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# Permafrost thaw induced abrupt changes in hydrology and carbon cycling in Lake Wudalianchi, northeastern China

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## ABSTRACT

**Lakes in the permafrost zone have been proposed to serve as key outlets for methane and carbon dioxide emissions. However, there has been no geological record of the hydrological and biogeochemical responses of lakes throughout the thawing of surrounding permafrost. We use multiple biomarker and isotopic proxies to reconstruct hydrological and biogeochemical changes in Lake Wudalianchi in northeastern China during regional thawing of the permafrost. We show permafrost thawing, as indicated by lignin degradation, initiated rapid lake water freshening as a result of the opening of groundwater conduits, and negative organic $\delta^{13}$C excursion due to increased inorganic and organic carbon fluxes. These hydrological changes were followed, with an ~5–7 yr delay, by abrupt and persistent increases in microbial lake methanotrophy and methanogenesis, indicating enhanced anaerobic organic decomposition and methane emissions from lakes as permafrost thaws. Our data provide a detailed assessment of the processes involved during permafrost thaw, and highlight the importance of lakes in ventilating greenhouse gases to the atmosphere.**

## INTRODUCTION

Permafrost soils, which currently cover ~14% ($21 \times 10^6$ km$^2$) of the global land surface (Obu et al., 2019) and store >30% (1330–1580 $\times 10^{15}$ g) of global soil organic carbon (Schuur et al., 2015), are particularly sensitive to climate warming. Warming-induced permafrost thaw greatly increases groundwater conductivity (Walvoord and Kurylyk, 2016; Lamontagne-Hallé et al., 2018) and rates of biogeochemical cycling (Vonk et al., 2015), leading to rapid loss of previously frozen organic carbon through microbial decomposition (Schuur et al., 2015). The resulting fluxes of the greenhouse gases methane ($CH_4$) and carbon dioxide ($CO_2$) create a positive feedback mechanism to accelerate global climate warming (Koven et al., 2011).

Lakes in the permafrost zone are considered "hotspots" for greenhouse gas release to the atmosphere via ebullition (Walter et al., 2006; Walter Anthony et al., 2016; Wik et al., 2016). Numerous incubation experiments and field observations based on $^{14}$C signatures have demonstrated that excessive $CH_4$ and $CO_2$ emitted from permafrost-affected lakes are derived primarily from old soil organic carbon with variable ages ranging from Pleistocene to late Holocene (Zimov et al., 1997; Walter et al., 2006; Walter Anthony et al., 2016). The strong correlation between the radiocarbon ages of the released $CH_4$ and those of the surrounding permafrost soil carbon suggests that the $CH_4$ ebullition is primarily produced from soil carbon inputs (Walter Anthony et al., 2016). However, results based solely on radiocarbon ages in $CH_4$ could be confounded by variably old carbon preserved in the seasonally frozen ground of the surrounding permafrost (Wild et al., 2019). Up until now, there has been no geological record from lakes indicating an abrupt or progressive input of carbon and the associated biogeochemical changes from regions undergoing permafrost thaw.

We present an annually to biannually resolved record of lignin degradation, lipid biomarkers, and isotopic ratios ($\delta^{13}$C and $\delta^{15}$N) in a sediment core of exceptionally high sedimentation rate from Lake Wudalianchi, northeastern China. The Wudalianchi region has undergone permafrost thaw starting from the 1970s and 1980s (Jin et al., 2007), creating a unique and valuable opportunity to investigate the full process of hydrological and biogeochemical changes throughout the permafrost thaw in the context of regional and global climate changes.

## MATERIALS AND METHODS

Lake Wudalianchi (48°40′N–48°47′N, 126°06′E–126°15′E) is currently located near the southern limit of permafrost (SLP, Fig. 1B) (Jin et al., 2007; Fig. S1 and Text S1 in the Supplemental Material[1]). A sediment core was collected from the center of Lake Wudalianchi Pool 2 (Fig. S2) and was well dated by $^{210}$Pb and $^{137}$Cs (Fig. S3; Text S2). We determined multiple proxies, including alkenone isomers, lignin

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[1]Supplemental Material. Supplemental information on the study sites, sampling, dating, methods, and significance of proxies. Please visit https://doi.org/10.1130/GEOL.S.14582745 to access the supplemental material, and contact editing@geosociety.org with any questions.

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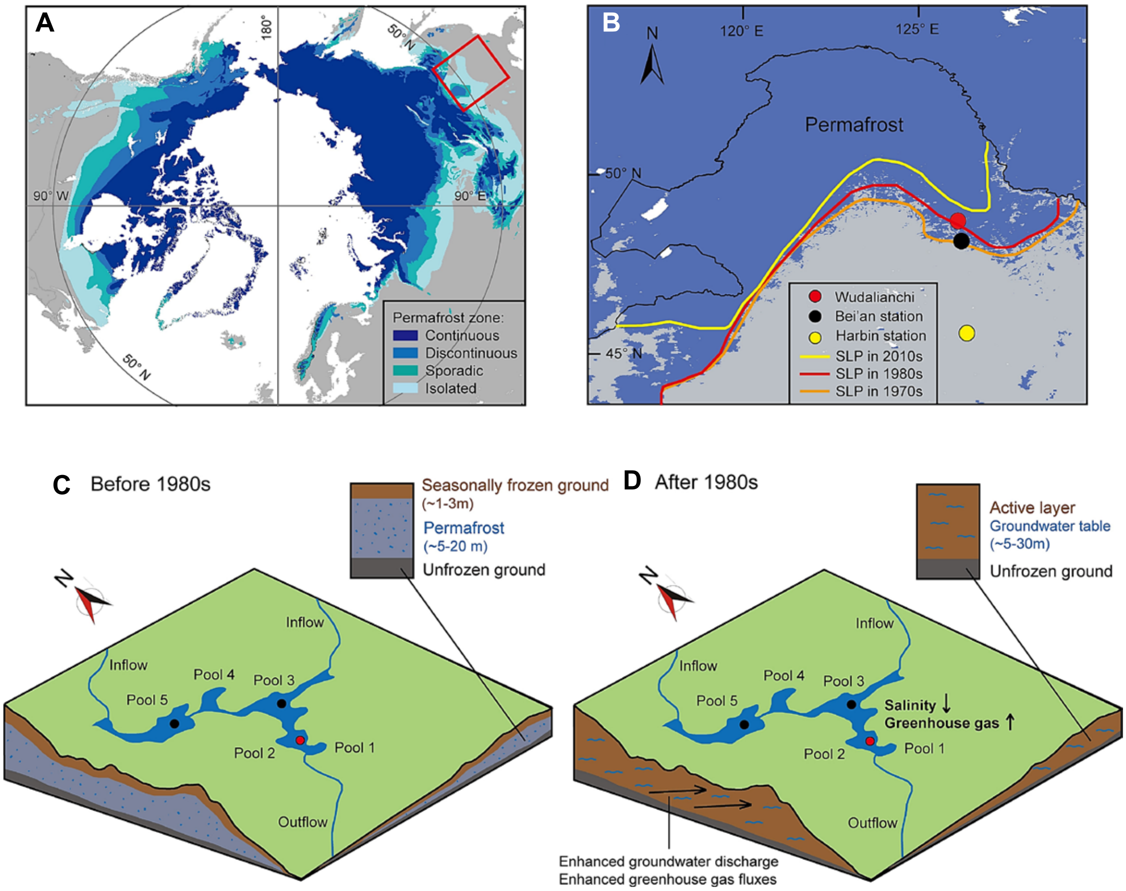


**Figure 1. Map of Lake Wudalianchi study site, with modeled changes in the southern limit of permafrost (SLP) boundary, in northeastern China. (A) Global map of permafrost zones (Brown et al., 2014). Red rectangle shows location of B. (B) Modeled SLP changes in northeastern China in the 1970s, 1980s, and 2010s (Fig. S1 and Text S5 [see footnote 1]) as well as the locations of Lake Wudalianchi and the Bei'an and Harbin weather stations. (C,D) Schematic diagrams illustrating hydrogeologic changes and enhanced greenhouse gas fluxes into Wudalianchi before and after the 1980s. (C) Before the 1980s, in our study region, thickness of seasonally frozen ground (Chen, 1994) was ~1–3 m and the permafrost soil layer had a variable thickness (Jin et al., 2007) of ~5–20 m. (D) After the 1980s, groundwater discharge and greenhouse gas fluxes through the active layer increased due to thawing of the permafrost. The depth of the groundwater table is ~5–30 m, in general, in our study region (Chen, 1994). Red circle represents our study sediment core site in Lake Wudalianchi Pool 2; black circles represent published sediment core sites in Pool 3 and Pool 5 (Gui et al., 2012).**

phenols, diploptene carbon isotopes, isoprenoid glycerol dialkyl glycerol tetraethers (iGDGTs), bulk nitrogen and organic carbon isotopes, and total nitrogen and organic carbon contents (Text S3), for reconstructing past hydrological and biogeochemical changes.

## RESULTS AND DISCUSSION

We used the alkenone isomer–based $RIK_{37}$ index as a salinity proxy (Kaiser et al., 2019; Yao et al., 2020a; Text S4) to reconstruct past hydrological changes in Lake Wudalianchi. Employing our regional $RIK_{37}$-salinity calibration (Figs. S4 and S5; Text S1), our reconstructed salinity has varied between 0.13‰ and 1.76‰ in Lake Wudalianchi since 1776 CE (Fig. 2A). Unfortunately, there are no continuously monitored salinity data available from Lake Wudalianchi to verify our reconstructed salinity values; our emphasis here is on the timing and rate of relative salinity changes (i.e., not absolute salinity values) since 1776. We observe an abrupt salinity decrease starting from 1975 to 1982 at a rate of ∼0.14‰ per year. After 1982, salinity generally continued to decrease but at a slower pace. The rapid freshening from 1975 to the present is not consistent with overall changes in rainfall and relative humidity, as illustrated by Palmer drought severity index (PDSI) variations (Cook et al., 2010) (Fig. 2), indicating precipitation and soil moisture content are not the primary controls on the lake water salinity.

The abrupt freshening of lake water coincides with significant regional warming (Figs. 2A and 2D). Due to the lack of glaciers in the Wudalianchi region (Chen, 1994), ice and/or snow meltwater has a negligible impact on changes in lake water salinity. Importantly, northeastern China has the second largest permafrost region in China, after the Tibetan Plateau (Jin et al., 2007), and warming-induced regional permafrost thaw is thus the most likely culprit for the observed hydrological changes since 1975. Our isotherm model results show that the Wudalianchi catchment crossed over the SLP from the 1970s to the 1980s (Fig. 1B; Fig. S1; Text S5), indicating the potential loss of regional permafrost or substantial deepening of the active layer (the layer of soil on top of permafrost that does not stay frozen all year) around that time. Before the 1980s, permafrost served as an impermeable barrier and restrained vertical and lateral groundwater flow (Walvoord and Kurylyk, 2016) due to the relative shallow seasonally frozen ground in our study region (Fig. 1C). With the accelerated warming after the 1980s (Fig. 2D), the regional permafrost loss or substantial deepening of the active layer opened up previously permafrost-blocked hydrogeological pathways (Walvoord and Kurylyk, 2016; Figs. 1B and 1D; Fig. S1). This provided a perennially open conduit for laterally transmitting underground fresh water into the Wudalianchi catchment and the connected rivers, leading to the persistent freshening of lake water since 1975 (Fig. 2A).

Lignin biomarkers derived from vascular plants are excellent tracers for thawing permafrost (Text S7). Vd/Vl ratio (vanillic acid/vanillin) and lignin acid (vanillic acid, syringic acid, *p*-coumaric acid, and ferulic acid) fluxes in our records abruptly increase from the 1970s to the 1980s, generally coinciding with periods of decreasing salinity (Figs. 3A–3C). These data indicate a rapid increase in the degree of lignin decomposition and increased fluxes of residual lignin degradation products into Lake Wudalianchi. Our lignin data are fully consistent with regional permafrost thaw from the 1970s to the 1980s. The several-year delay in changes in the Vd/Vl ratio relative to total lignin acids may be due to the relatively slow degradation rate of vanillic acid.

To examine how permafrost thaw affects carbon cycling in Lake Wudalianchi, we measured $\delta^{13}C$ values of the bulk organic matter to decipher the overall changes in carbon cycling. We also analyzed two sets of lipid biomarkers,

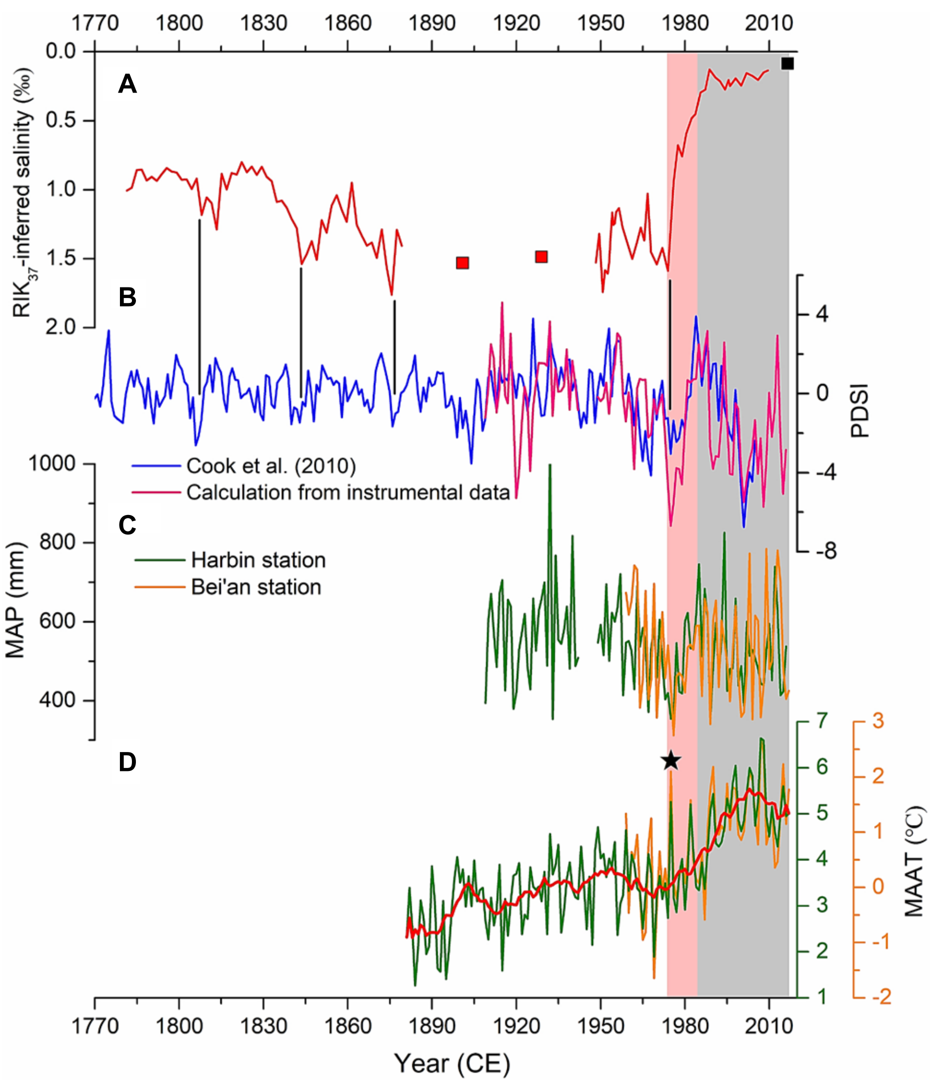


**Figure 2. Comparison between salinity changes reconstructed from a Lake Wudalianchi (northeastern China) sediment core and regional environmental conditions. (A) $RIK_{37}$-inferred salinity changes in the Lake Wudalianchi sediment core (we did not find detectable alkenones except for samples marked with red squares during 1880–1955 CE). Black square represents measured salinity (0.09‰) in the summer of 2016 (Yao et al., 2019a). (B) Palmer drought severity index (PDSI) in our study region (blue line, 47.5°N–50°N, 125°E–126.5°E) extracted from gridded PDSI data set during the past millennium (Cook et al., 2010). Another PDSI (pink line) is calculated based on instrumental data from Harbin station (Fig. 1B). (C) Instrumental mean annual precipitation (MAP) from the Harbin and Bei'an weather stations (Fig. 1B; Text S6 [see footnote 1]). (D) Instrumental mean annual air temperature (MAAT) from Harbin and Bei'an weather stations (Fig. S6; Text S6; red line represents 10-point running means for Harbin station; black star represents exceptionally high temperature in 1975). Red shaded area represents age range from 1975 to 1982; gray shaded area represents age from 1982 to 2016.**

diploptene from bacteria and iGDGTs from archaea, to reconstruct changes in $CH_4$ cycling regulated primarily by microbial methanotrophy and methanogenesis. Bulk organic $\delta^{13}C$ values range from −27.8‰ to −25.3‰ at our study site, showing a trend of increasing $^{13}C$ depletion from ca. 1975 to present (Fig. 3J). Our record shows a trend broadly similar to previously published data from two other Wudalianchi pools (Gui et al., 2012; Pool 3: −26.8‰ to −25.7‰; Pool 5: −24.9‰ to −22.3‰), although Pool 3 appears to display the most abrupt changes starting from ca. 1978 (Fig. S7A). The slight differences among the bulk $\delta^{13}C$ records from the three pools may be attributed to different pool size, depth, and volume; local hydrological factors; and sediment core dating uncertainties. Several factors may have contributed to the observed decrease in bulk organic matter $\delta^{13}C$ values since ca. 1975: (1) input to Lake Wudalianchi of $^{13}C$-depleted $CO_2$ from decomposing permafrost organic carbon in the permafrost soil (Elder et al., 2018), which is subsequently incorporated into aquatic biomass through photosynthesis and eventually sediments; (2) increased $CH_4$ fluxes (Paytan et al., 2015) leading to enhanced microbial methanotrophic biomass in the lake; (3) oxidation of $CH_4$ into $CO_2$ with low $\delta^{13}C$ values (Segarra et al., 2013), which is subsequently incorporated into the aquatic biomass through photosynthesis; and (4) direct input of dissolved and particulate organic matter with low $\delta^{13}C$ values from decomposing permafrost soils (Wild et al., 2019). Overall, the observed negative $\delta^{13}C$ excursion likely results from enhanced fluxes of $^{13}C$-depleted carbon and strengthening of the $CH_4$ cycle due to permafrost thaw.

Diploptene $\delta^{13}C$ values show an abrupt decrease, by >10‰, from 1982 (Fig. 3E). At the same time, diploptene flux increased by ~9× (Fig. 3D). The average $\delta^{13}C$ value of diploptene is −50.3‰ ± 1.9‰ after 1982, which is commonly associated with consumption of highly $^{13}C$-depleted biogenic $CH_4$ by methanotrophic bacteria (Pancost and Sinninghe Damsté, 2003; Davies et al., 2016; Elvert et al., 2016). We also estimated temporal changes in the contributions of methanotrophs to diploptene using a carbon isotopic mass-balance approach (Text S8). Our results show that ~13% of carbon in diploptene originates from methanotrophs before 1982, whereas the methanotroph contribution increases to ~38% after 1982 (Fig. 3F). This approximately three-fold increase in bacterial methanotrophic contribution to diploptene indicates substantially elevated $CH_4$ fluxes from the early 1980s.

The GDGT-0/crenarchaeol ratio, considered an indicator for methanogenic archaea (Blaga et al., 2009; Naeher et al., 2014), also shows an abrupt increase from 1982 to 1985, followed by a persistent (but slower rate of) increase to the present (Fig. 3H). Similarly, GDGT-0 fluxes increased since 1982, and continue to rise until the present (Fig. 3G). The enhanced biomarker fluxes of archaeal methanogenesis since 1982 observed in our record may reflect (1) an increase in $CH_4$ flux to Lake Wudalianchi from decomposing organic matter in permafrost soils (Paytan et al., 2015); (2) enhanced flux of methanogenic archaeal lipids to the lake from thawing permafrost soils; and (3) enhanced methanogenic activity within the lake, given that increased supply of allochthonous organic matter from permafrost soils and enhanced lake primary productivity provide additional substrates for methanogenesis (Fig. 3K). Because the lake does not become anoxic after permafrost thawing, as indicated by our HP5 index as a redox proxy (Fig. 3I; Yao et al., 2020b), the positive $\delta^{15}N$ trend also mainly reflects increased lake primary productivity (Fig. 3J; Text S9). Our recent study of surface sediments, suspended particulate matter, and surrounding surface soils in volcanic lakes in northeastern China indicate that iGDGTs in Lake Wudalianchi are derived primarily from in situ production of lakes (Yao et al., 2019b). Thus, the enhanced archaeal methanogenesis mainly reflects increased contribution from autochthonous sources in modern times.

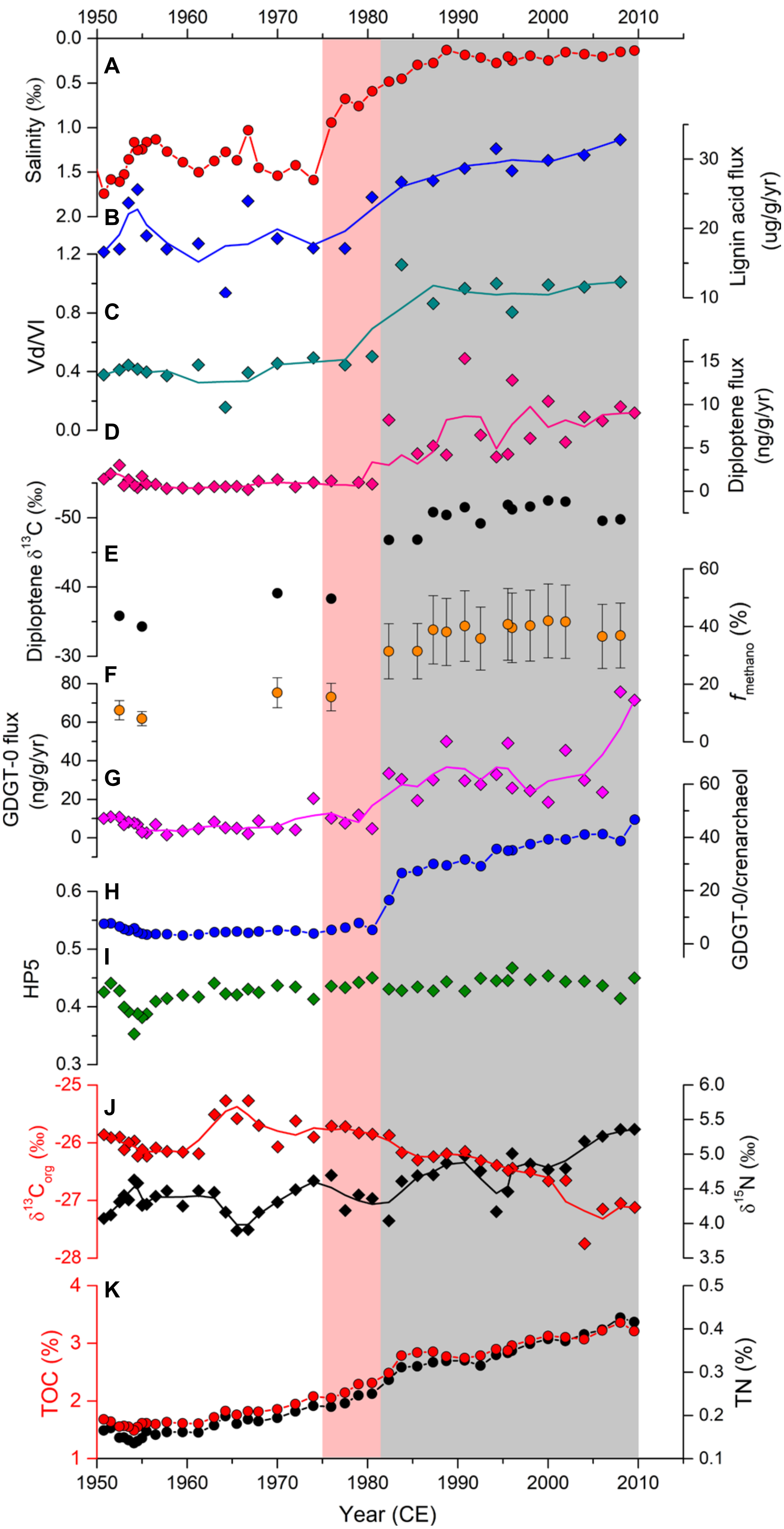


**Figure 3. Comparison of reconstructed salinity changes and carbon and nitrogen cycling from Lake Wudalianchi (northeaster China) sediment core. (A) $RIK_{37}$-inferred salinity. (B) Lignin acid flux (line is two-point running mean). (C) Vd/Vl (vanillic acid/vanillin) ratio (two-point running mean). (D) Diploptene flux (two-point running mean). (E) Diploptene $\delta^{13}$C values (before 1980 CE, most samples have very low concentrations of diploptene that are insufficient for carbon isotope measurements). (F) Average contributions of methanotrophic bacteria to diploptene ($f_{methano}$) using carbon isotopic mass-balance equation (see Text S8 [see footnote 1]). (G) Glycerol dialkyl glycerol tetraether 0 (GDGT-0) flux (two-point running mean). (H) GDGT-0/crenarchaeol ratio. (I) HP5 index. (J) Bulk organic matter $\delta^{13}$C values and bulk $\delta^{15}$N values (two-point running means). (K) Total organic carbon (TOC) and total nitrogen (TN). Red shaded area represents age range from 1975 to 1982; Gray shaded area represents age from 1982 to 2010.**

We note that there is a 5–7 yr delay in the major rises in methanogenic and methanotrophic activities relative to lake water freshening and bulk organic $\delta^{13}$C decrease in our Wudalianchi records (Fig. 3). We attribute this delay to the progressive deepening of the permafrost active layer starting from ca. 1975 and the vertical profile of redox potential in the decomposing organic-rich soils. As permafrost starts to thaw and the active layer deepens, the greenhouse gases released from decomposition gradually change from those with a relatively high $CO_2/CH_4$ ratio (when a sufficient amount of oxygen is available to serve as the dominant electron acceptor in shallower soils) to those with a relatively low $CO_2/CH_4$ ratio (when thawing reaches deeper soil where soil water is depleted in oxygen) (Hodgkins et al., 2014). This change in gas composition may have taken ~5–7 yr to occur, resulting in our observed timing difference in the bulk organic $\delta^{13}$C excursion and rapid ramping up of the $CH_4$ cycle in Lake Wudalianchi.

In summary, our biomarker proxies from Lake Wudalianchi indicate abrupt changes in hydrology and $CH_4$ cycling as regional permafrost thaws. Enhanced $CH_4$ cycling due to warming of permafrost soils has been previously reported in high-latitude lake sediment records (e.g., Wooller et al., 2012; van Hardenbroek et al., 2013; Elvert et al., 2016). Our analysis further demonstrates the important role that lakes play in ventilating greenhouse gases to the atmosphere as mid- to high-latitude warming speeds up, which would further form a positive feedback cycle and amplify global warming.


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