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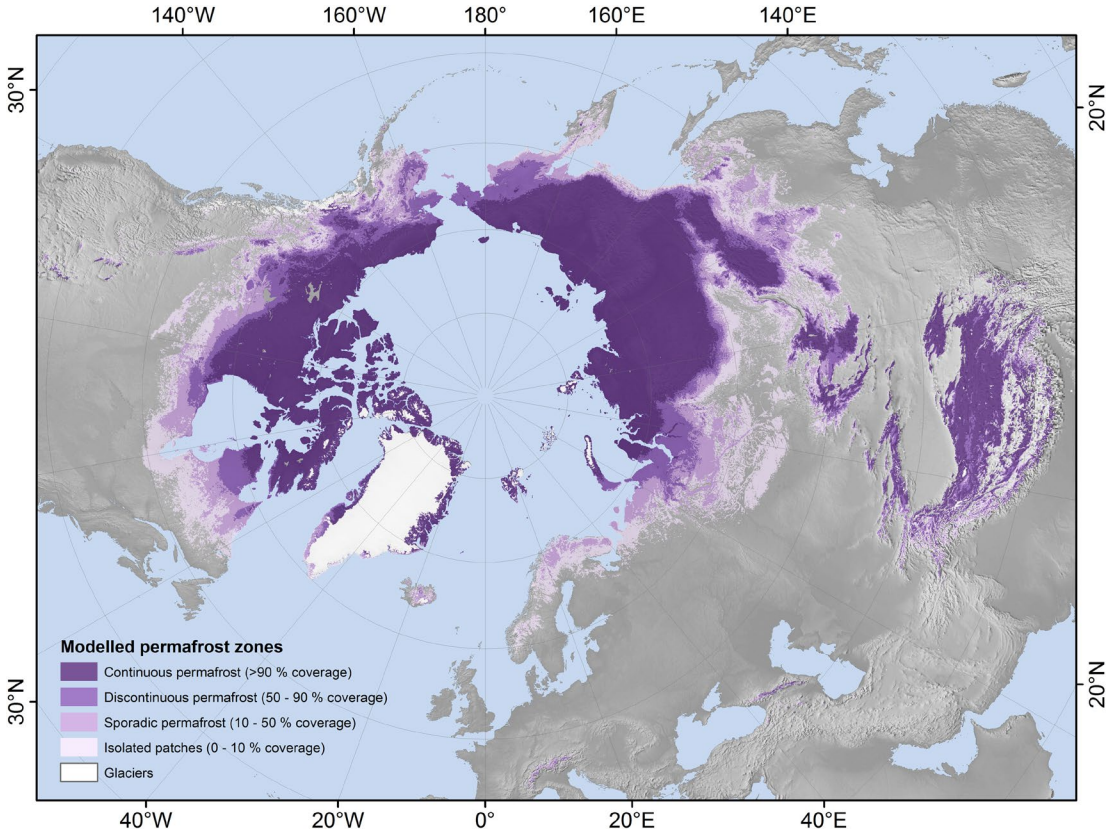
Microbial Mercury Methylation in Tibetan Thermokarst Lakes

Xia Lu, Xiaosha Liu, Liang Li, Zhixia Yang, Cuicui Mu, Man Yang, Yurong Liu, Baohua Gu

College of Earth and Environmental Sciences, Lanzhou University
College of Resources and Environment, Huazhong Agricultural University
Environmental Sciences Division, Oak Ridge National Laboratory

Permafrost stores a significant amount of Hg



- Permafrost is widespread in the Northern Hemisphere; permafrost region ~20.8×10^6 km^2 (**22%** of exposed land area)
- The high latitudes and the high altitudes
- The Tibetan Plateau is the largest alpine permafrost region in the world (**8%** of permafrost area in the Northern Hemisphere)

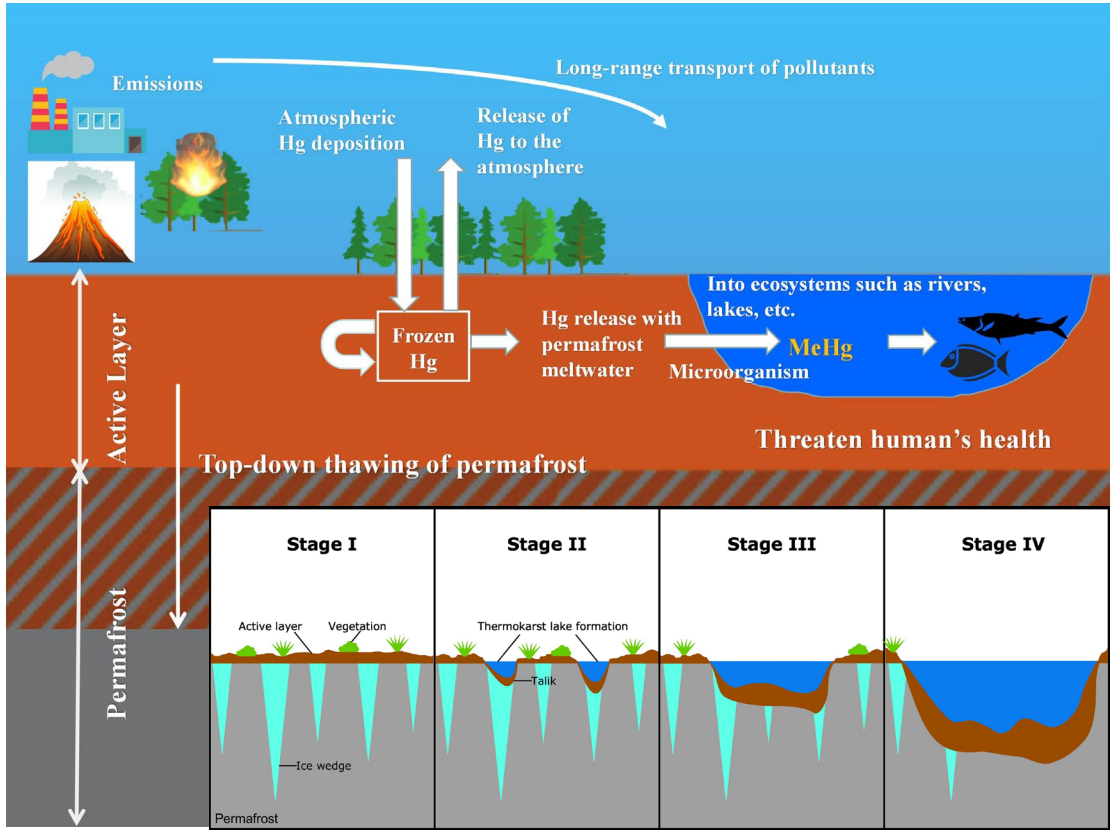
(Obu et al., 2019; Zou et al.,2017)

Permafrost constitutes the largest long-term reservoir of Hg (by long-range transport)

- 597 Gg (384~750 Gg, IQR) of Hg in 0~300 cm pan-Arctic permafrost soils
- 21.7 Gg (18.4~24.2 Gg) of Hg in 0~300 cm Tibetan Plateau permafrost soils

(Lim et al., 2020; Mu et al., 2020)

Permafrost degradation and thermokarst lakes formation



Ice wedge melting

↓

Subsidence, collapse or erosion of the surface

↓

Fill with water, thermokarst lakes form

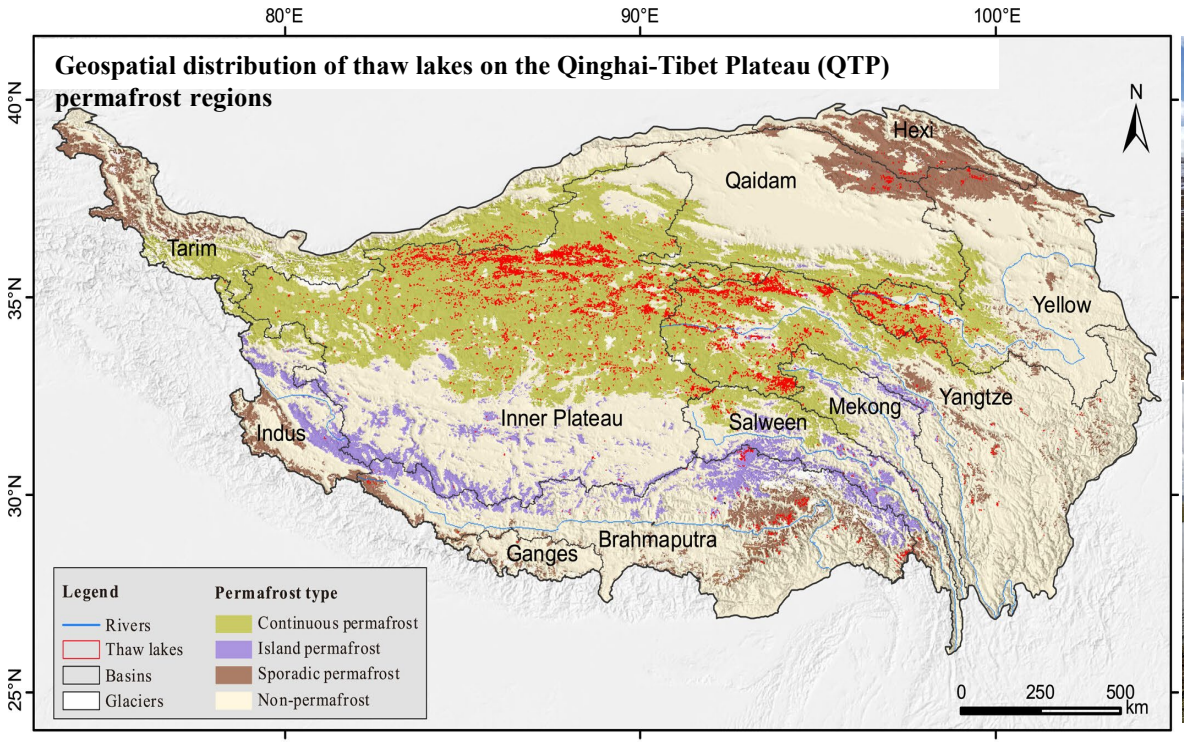
↓

Horizontal and vertical lake expansion

- At climate-sensitive high-latitude and high-elevation regions, air temperatures have risen about **twice** as fast as the global average in recent decades, resulting in widespread permafrost thaw and degradation.

(Schuster et al.,2018; Sun et al.,2023)

Tibetan Thermokarst Lakes

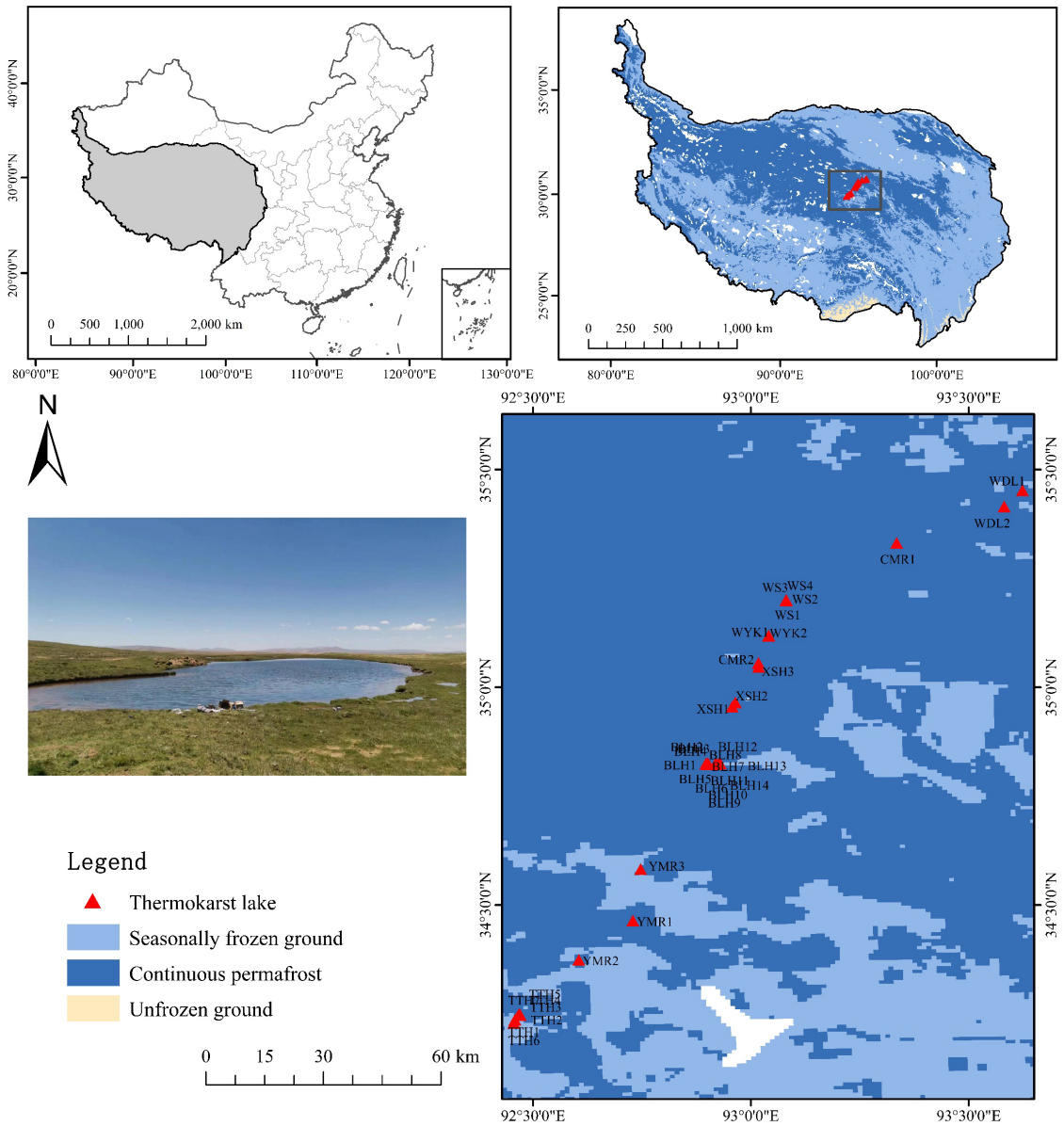


- The Tibetan Plateau is known as the third pole of Earth and the water tower of Asia.
- In recent decades, with global warming, the permafrost area on the Tibetan Plateau has decreased by 20%.There are ~**161,300** thermokarst lakes with sizes of 500 m^2 ~3 km^2.

(Wei et al., 2021; Cheng et al., 2019)

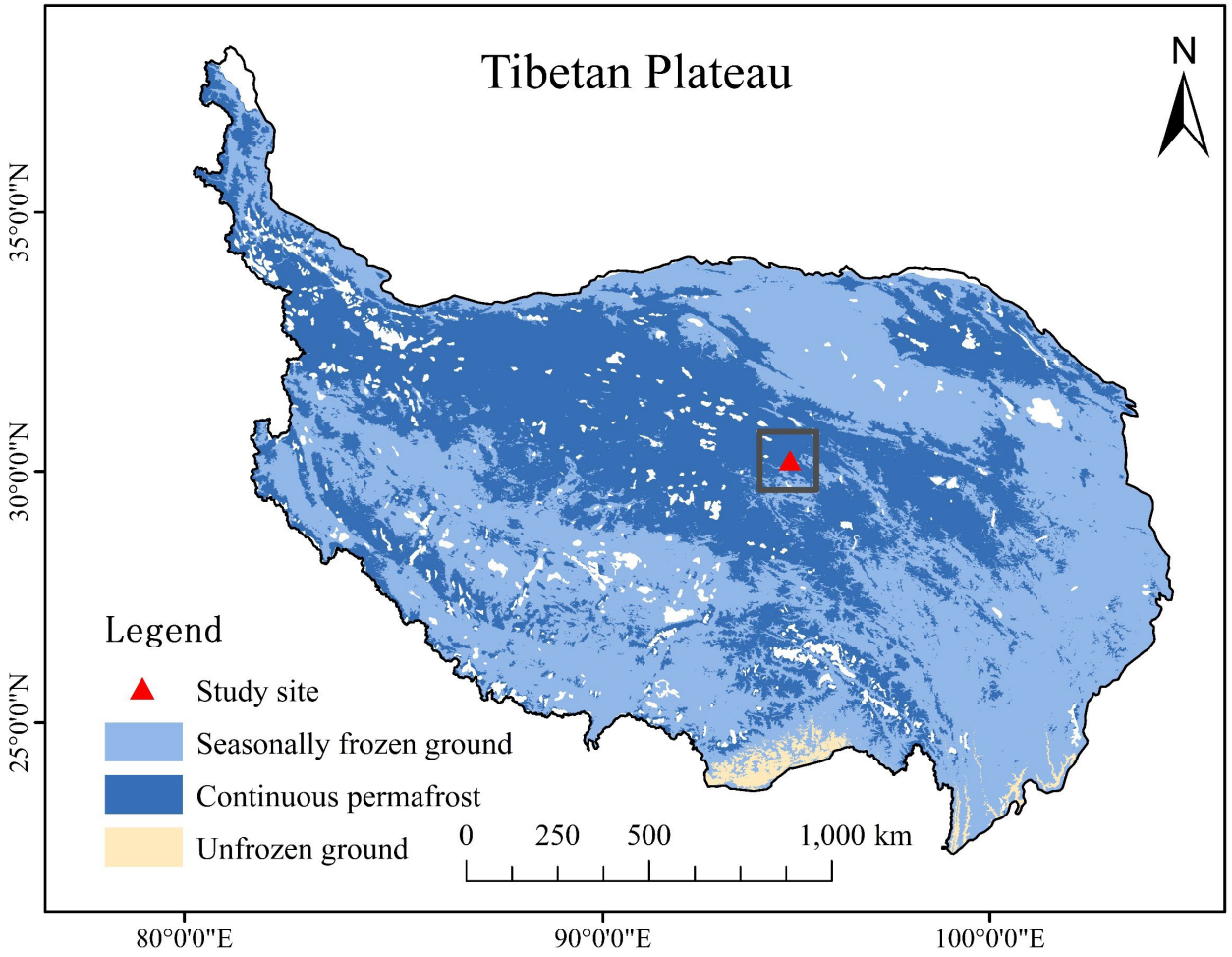
This work aims to investigate the effects of thermokarst lakes formed by permafrost thaw on Hg methylation in Tibetan Plateau ecosystems.

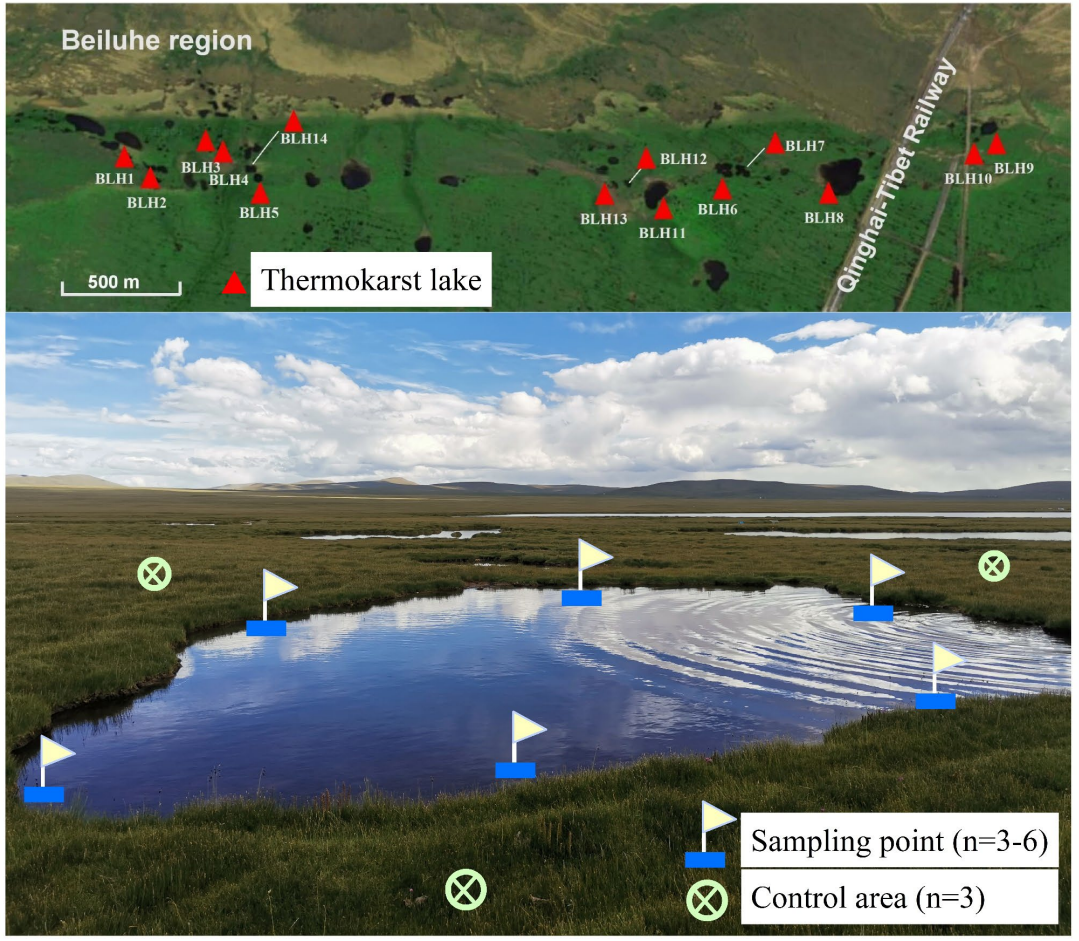
Study site and field sampling



- Continuous permafrost regions in central Tibetan Plateau
- Altitude: 4400-4800 m
- Mean annual air temperature: -5.0 °C to -3.8 °C
- Thermokarst lake water and sediment (top 10 cm)
- Ice-free period (Jun, Jul, Sep, Oct) in 2020 (n=37), and July in 2023 (n=14)

Study site and field sampling





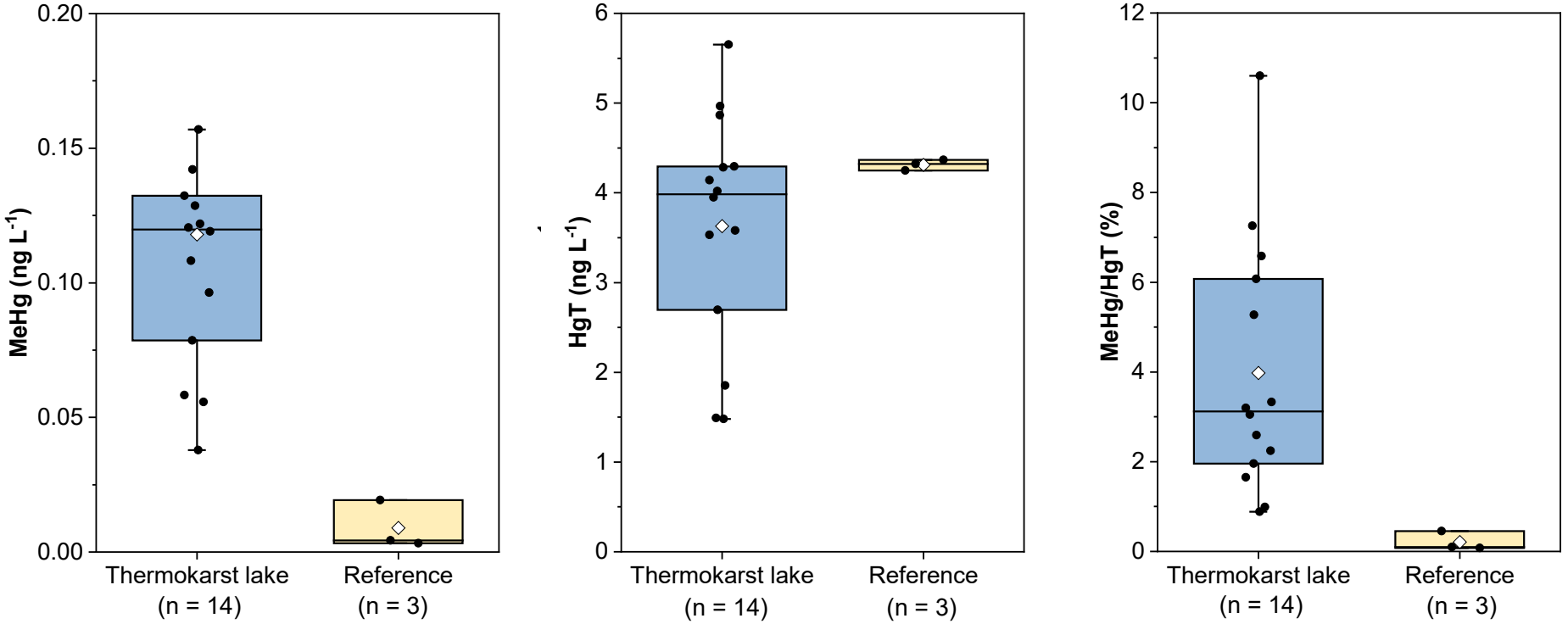
Hg analyses and sediment microcosm incubation

Brooks Rand MERX system

Anaerobic incubation (0-12 days, 10 °C)

- **MeHg in sediment/water**: KBr-H_2SO4-$CuSO_4$-CH_2Cl_2 method/distillation
- **HgT in sediment/water** : concentrated HCl and HNO_3 (v/v = 4:1)/BrCl
- **Incubation**: unamended control, Hg, Hg+BES (2-bromoethanesulfonate), Hg+Mo (molybdate)

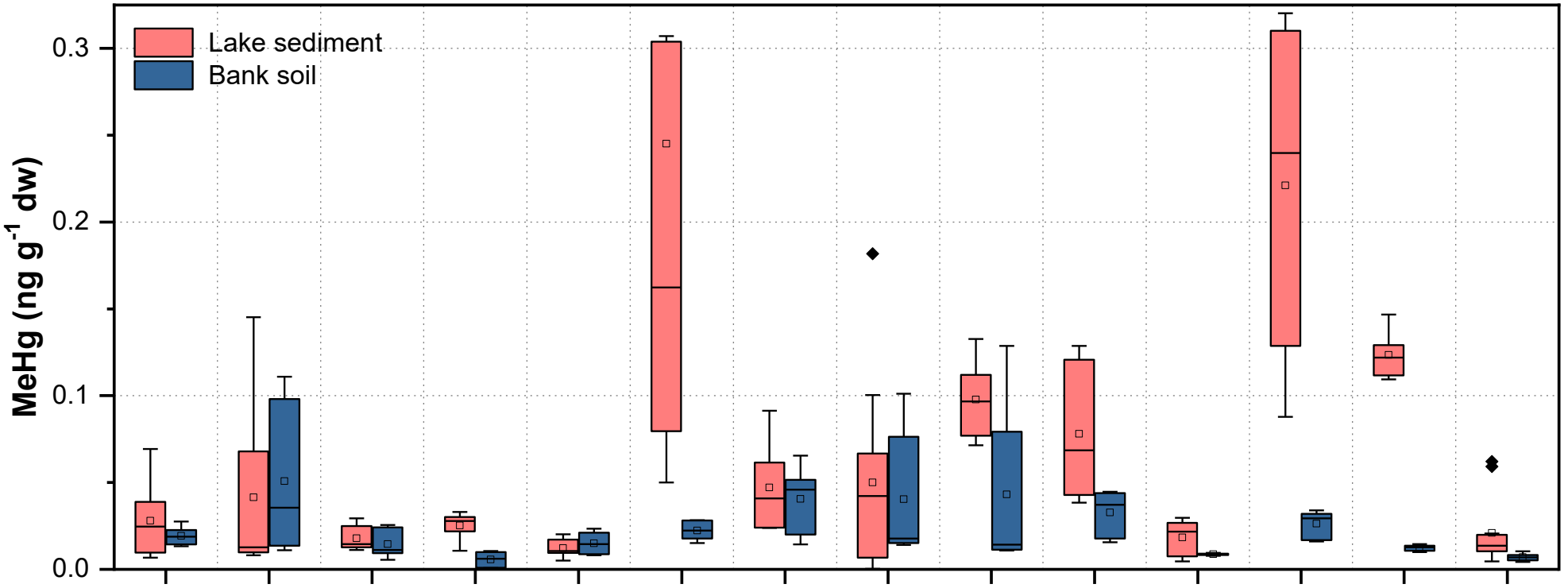
Hg concentration in thermokarst lakes water



	Thermokarst lake	Control	Compare
MeHg (ng L^{-1})	0.118 (0.062)	0.009 (0.009)	*13 × higher (P<0.01)*
HgT (ng L^{-1})	3.63 (1.30)	4.31 (0.06)	*NS*
MeHg/HgT (%)	3.98 (2.81)	0.21 (0.21)	*19 × higher (P<0.05)*

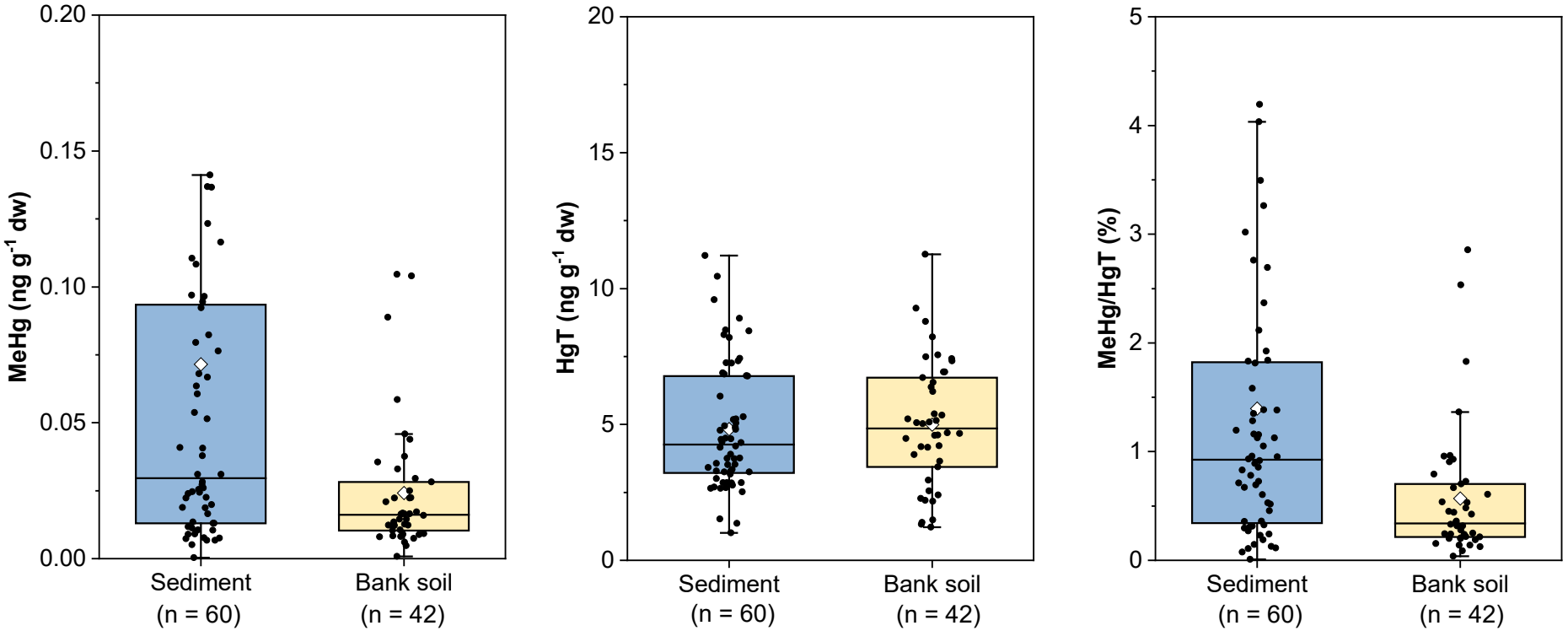
(Lu et al., 2024, in prep)

MeHg concentration in thermokarst lakes sediment



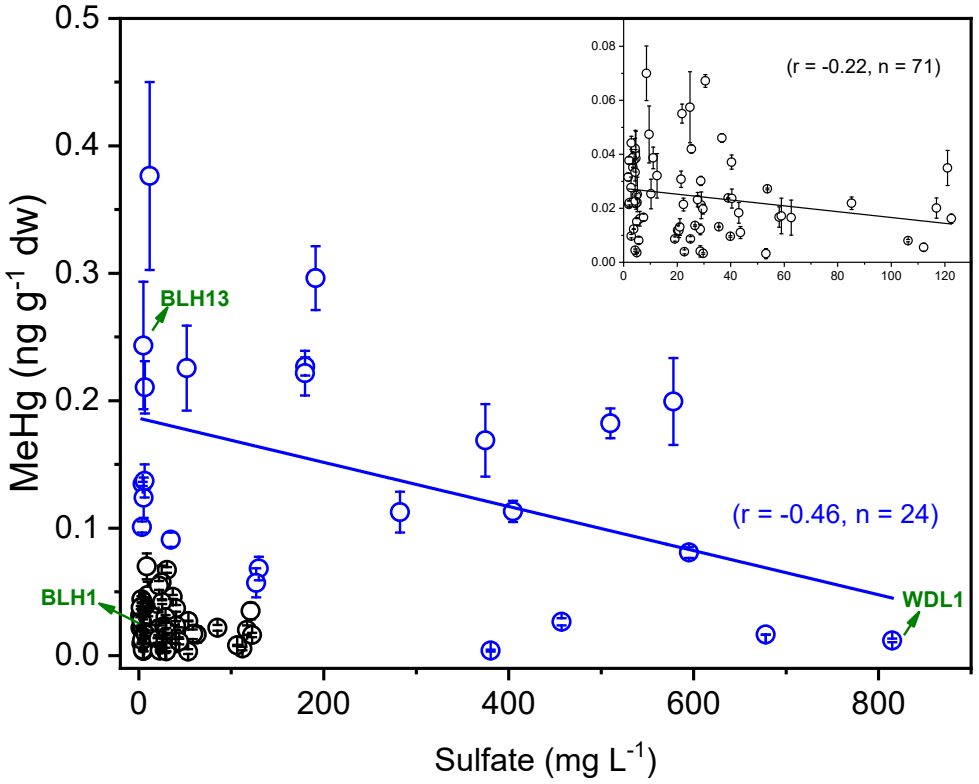
- Large differences in MeHg concentrations among 3-6 sampling points in each thermokarst lake, due to spatial heterogeneity
- For most thermokarst lakes, MeHg concentration in lake sediments were higher than in bank control areas

Hg concentration in thermokarst lakes sediment



	Thermokarst lake	Control	Compare
MeHg (ng g^{-1} dw)	0.072 (0.109)	0.024 (0.024)	*3 × higher (P<0.01)*
HgT (ng g^{-1} dw)	4.83 (2.32)	5.00 (2.34)	*NS*
MeHg/HgT (%)	1.39 (1.50)	0.57 (0.60)	*2.5 × higher (P<0.01)*

Sediment MeHg concentration and water sulfate concentration

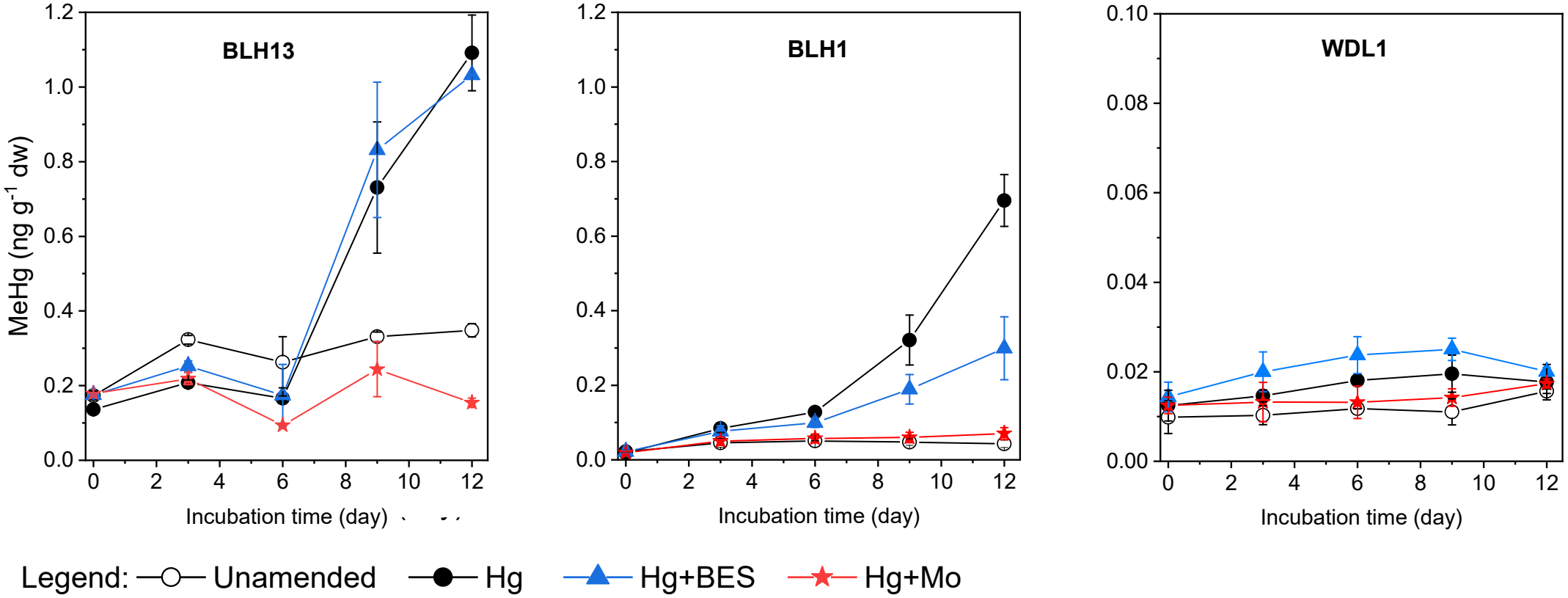


- MeHg concentration in thermokarst lake sediment negatively correlated with sulfate concentration in lake water
- **BLH13, BLH1, WDL1** for following sediment microcosm experiments

Sediment MeHg concentration and water sulfate concentration

Thermokarst lake	Vegetation cover	Area	Water sulfate	CH_4	Sediment OC	Sediment water content	MeHg	HgT
		m^2	mg L^{-1}	mmol·m^{-2}·d^{-1}	%	%	pg g^{-1} dw	ng g^{-1} dw
BLH13	alpine meadow	399	5.03	10.77	0.47	33	243.4	11.17
BLH1	alpine wet meadow	5793	4.99	18.61	0.43	30	22.1	3.46
WDL1	alpine desert steppe	5702	814	0.08	0.1	9	11.9	3.12

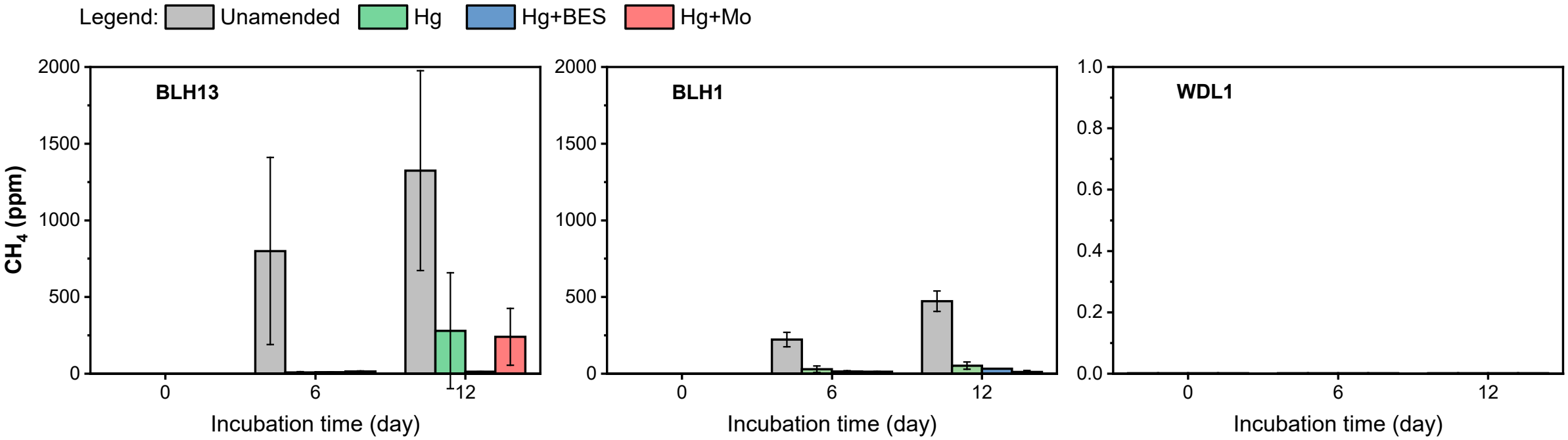
Hg methylation potential in thermokarst lake sediment



- BLH13: Hg (8×), Hg-BES (no effect), Hg-Mo (100% inhibition)
- BLH1: Hg (38×), Hg-BES (60% inhibition), Hg-Mo (100% inhibition)
- WDL1: changed slightly, Hg-Mo (inhibition)

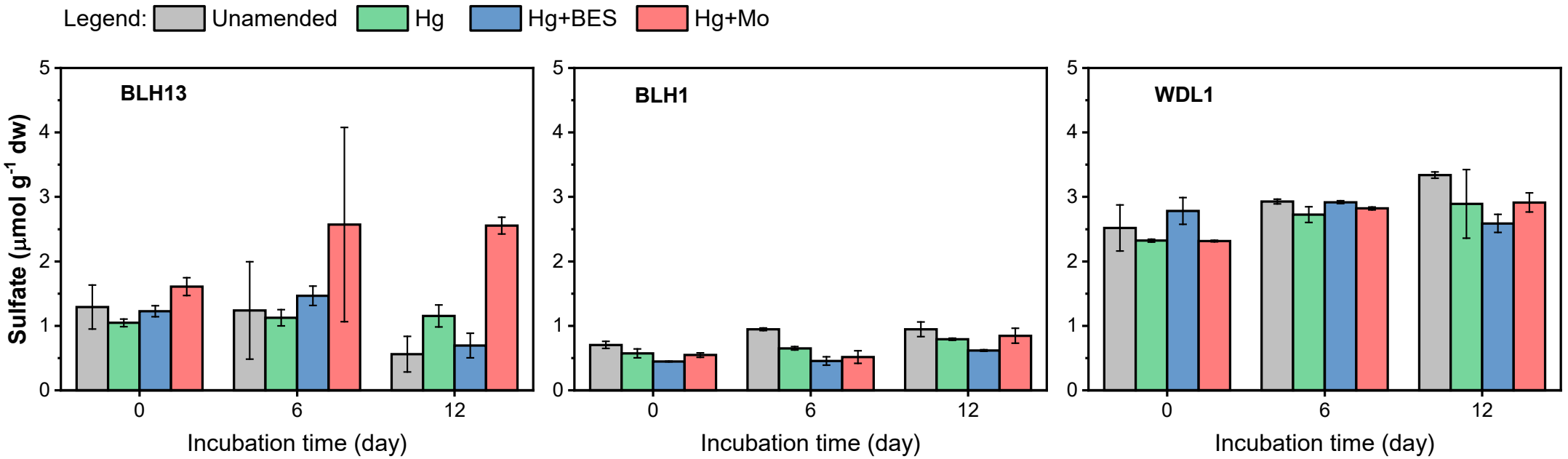
(Lu et al., 2024, in prep)

Methane production



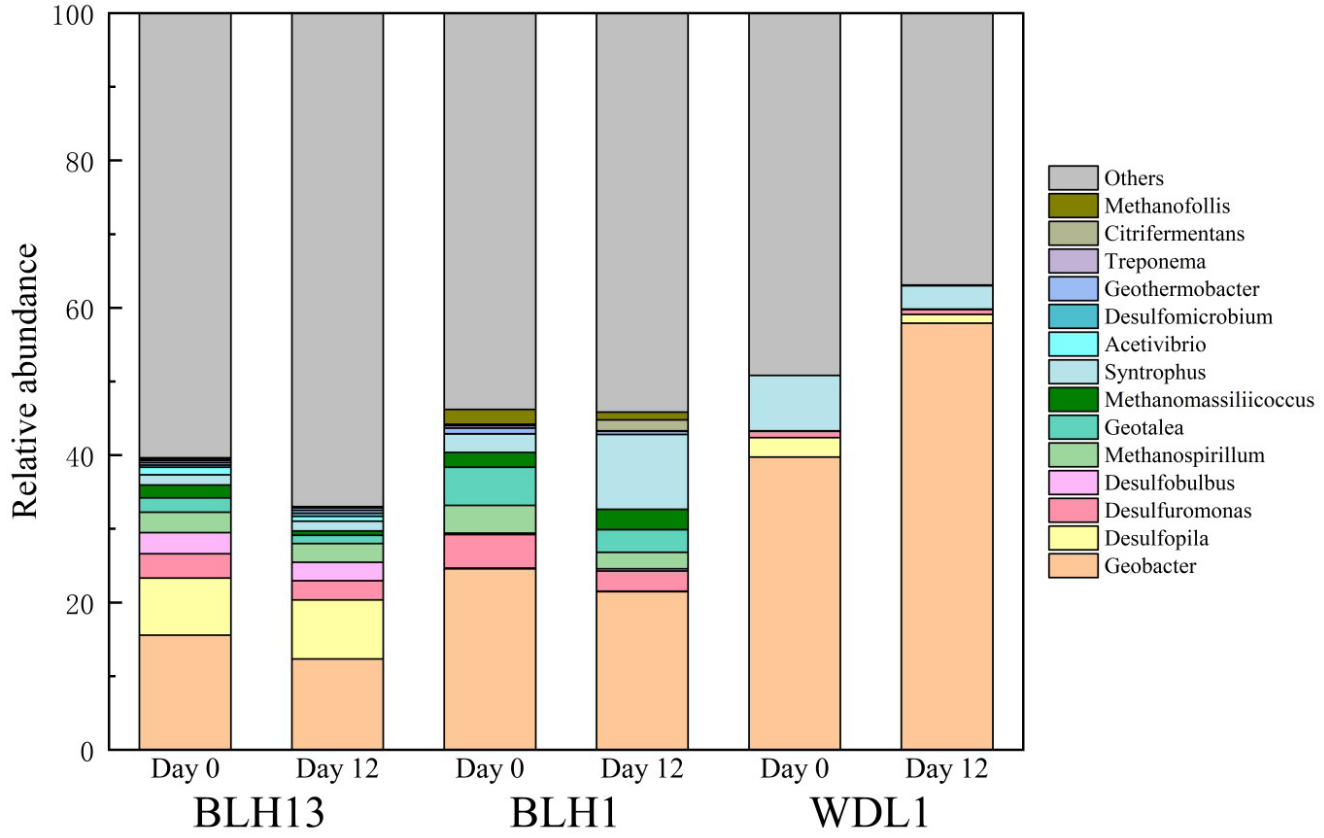
- In the BES-amended incubation, methane production was completely stopped.

Sediment sulfate concentration



- The sulfate concentration increased in molybdate-amended incubation, rather than decreased, due to molybdate addition inhibits sulfate reduction.

Dominant Hg methylators



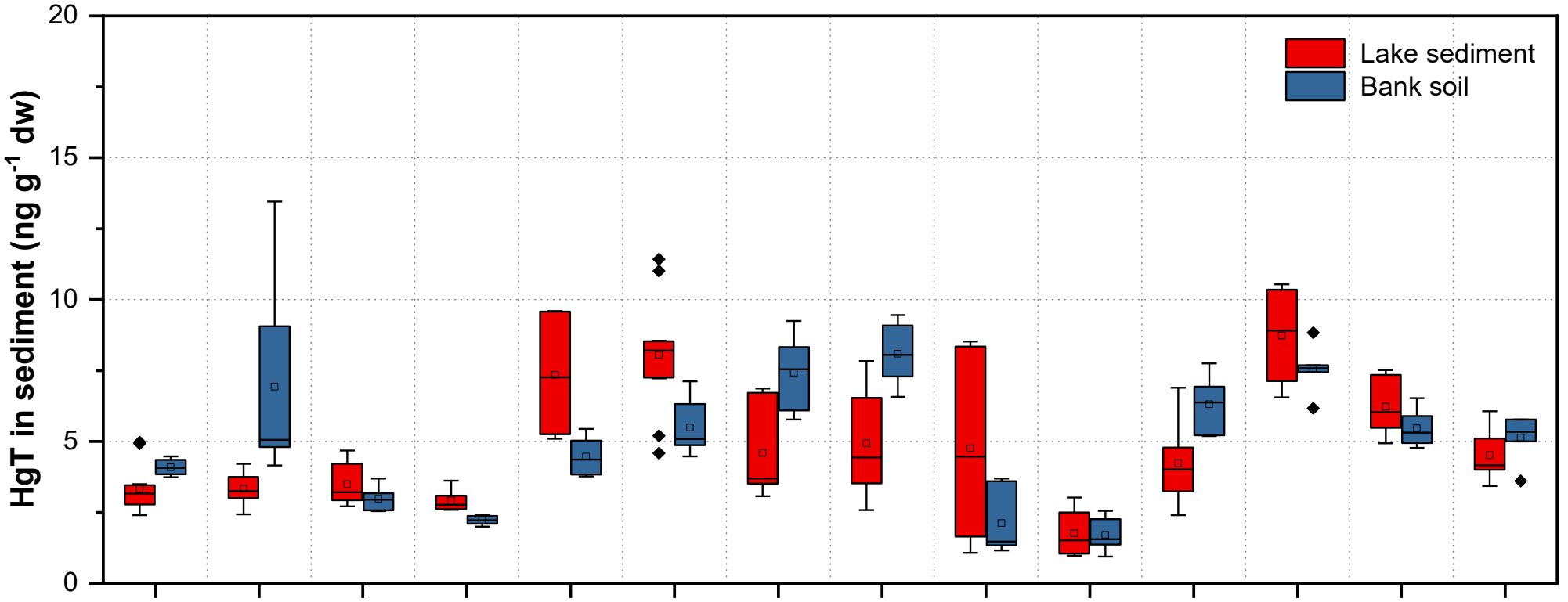
- Geobacter and SRB (*Desulfopila*, *Desulfuromonas*, *Desulfobulbus*) were the top abundant genus

Conclusions

- In Beiluhe region in central Tibetan Plateau, MeHg concentration in the thermokarst lakes is **an order of magnitude higher** than that of other aquatic ecosystems, and MeHg concentration in sediments is **3 times higher** than that in bank soils, while HgT concentrations are **comparable**.
- **Low sulfate** thermokarst lakes (BLH13, BLH1) show high Hg methylation potential, converting **7%** of new added Hg(II) to MeHg at day 12, whereas high sulfate lake (WDL1) shows little methylation potential.
- Molybdate addition was effective in inhibiting Hg(II) methylation in all three lakes, indicating that **sulfate-reducing bacteria** were the dominant Hg(II) methylators.
- Our study reveals that thermokarst lakes formed by permafrost degradation on the Tibetan Plateau could potentially lead to **elevated MeHg levels** in Tibetan ecosystems and may thus pose health risks to humans and wildlife, particularly under future climate warming scenarios.

Supplementary Materials

HgT concentration in thermokarst lakes sediment



HgT concentration