)- and \(^{210}\text{Pb}\)-based age models show that the fine sand–coarse silt-dominated core contains ~170 years (c. AD 1842–2011) of continuous sedimentation. Sediments with fine sand, low MS values, high water content, high TOC content and a high C:N ratio from c. AD 1842 to 1897 suggest intense hydrological conditions and strong runoff in the catchment, probably because of a humid climate. From AD 1897 to 1990, sediments with very fine sand and coarse silt, high MS values, low water content and unchanged TOC and C:N ratios indicate normal hydrological conditions and in-lake algal-derived organic matter. During this interval, the chemical weathering indicators show stronger weathering conditions compared with sediments deposited during AD 1842–1897, supporting the dominance of weathered surface soil input in the earlier interval and physical erosion dominance in the later period, respectively. Since AD 1990, the continuous decrease of geochemical proxies suggests human-interacted Earth surface processes in the catchment of Tian Lake. A PCA revealed four dominant geochemical controlling factors – detrital input, trophic status, grain size and early diagenesis –, accounting for 26, 20, 18 and 16% of total variance, respectively. This study for the first time provides lacustrine geochemical evidence for the most recent two centuries of climate and environmental changes in coastal southeastern China, a region that is currently undergoing an inversion of critical zone, i.e. an overturning of its soil profile, owing to swift modernization.

Lake sediments have been widely regarded as an important natural archive for the reconstruction of past climatic and environmental changes over centennial–millennial time scales in both humid and arid parts of China (Yancheva et al. 2007; Chen et al. 2010, 2013; An et al. 2012; Selvaraj et al. 2012). However, similar studies on shorter, decadal–multidecadal time scales focusing on the last millennium and part thereof using lake sediments from this region are rare. This may hamper our understanding of potential future environmental changes, given the drastically changing land-use pattern of China during the last two decades that might have a great impact on the sedimentation pattern of inland aquatic systems in the near future. Investigation of the hydrodynamic characteristics and erosion-weathering processes of a lake basin using multi-proxy analyses is common in palaeoenvironmental research (Selvaraj et al. 2007; Nahm et al. 2010; Chen et al. 2013). For example, grain-size variations of lake sediments are generally used as a reliable proxy for past changes in precipitation and eolian activity (e.g. Yan et al. 2011; Qiang et al. 2014). Likewise, sediment magnetic susceptibility (e.g. Warrier & Shankar 2009) and colour intensities (e.g. Moy et al. 2002) are used to infer past climate changes. In addition, sedimentary inorganic (Ti, Al, Si, K, Fe, Mg, Ca and other trace elements) and organic geochemical proxies (TC, TOC and C:N ratio) reflect information about physical erosion and chemical weathering processes as well as vegetation types in the catchment (Meyers 2004; Selvaraj et al. 2007, 2011, 2012; Giguet-Covex et al. 2011; Liu et al. 2014). Hence, a combination of diverse geochemical parameters is useful to extract information about past climate and environmental changes depending upon the sediment accumulation rate as well as the length of sediment core under investigation.

A substantial increase in climate and environmental reconstructions using lake sediment records has recently been achieved in China (e.g. Yancheva et al. 2007; Chen et al. 2010; An et al. 2012). However, most researchers have mainly focused upon longer-term climate changes, with only a few studies addressing environmental changes at shorter, centennial–decadal, time scales, especially in southeastern China, including Taiwan Island (Lou & Chen 1997; Selvaraj et al. 2007, 2011, 2012; An et al. 2011), with a complete lack of...
geochemical investigation on island lakes along the SE coast of China. There are around 6500 islands along the 18,000 km coastline of China, of which 93% are considered as not suitable for human settlement because of an acute shortage of fresh water. Nonetheless, Yushan Island, located in Fujian Province (Fig. 1A), is one island that has a lot of fresh water to support human habitation, with a current population of ~6500. Tian Lake, a relatively pristine lake and an important source of fresh water in this island, has never been investigated either in terms of water chemistry or sediment geochemistry (Fig. 1B). Here, we used an integrated sedimentological, physical and geochemical approach to infer the modern climate and environmental changes of Tian Lake using a short sediment core collected from the lake in May 2012. We aimed to investigate (i) the hydrological changes in the catchment surrounding the lake; (ii) the trophic status within the lake; (iii) the factors that drive the sediment input; and (iv) the processes that control the geochemical composition of sediments.

Study area

In China, the subtropical region falls between ~22 and 34°N latitude and is bordered by the Qingling Mountain and Huaihe River in the north, the Leizhou Peninsula in the south and extends to the Hengduan Mountain in the west (Wu 1980). Fujian Province, located at the southeastern boundary of the subtropical area (Fig. 1A), consists mostly of mountains with undulated topography and faces the East China Sea to the southeast. Tian Lake (latitude 26°56’52”N, longitude 120°20’56”E) is located on Yushan Island of Fuding City, northeast of Fujian Province and Yushan Island’s southeast border is the East China Sea (Fig. 1A). Covering an area of 21 km², Yushan Island is the seventh largest island in the province and has a coastline of ~30 km surrounded by the sea. The local bedrock of Fuding City consists of various rock types, including Carboniferous, Jurassic, Cretaceous and Quaternary ages, of which Jurassic and Cretaceous rocks are most developed, covering an area of about 1200 km², and accounting for ~78% of the total land area. Yushan Island is underlain mainly by Jurassic granites, dark grey intermediate acidic dacite and tuff lava (source: Compiling Team for Annals of Fuding County). The morphology of Yushan Island is largely defined by an abrasion landmass covered mostly with both coarse- and fine-grained granites (Fig. 2A–C).

Yushan Island is strongly influenced by subtropical oceanic monsoons and thus has a mild and moist climate, with mildly cold winters and humid summers. The rainfall is controlled dominantly by the East Asian monsoon, and has an annual mean of ~1300 mm. The mean annual temperature is 15.1 °C and the relative humidity is ~100%. The average number of foggy days per month is 6.2 days and the soil is mostly fertile red clay type, which is suitable for grass growth. Therefore, Yushan Island has widespread grassland (Fig. 1B), covering an area of around 667 km². The dominant species of grass is Miscanthus floridulus. Affected by the topography, the land hydrology in this island is mostly recharged from both groundwater and rainfall. Oceanographically, Yushan Island is a region of regular semidiurnal tide with mean annual macro-tidal variation of 5.01–5.83 m and mean neap tide varying between 3.37 and 4.10 m. Yushan Island contains three natural lakes, namely Large Tian Lake, Small Tian Lake and Jiuzhuxiachao Lake, with a combined gross water storage capacity of up to 1.7 million m³. Tian Lake has an average altitude of 306 m a.s.l., with minimum and maximum altitudes of 303 and 310 m.

Fig. 1. A. Map showing the location of Tian Lake on Yushan Island (black dot), southeastern China. B. A panoramic view of Tian Lake. Red arrow shows the location of the sediment core TH1 investigated in the present study.
a.s.l., respectively. As the lake direction is the same as the rock crevice direction, Tian Lake has been categorized as a tectonic fissure lake. Small and Large Tian lakes have areas of 0.1 and 0.7 km², respectively, with maximum depths of around 5 and 18.5 m. As Large Tian Lake has recently been dammed artificially for drinking and agricultural purposes, we selected the pristine Small Tian Lake, which is surrounded by 100–200 m high hills (Fig. 1B), for sediment coring, aiming to investigate climate and environmental changes for the last ~170 years.

Material and methods

Four short (<1 m long), push sediment cores were manually collected from chest-deep waters in Tian Lake during May 2012, using 4-cm diameter transparent plastic tubes. Amongst them, the longest sediment core (89 cm), named as TH1, was chosen for this study (Fig. 4). The core was subsampled at 1-cm intervals, except for the top depth of 1–2 cm and bottom depth of 86–87.5 cm, for sedimentological (grain size), physical (magnetic susceptibility and red, green and blue (RGB) colour intensities) and organic (total carbon (TC), total organic carbon (TOC), total nitrogen (TN)), as well as inorganic (major and trace elements) geochemical analyses.

Prior to the detailed laboratory work, all subsamples were freeze-dried for 24 h to achieve a constant weight and were homogenized. A low-frequency (0.47 kHz) magnetic susceptibility analysis was performed using a Bartington MS-2B sensor attached to a MS-2 susceptibility meter, and each sample was measured in triplicate. Grain-size analysis was carried out with a Coulter LS-100 laser particle size analyser, as detailed in Janitzky (1987). Briefly, samples were sieved to remove grains larger than 1000 μm and then treated with HCl and H₂O₂ to remove carbonate and organic matter, respectively. In the final stage, sodium hexametaphosphate was added to prevent the flocculation of clay minerals and the samples received a short period of ultrasonic agitation. RGB colour intensities of samples were measured by digital colour scale. Grain size, magnetic susceptibility and colour intensities were measured using instruments located at the Department of Marine Sciences, Chinese Naval Academy, Taiwan.

Forty-nine bulk sediment subsamples from every 2-cm interval were ground to /C24 200 micrometer in an agate mortar for geochemical analysis. Sediments were acidified with 1 N HCl to remove carbonates and then rinsed three times with distilled water, dried at 50 °C and homogenized using a mortar and pestle. Precisely 20 mg of sediment was weighed into a tin boat, which was then folded into a pellet. These pellets were measured for TOC and TN using a Vario EL III elemental analyzer (Selvaraj et al. 2007). The standard reference materials used were BCSS (2.24% C, 0.24% N) and NIST2704 (3.34% C, 0.22% N). C:N ratios were calculated as molar proportions. Selected major (Al, Ca, Fe, Mg, Mn, Na, K, P, S, Si and Ti) and trace (Ba, Rb, Sr and Zr) elements were determined using an X-ray fluorescence (XRF) spectrometer equipped with an Rh tube located at the State Key Laboratory of Marine Geology, Tongji University, China. Details of the XRF method have been described in Selvaraj & Chen (2006) and Selvaraj et al. (2010). Activities of ²¹⁰Pb and ¹³⁷Cs radionuclides in subsamples of core TH1 were measured with EG and G Ortec gamma spectrometry using

Fig. 2. A. Grey granite rocks on the mountain surrounding Tian Lake. B. Pink granite boulders, with a broken surface, on the mountain slope. C. Hammered rock pieces of coarse-grained intermediate dacite along the mountain slope.
a germanium detector located at the Institute of Limnology and Geography, Chinese Academy of Sciences, Nanjing, China (Yao et al. 2008).

Results and discussion

Chronology: 137Cs dating

The radioisotope caesium-137 ($^{137}\text{Cs}$, $t_{1/2} = 30.1$ years) is a fission product and was initially introduced into the environment in significant quantities as a result of atmospheric nuclear weapons tests conducted in the early 1950s (Nahm et al. 2010). Widespread global dispersal of $^{137}\text{Cs}$ to the environment began with high-yield thermonuclear tests in November 1952/C62 (Perkins & Thomas 1980). Also, the maximum fall-out of $^{137}\text{Cs}$ has been observed in the Northern Hemisphere during 1963, and in the Southern Hemisphere during 1964 (Longmore et al. 1983). The fall-out of $^{137}\text{Cs}$ from the 1986 Chernobyl accident was mainly observed at sites in Europe and has been used as a time marker for the sediment deposited during that year (Wieland et al. 1993; Fan et al. 2010).

The profiles of $^{137}\text{Cs}$ in coastal sediments of China generally have two to three distinct chronological markers (Chen et al. 2009; Sun et al. 2011). As for many places in the Northern Hemisphere, the time markers include the onset of measurable amounts in soils in 1954 and the peak fall-out in 1963. In some areas of China, there may also be another smaller peak of $^{137}\text{Cs}$ fall-out that probably derives from the Chernobyl fall-out in 1986 (Xiang 1998; Zhang 2005). However, in some situations, the 1954 time marker may give a false result because of the detection limit of $^{137}\text{Cs}$, which has a value of $\sim 0.5$ Bq kg$^{-1}$ (Campbell 1983).

Figure 3 shows the variations in $^{137}\text{Cs}$ activity with depth for our sediment core TH1. The $^{137}\text{Cs}$ pattern is very similar to the theoretical fall-out pattern, suggesting that such processes might not have occurred by chance (Grousset et al. 1999). The first peak, which had the highest $^{137}\text{Cs}$ activity (8.33 Bq kg$^{-1}$), may record the nuclear weapons fall-out peak that occurred in 1963, which appeared at 25–26 cm in core TH1. Secondary small peaks at around 13–14 cm (~6.05 Bq kg$^{-1}$) are likely to have been caused by the Chernobyl fall-out. As sampling was carried out in May 2012, we assumed that the surface layer of core TH1 corresponds to the sediment accumulated in AD 2011. The average sedimentation rate (SR) deduced from the 1963 time marker is 0.53 cm a$^{-1}$. The SR derived from the 1986 time marker is 0.54 cm a$^{-1}$. It is apparent from Table 1 that the SR given by the 1986 marker is the same as the SR determined from the 1963 peak $^{137}\text{Cs}$ activity.

$^{210}\text{Pb}$ dating

Lead-210 ($^{210}\text{Pb}$, $t_{1/2} = 22.3$ years) is a natural radioactive by-product of the $^{238}\text{U}$ decay series that is supplied to the environment via atmospheric precipitation (e.g. Selvaraj et al. 2010). The $^{210}\text{Pb}$ activity of lake sediments has two components, a supported component ($^{210}\text{Pb}_{\text{ss}}$) derived from $^{222}\text{Rn}$ decay within the

<table>
<thead>
<tr>
<th>Time of peak activity</th>
<th>Depth (cm)</th>
<th>Time-span (year)</th>
<th>Sedimentation rate (cm a$^{-1}$)</th>
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<tbody>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>1986</td>
<td>13.5</td>
<td>1986–2011</td>
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<tr>
<td></td>
<td>1963</td>
<td>25.5</td>
<td>1963–2011</td>
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<tr>
<td>$^{210}\text{Pb}_{\text{ss}}$</td>
<td>13–41</td>
<td>1932–1985</td>
<td>0.51</td>
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Figure 3. Down-core variations in $^{137}\text{Cs}$, $^{226}\text{Ra}$, excess $^{210}\text{Pb}$ ($^{210}\text{Pb}_{\text{ex}}$) and total $^{210}\text{Pb}$ activities in sediment core TH1 from Tian Lake. The profile of $^{210}\text{Pb}_{\text{ex}}$ is plotted on a logarithmic scale for the depth of 13–41 cm, giving a sedimentation rate of 0.51 cm a$^{-1}$. 

$y = 112.33 - 16.51 \ln x$
$R^2 = 0.84$
$\text{SR} = 0.51 \text{ cm a}^{-1}$
sediment column, and an unsupported (or excess) component \((^{210}\text{Pb}\text{ex})\) derived from the atmospheric fall-out of \(^{210}\text{Pb}\). \(^{210}\text{Pb}\) can, for most purposes, be approximated by the \(^{226}\text{Ra}\) concentration. In the absence of \(^{210}\text{Pb}\) fall-out, \(^{210}\text{Pb}\) and \(^{226}\text{Ra}\) would be in radioactive equilibrium. \(^{210}\text{Pb}\text{ex}\) is determined by subtracting \(^{226}\text{Ra}\) from the total \(^{210}\text{Pb}\) concentration (Appleby & Oldfield 1983). The unsupported \(^{210}\text{Pb}\) concentration in each sediment layer declines with its age in accordance with the usual radioactive decay law. This law can be used to calculate the age of the sediment provided that the initial unsupported \(^{210}\text{Pb}\) concentration when deposited on the lake bottom can be estimated (Appleby & Oldfield 1983).

Figure 3 shows the activity profiles of \(^{210}\text{Pb}\text{ex}, ^{210}\text{Pb}, \) and \(^{226}\text{Ra}\) in core TH1. The profile of excess \(^{210}\text{Pb}\) \((^{210}\text{Pb}\text{ex})\) shows a tendency to decrease with depth to \(~41\) cm, but generally homogenous activity occurs at \(~41\)–\(~86\) cm intervals, which is supported by the decay of \(^{226}\text{Ra}\). The profile of \(^{210}\text{Pb}\text{ex}\) activity at the depth of \(~13\)–\(~41\) cm shows an exponential decrease with minor fluctuations (Fig. 3). The age of sediments of depth \(m\) is thus calculated using the equation: \(t = \frac{A_{0}}{A} \frac{V}{k} - \frac{1}{k} \ln (A_{0}/A)\), where \(t\) is the age of sediments of depth \(m\); \(A_0\) represents the total excess \(^{210}\text{Pb}\) in the sediment column; \(A\) is the cumulative residual \(^{210}\text{Pb}\) in the depth \(m\) in the sediment and \(k\) is the \(^{210}\text{Pb}\) radioactive decay constant \((0.03114 \text{ a}^{-1})\). The SR calculated from \(^{210}\text{Pb}\text{ex}\) is 0.51 cm a\(^{-1}\) and this value is very similar to the SR \((0.53 \text{ cm a}^{-1})\) derived from the \(^{137}\text{Cs}\) dating (Table 1).

Figure 3 also shows that \(^{210}\text{Pb}\text{ex}\) profile is nonlinear from the surface to the bottom. This nonlinearity suggests that either sediment accumulation rates have varied slightly over time, or that a number of other factors are involved, such as migration of \(^{210}\text{Pb}\) through interstitial waters near the sediment–water interface (Koide et al. 1973), mixing of near-surface sediments by physical (Petit 1974) or biological (Robbins et al. 1977) processes, and postdepositional redistribution of sediments either discontinuously through slumping (Edginton & Robbins 1976) or more or less continuously by sediment erosion. The disequilibrium between \(^{210}\text{Pb}\) and \(^{226}\text{Ra}\) activity at the bottom of the core may mean that the lowermost appearance of \(^{210}\text{Pb}\) has not been reached in our sampling core.

Considering the uncertainty of \(^{210}\text{Pb}\) dating because of the above-mentioned factors and the close similarity of the SRs obtained between the \(^{137}\text{Cs}\) and \(^{210}\text{Pb}\) methods (Table 1), we adopted \(^{137}\text{Cs}\) dating to calculate the age of core TH1, given that the peak \(^{137}\text{Cs}\) activity in our sediment core is consistent with the peak \(^{137}\text{Cs}\) activity of the Northern Hemisphere reported in a number of previous studies (Zhang et al. 2005; Arnaud et al. 2006; Liu & Fan 2011). If we assume that the surface layer of core TH1 corresponds to AD 2011, then the bottom of the core (89 cm) corresponds to about AD 1842 (AD 1836), according to a linear extrapolation using the SR of 0.53 cm a\(^{-1}\) (0.51 cm a\(^{-1}\)) derived from the \(^{137}\text{Cs}\) \((^{210}\text{Pb})\) method.

**Lithology and magnetic susceptibility (\(\chi\))**

Variations in the grain size of lake sediments may be influenced by changes in lake level, palaeoenvironmental zones of deposition, and transport energy related to variations in rainfall and/or wind (Chen et al. 2004; Conroy et al. 2008). As the relative humidity in Yushan Island is ~100%, the grain size of Tian Lake may be controlled by overland flows from the catchment. Magnetic susceptibility is widely used to provide information on the concentration of ferromagnetic minerals present in soils and sediments (Dearing 1999). Magnetic properties of soil and sediments depend not only on minerals inherited from parent rocks, but also on pedogenic processes such as weathering and biogenic/authigenic formation of minerals (Ortega-Guerrero et al. 2004).

Lithologically, core TH1 consists of find sand, silty sand and sandy silt (Fig. 4). Mean contents of sand, silt and clay are about 72, 25 and 4%, respectively, throughout the core. The mean grain size ranges from 34 to 327 \(\mu\text{m}\) with a mean value of 129 \(\mu\text{m}\), indicating the dominance of fine–very fine sand and coarse silt. The median grain size varies between 35 and 618 \(\mu\text{m}\) with a mean value of 170 \(\mu\text{m}\) (Fig. 4). The trend of median grain size is similar to the mean grain size. There is not much change in the composition of sediment in the bottom 15 cm and top 13 cm. In the 67–13 cm interval, which roughly equals the time interval from AD 1885 to 1986 (AD 1880–1985), the grain size fluctuated significantly, with two peaks at ~29.5–41.5 cm and ~51.5–55.5 cm. The average \(\chi\) value in our core TH1 is 3.2 SI. \(\chi\) is close to zero from the base of the core to ~70 cm. The middle section (70–13 cm) of the record is characterized by fluctuating \(\chi\) values (0.8–11.5 SI), whereas the upper portion of the record shows a decreasing trend towards the core top.

**TC, TN, TOC and C:N ratio**

The atomic ratio of total organic carbon to total nitrogen (C:N ratio) of organic matter is little influenced by decomposition and can be a reliable proxy for organic matter sources (algae-derived or land-derived), albeit with some small early diagenetic alterations (Meyers 2004). Algae generally have atomic C:N ratios <10, whereas land plants commonly have ratios >20 (Meyers & Ishiwatari 1993). The TOC concentration can provide important information about variations in organic matter derived from land plants, which is brought into the lake via runoff when precipitation is high (Meyers et al. 2006; Selvaraj et al. 2007). Therefore, a combination of high TOC content and high C:N ratios is probably related to higher inputs of soil
material accompanied by organic matter from the catchment during enhanced rainfall. By contrast, a small amount of land plant material was delivered when rainfall was low and during which the static water column may also favour aquatic productivity and thus sediments in general have low C:N ratios.

The TOC content in core TH1 ranges from 0.5 to 10.9%, with an average TOC content of 2.3% (Fig. 5). In the bottom ~10 cm, the TOC content is high and ranges from 4.3 to 10.9%, with large oscillations compared with the top 78.5 cm of the core, where the TOC content is comparatively less, ranging from 0.5 to 3.5%. TOC shows an increasing trend from 78.5 cm, corresponding to AD 1863 (AD 1857). TN content (0.09–0.53%) in the sediment core varies in concert with TOC toward the top of the core. C:N ratios are high in the bottom ~10 cm of the core, whereas in the top 78.5 cm of the core they remain mostly at ~8–10 with fewer fluctuations. The TN shows a strong linear correlation with TOC ($R^2 = 0.88$, $p < 0.0001$; Fig. 6) with a positive intercept on the TN axis for sediments from the upper 78.5 cm, suggesting the presence of a small amount of inorganic N (~0.03%) in TN. TOC-rich sediments from the singular indicate larger proportions of land-derived organic matter, as also evidenced by higher C:N ratios in these sediments (Fig. 5), although they contain proportional content of inorganic N (~0.17%; Fig. 6), probably because of the presence of clay-associated N in terrestrial materials in the bottom part.

**Proxies of chemical weathering**

As soils and rocks in the catchment represent major sources of terrigenous input into the lake (Bertrand et al. 2005), elemental concentrations in sediments are

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**Fig. 4.** Litho-log of core TH1 along with down-core distributions of clay (<4 μm), silt (4–64 μm) and sand (>64 μm) contents (all in %) as well as mean grain size (Md, μm), median grain size (Md, μm) and magnetic susceptibility ($\chi$ in SI) against year (in AD). The two horizontal shaded bars mark the dominance of medium-size sand in core TH1 during AD 1906–1913 and AD 1935–1953, suggesting a stronger erosion in the catchment, probably as a result of the influence of typhoons in SE China.

**Fig. 5.** Total carbon (TC), total organic carbon (TOC), total nitrogen (TN) and the TOC to TN (C:N) ratio vs. depth and age in sediment core TH1.

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helpful to reconstruct the intensity of chemical weathering, an important process in the global hydro-geochemical cycle of elements (Gaillardet et al. 1999; Warrier & Shankar 2009; Gupta et al. 2011). As this process is strongly influenced by rainfall and temperature (Ollier 1984; Gaillardet et al. 1999), humid tropical and subtropical climates with denser vegetation in general enhance the weathering of source rocks (Selvaraj & Chen 2006). Conversely, in arid or polar climates, weathering intensity is weak to non-existent owing to either low precipitation or water locked in ice and thus erosion predominates over weathering with few to no new secondary mineral formation (Schönborn & Fedo 2011).

The degree of weathering can be obtained by calculation of the chemical index of alteration (CIA): 

\[ \text{CIA} = \frac{100\text{Al}_2\text{O}_3}{\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O}} \] (Nesbitt & Young 1982), and the plagioclase index of alteration (PIA): 

\[ \text{PIA} = 100\left[\frac{\text{Al}_2\text{O}_3 - \text{K}_2\text{O}}{\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} - \text{K}_2\text{O}}\right] \] (Fedo et al. 1995), where CaO* is the amount of CaO incorporated in the silicate fraction. These indices indicate the degree of decomposition of total feldspar (both alkali and plagioclase) and plagioclase feldspar, respectively, to secondary clay products, where CIA (PIA) values of about 50 indicate unweathered bedrock (plagioclase), and values of 75–100 indicate complete conversion of feldspars to aluminous clay minerals (intense chemical weathering; see Fedo et al. 1995). Intensely weathered rock thus yields kaolinite or gibbsite with CIA/PIA values of 100 (Fedo et al. 1996, 1997).

Similar to CIA and PIA, ratios such as Rb/Sr, K/Al and Al/Na are also good proxies of chemical weathering intensity (Selvaraj & Chen 2006; Clift et al. 2008; Selvaraj et al. 2010), as increasing weathering rapidly leaches Sr (Na) compared with Rb (Al) (Nesbitt & Young 1982) and, therefore, the Rb/Sr and Al/Na ratios increase in the weathered product (Ma et al. 2000). K/Al has been used as a proxy for the illite/kaolinite ratio (Yarincik & Murray 2000), as illite represents the dominance of physical weathering in temperate to arid climates, whereas kaolinite indicates intense weathering in tropical, humid climates (e.g. Bonatti & Gartner 1973).

As shown in Fig. 7, the CIA values for core TH1 range from 79 to 86 with an average of 84. Such high CIA values indicate that most primary feldspar minerals in the catchment have been converted to aluminous clay minerals (Fedo et al. 1996). Consistent with the
CIA values, the PIA values for core TH1 vary from 89 to 96 with an average of 94, indicating that all primary plagioclase feldspars have been converted to secondary clay minerals. High Rb/Sr ratios (1.21–2.05) with a mean value of 1.84, which is higher than the Rb/Sr ratios of different shales (0.80–0.88; Selvaraj & Chen, 2006), supporting the idea that source rocks in the catchment might have been intensively weathered because of favourable climatic conditions (high rainfall and high temperature) in tropical and subtropical regions (Selvaraj & Chen 2006). Rb/Sr ratios in illite minerals are usually >1, with a maximum value of 6.46 (Chaudhuri & Brookings 1979), suggesting that the high illite content in the lake sediments might be responsible for the high Rb/Sr ratios. Values for K/Al range from 0.16 to 0.27, averaging 0.21. The variations in K/Al follow CIA, with the ratio increasing during warm periods and decreasing during cold periods. The degree of chemical weathering of lake sediments is high, as shown by their very high Al/Na ratios (32–95 with an average value of 61), resulting from the extreme dissolution of plagioclase (Fig. 7). This weathering condition is comparable to lake sediments from the sub-alpine region of nearby Taiwan Island, which show extreme weathering because of the high precipitation and dense vegetation (Selvaraj & Chen 2006; Selvaraj et al. 2007). This similarity also suggests regional uniformity of chemical weathering of rocks in subtropical Taiwan and SE China, although the catchment of Tian Lake is devoid of meta-sedimentary rocks, which are dominantly present in Taiwan Island.

**Principal component analysis (PCA)**

Geochemical factor analyses have been used for determining the factors that control the sources of sediment (Selvaraj et al. 2010) and processes that control the geochemical composition of coastal and river sediments (Chen & Selvaraj 2008). Therefore, to disentangle significant grouping of factors and relationships to possible geochemical controlling processes, all of the sediment geochemical data, including grain size, magnetic susceptibility and colour intensities, were subjected to factor analysis using SPSS software (version 17.0, Chicago, IL, USA). The factors were extracted using the Varimax rotation scheme with Kaiser normalization.

The results reveal that four factors account for 80% of the total variance (eigenvalue >2, Table 2). Factor 1 accounts for 26% of the variance and Ti, Al, Mg, K, Pb, Ba, Rb and Sr are positively loaded in this factor. The strong positive correlations of Ti (r = 0.85), Ba (r = 0.79), Pb (r = 0.75), Rb (r = 0.74), Sr (r = 0.62), K (r = 0.79) and Mg (r = 0.83) with Al indicate that they are associated with aluminous clay minerals such as kaolinite, illite and/or smectite Table 3. In general, all these elements showed a similar pattern over the whole core (Fig. 8). Ti, Rb, Ba, Sr, K and Mg are in general incorporated into clay minerals during chemical weathering, whereas Ca and Na tend to be leached (Nesbitt et al. 1980; Fedo et al. 1996). These elements are mainly derived from the catchment and thus related to detrital input into the lake.

Variables such as intensities of red, green and blue colours, and magnetic susceptibility are positively loaded and Ca and S are negatively loaded in factor 2, which accounts for 20% of the total variance. The relationships between colour and the physical and chemical characteristics of soil, such as iron, organic matter, moisture content, mineralogy and structure, have been studied (Krishnamurti & Satyanarayana 1971; Davey et al. 1975). It is evident from Fig. 9 that Ca and S may be responsible for the low values for colour intensities as well as for magnetic susceptibility in the bottom 10 cm of the core. The covariance of Ca, S, RGB and magnetic susceptibility suggests that similar environmental conditions caused their accumulations/ variations. High C:N ratios are generally associated with land plants and the increase in sediment TOC in the bottom part of the core may reflect an increase in land-derived organic matter in the lake (Fig. 5). When land-derived organics increase, lakes become anoxic, resulting in high TOC preservation in sediment and thus reducing sediment magnetic susceptibility values. Such an interpretation receives support from the S profile in our study.
Therefore, factor 2 may be interpreted as a representation of the trophic status.

Factor 3 explains about 18% of the total variance and has negative loadings for clay and silt contents and positive loadings for sand content, mean grain size and median grain size. As variations in the grain-size data are primarily attributed to catchment runoff variability, factor 3 can be interpreted as representing rainfall variations in the study area. Two peak values of mean and median grain sizes around AD 1906–1913 and AD 1935–1953 (Fig. 4) perhaps seem to be attributed to the influence of typhoons in the study area, given that heavy downpours associated with tropical storms can transport coarser sediments, although a causal link needs to be clarified in future studies.

Factor 4 accounts for 16% of the total variance and shows positive loadings with Fe, Na, Zr and Mn and a negative loading with Si, probably suggesting the influence of Fe-Mn oxyhydroxides in the sediments. Figure 10 shows enrichment of Fe and Mn in the surface sediment, which may be attributed to the redox-related diagenetic recycling of iron and manganese in sediments (Boyle et al. 1999; Boyle 2001). Mn and Fe in sediments are dissolved under reducing conditions, migrate upward and accumulate in the oxidized surface sediments. Si is not only associated with many aluminosilicate minerals, but is also related to diatom productivity as a component in their frustules (Peinerud 2000). Based on poor correlations with K, Ti and Rb (Table 3), Si behaviour seems to be controlled by primary productivity. The negative relationship between Si and Zr found here substantiates a biogenic source of Si (Table 3).

Consistently, the depth profile of the Si:Al ratio (Fig. 11), an approximate indicator of biogenic silica, shows that the Si:Al ratio of the lake sediments is mostly less than the mean ratio of 4.13 obtained from eight rock samples investigated in the catchment (range: 3.07–4.66). The plot also includes the Si:Al ratio of two soil samples (2.01 and 2.32) investigated from the catchment with a mean ratio of 2.17. The Si:Al ratio in sediments depends upon the Si:Al ratio of the settling suspended particles. If the suspended matter is dominated by detrital particles (i.e. physically eroded soil or bedrock particles), the Si:Al ratio will be
Table 3. Correlation coefficients ($r$) between different pairs of variable ($n = 49$). Bold values are significant at the 0.01 level. MGS = mean grain size;Md = median grain size; MS = bulk magnetic susceptibility.

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close to the ratio in the soil or bedrock within the catchment. If we assume that the soil in the catchment is free of biogenic silica, then the depth profile of the Si:Al ratio suggests the presence of biogenic silica (5–10%), especially in the sediments deposited from 68.5–86 cm (20 cm) and sediments deposited from 12.5–3.5 cm in core TH1 (Fig. 11). However, the total CaO content of the sediments measured using XRF was always <0.6% (Fig. 11). This amount is very low compared with the mean content of CaO (1.99%) in eight rock samples analysed from the catchment of Small Tian Lake, suggesting that the sediments of this lake contain insignificant amounts of biogenic materials in the form of carbonate and thus are dominantly detrital in nature. The negative relationship of Si with Fe and Mn therefore indicates the dilution effect of authigenic Fe-Mn hydroxides on detrital silica rather than biogenic silica (Boyle et al. 1998). Thus, factor 4 may be regarded as representing the dilution of detrital input by Fe-Mn enrichment resulting from early diagenetic reactions.

Thus, when arranged all four factors depth-wise (Fig. 12), changing PCA scores reveals the importance of relative components to the overall geochemical record. PCA1 shows larger variation at a depth of 86–56 cm, corresponding to AD 1848–1905 (AD 1842–1901) and then decreases slightly to a depth of 13.5 cm. PCA1 decreases significantly over the top 13 cm. PCA2 increases from the bottom to a depth of
around 33 cm and then decreases slightly to the top. PCA3 shows a decreasing trend from the bottom to a depth of 63.5 cm and then varies relatively significantly. PCA4 decreases from the bottom to a depth of 66.5 cm and then increases to the top.

Palaeolimnological interpretation

The bottom layer of core TH1 is dated at AD 1842 (AD 1836), thus enabling us to reconstruct the last 170 years of climate and environmental changes in southeastern China. Three periods can be distinguished: two short, erosion-dominated terrigenous input intervals at 1842 to 1900 (89–58 cm) and 1986 to 2011 (13–0 cm), and a relatively long period (1900–1986) of weathering-dominated terrigenous input (Fig. 8).

During the period from AD 1842 to 1900, the mineral load was generally low and punctuated by extreme events of relatively short duration, probably related to extreme cold fluctuations that occurred during this time (Figs 7, 8). It has been suggested that southeastern China experienced a very severe cold winter in AD 1892, when many lakes (e.g. Tai Lake and Dongting Lake) and rivers (e.g. low reaches of Huaihe, Huangpu and Yangtze Rivers) became frozen for a long duration, as recorded in Chinese historical documents (Zheng et al. 2012). After AD 1891, the mineral loads show an abruptly decreasing trend, which may correspond to this severe cold event. In this interval, some peaks of TOC, TN and the C:N ratio are consistent with mineral loading, indicating that the organic source of sediment was mainly terrestrial material (Fig. 5). Down-core variations in CIA, PIA, K/Al, Rb/Sr and Al/Na also show relatively lower values (Fig. 7), indicating relatively weak chemical weathering of rocks in the catchment during this interval. During AD 1842 to 1876 (71–86 cm), the relatively high value and stable level of grain size and high water content suggest wetter than normal hydrological conditions during the formation of sediment, probably related to high precipitation. High values of TOC (>4%), TN (>0.2%) and C:N ratios (>10) further support high levels of runoff in the catchment as a result of high rainfall and thus delivery of terrestrial materials into the lake. The χ-value is close to zero from the base of the core to 71 cm, which is consistent with high water content and TOC values of sediments, suggesting that the organic matter is diamagnetic in nature (Figs 4, 5). High concentrations of S and Ca in the bottom 15 cm suggest a sub-oxic/anoxic environment within the lake probably because of high water levels, supporting the theory of heavy rainfall during AD 1842–1876 (Fig. 9). Therefore, the climate was generally cold from AD 1842 to 1900, although the climate from AD 1842 to 1876 was more humid than during AD 1876–1900. Such an interpretation of less humid conditions from AD 1876–1900 is roughly consistent with a higher number of typhoons observed in c. AD 1870–1930 in Taiwan when low solar activity prevailed (Hung 2013).

Overall, from AD 1900 to 1986, the levels of elements related to detrital input show a highly weathered nature of sediments, although they decreased up-core slightly (Figs 7, 8). All chemical weathering indices indicate greater chemical weathering of rocks in the catchment compared to sediments that accumulated during AD 1842–1900, probably because of a warm and humid climate (Fig. 7). However, distinct variations in grain size suggest that the depositional environment was relatively turbulent (Fig. 4). For instance, two medium-size sand-dominated layers in the middle of core TH1 correspond to AD 1906–1913 and AD 1935–1953 (137Cs dating; Fig. 4) and AD 1902–1910 and AD 1931–1951 (210Pb dating), suggesting that the hydrodynamic conditions in the catchment were probably wetter with high surface runoff during these intervals. This may imply the influence of typhoons around these time intervals (Fig. 4). Given that there is no prominent river input into the lake and that the lake is surrounded by hills (~100–200 m a.s.l.), the deposition of medium-size sand-dominated layers in these intervals might have contributed to greater hill slope erosion in the catchment. Consistent with this inference, the reconstruction of temperatures for the last 500 years from historical records in China revealed that the warmest interval was from the 1920s to the 1940s (Wang et al. 1991). Likewise, the historical inventory contains 41 typhoon strikes during the 26-year interval between AD 1884 and 1909 (Liu et al. 2001). The position of the medium-size sand-dominated layers in core TH1 roughly corresponds to the warmest epoch and the period of intense typhoon strikes, indicating that our sedimentary proxy records, in particular the grain-size proxies, may respond to the influence of stronger typhoons in SE China and thus may be an ideal natural archive for the reconstruction of decadal–centennial scale climate changes in SE China. The relatively low C:N ratios with few fluctuations suggest an algal source of organic matter and thus a lacustrine environment during this interval. The amounts of Fe and Mn increase as a result of precipitation of Fe-Mn oxyhydroxides, resulting in increases in χ and RGB intensity (Figs 9, 10). Low values of Ca and S also substantiate the existence of a warm and humid climate during this interval (Fig. 9).

In the last ~25 years (AD 1986–2011), the mineral load reduced abruptly and then slightly increased (Fig. 8). Chemical weathering proxies also show trends similar to the mineral loads (Fig. 7). The mean grain size shows smaller fluctuations, probably indicating only one source of sediment. The median grain size is also relatively constant, indicating a stable depositional environment (Fig. 4). These changes may be attributed...
to changes in land use patterns over the last two decades, an interpretation that is roughly consistent with the expansion of the urban construction area in nearby Fuzhou City to 64.2 km² during the AD 1988–2000 period (Bai & Chen 2013). High concentrations of Fe and Mn, however, suggest a well-oxygenated sediment–water interface probably because of low rainfall, resulting in a lower lake water level. When the lake level is low, it is expected that the core location become closer to the catchment, resulting in either the accumulation of larger grain sizes at the core site, or the precipitation of Fe-Mn oxyhydroxides, both might have diluted the detrital input (Figs 7, 8, 10).

Conclusions

Sedimentological, physical, and organic and inorganic geochemical records of a sediment core from Tian Lake in SE China have provided unprecedented records of climatic and environmental changes and related catchment processes for southeastern coastal China over the last ~170 years. Our study revealed that the climate was mostly wet during this time period. Although generally wet, the geochemical parameters indicate the dominant input of physically eroded materials during c. AD 1842–1900, whereas chemically weathered surface soil input seems to have dominated over a relatively longer period from c. AD 1900–1986. The situation was completely different since AD 1986 until the end of the core dating period in 2012, during which time geochemical parameters suggest the influence of land use changes on the sedimentation pattern and/or dilution of detrital elements by early diagenetic enrichments of Fe and Mn. Given that most coastal estuaries and shallow coastal systems have already been altered because of intense economic development along the coastal zone of SE China, the wide variety of physical and geochemical data provided here will be useful for the future assessment of climate and environmental, including man-made, changes to the coastal and inland aquatic systems of SE China.

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