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Microbial mercury methylation in paddy soils

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16th ICMGP, Cape Town, South Africa

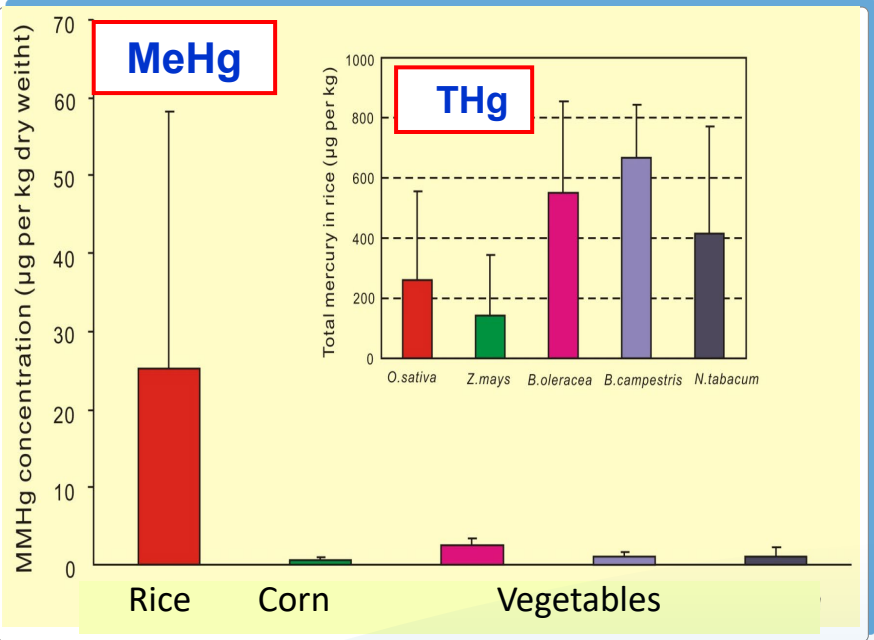


Outline

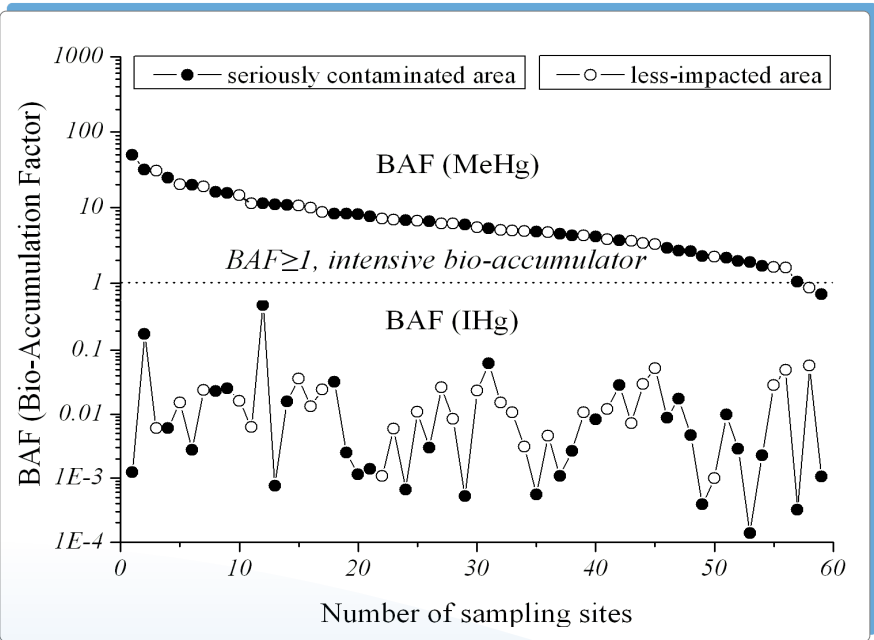
- Issue of MeHg contaminated rice and related human health risks
- Hg methylation in rice paddy ecosystem
- Challenges and future research

Rice has been highlighted as a bioaccumulator plant species of MeHg

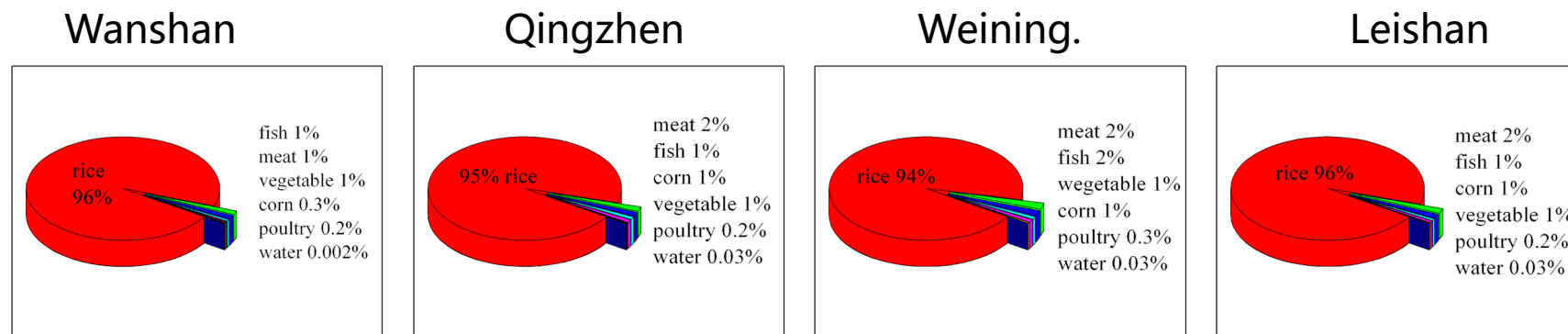
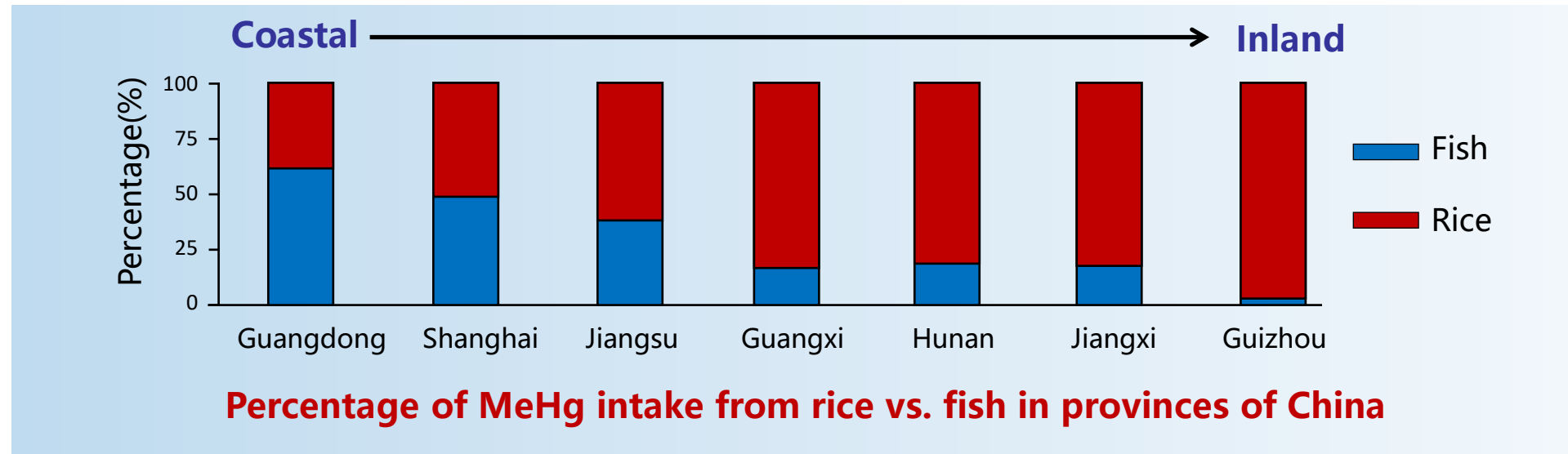
THg and MeHg concentrations in crops



Bio-accumulation factors of MeHg and IHg in rice

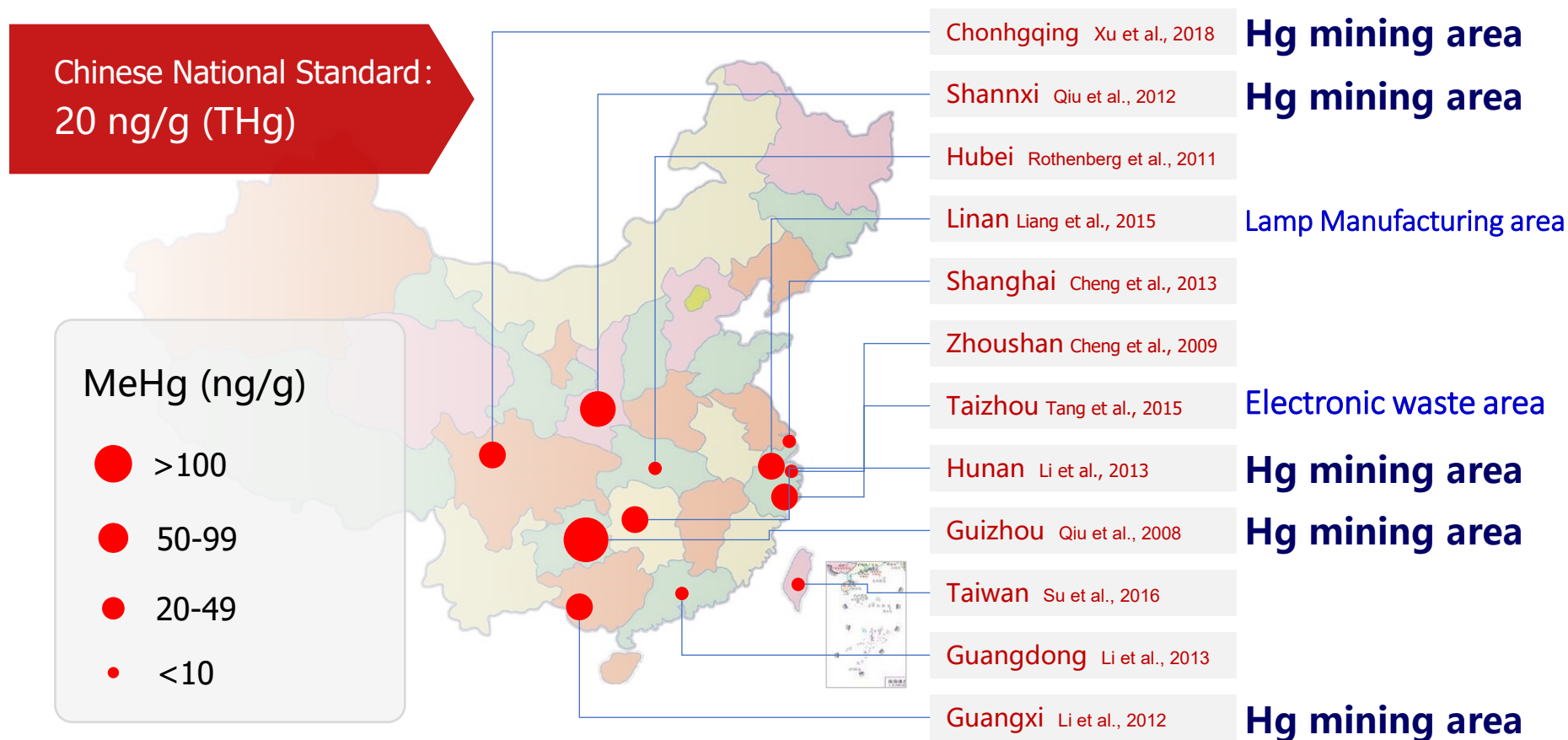


Rice, rather than fish, is the major pathway for MeHg exposure in inland China



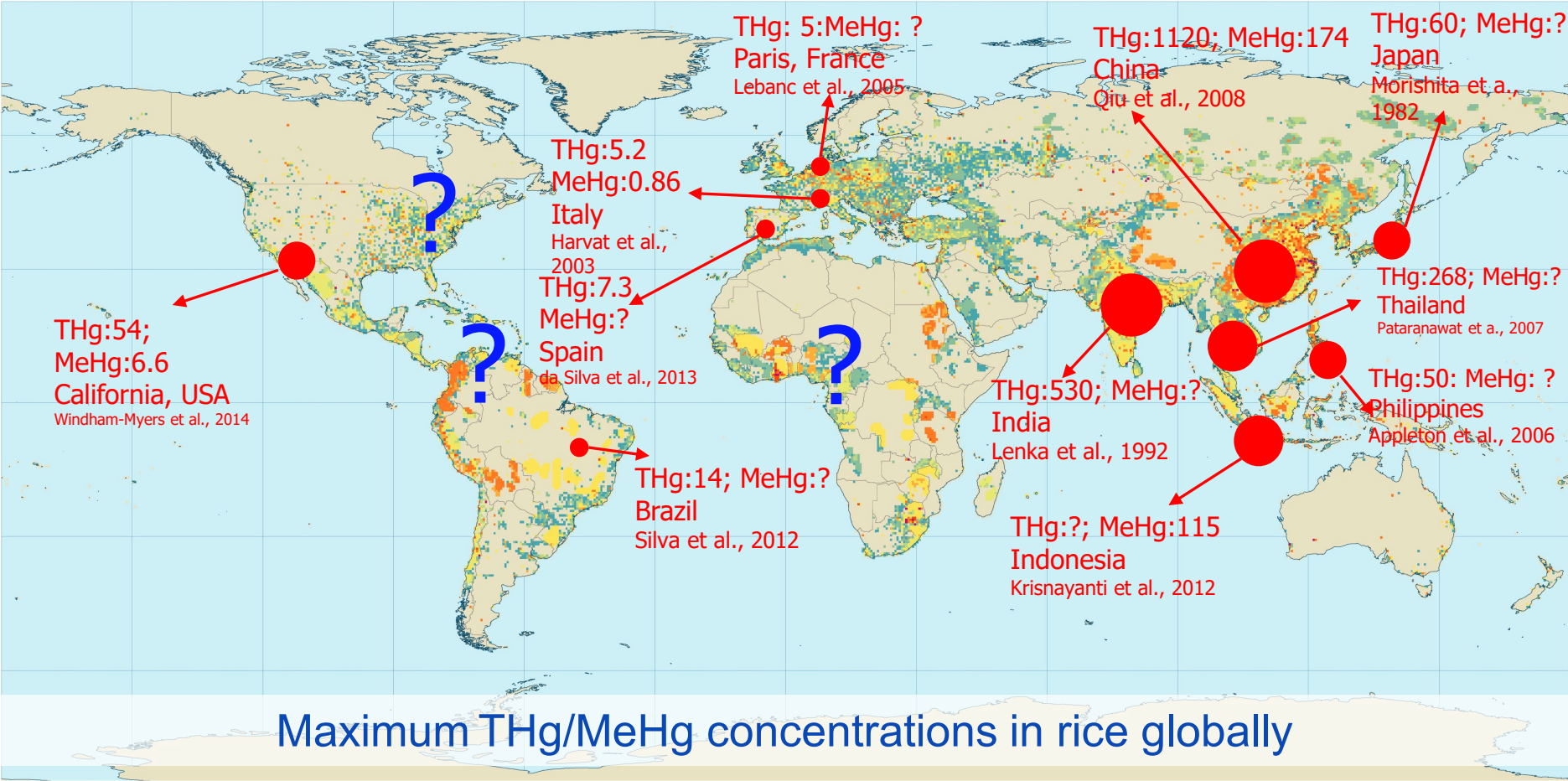
Percentage of MeHg intake from different media in Guizhou, China

MeHg contamination in rice is a common phenomena in Hg polluted area



Maximum MeHg cons. in rice from provinces of China

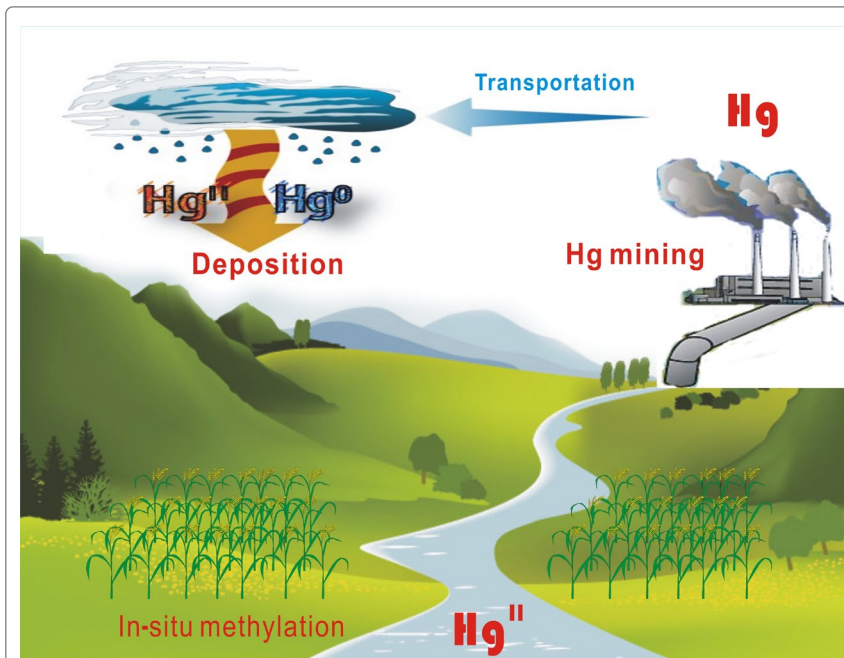
Hg bioaccumulation in rice is a worldwide issue, and need to be paid enough attention



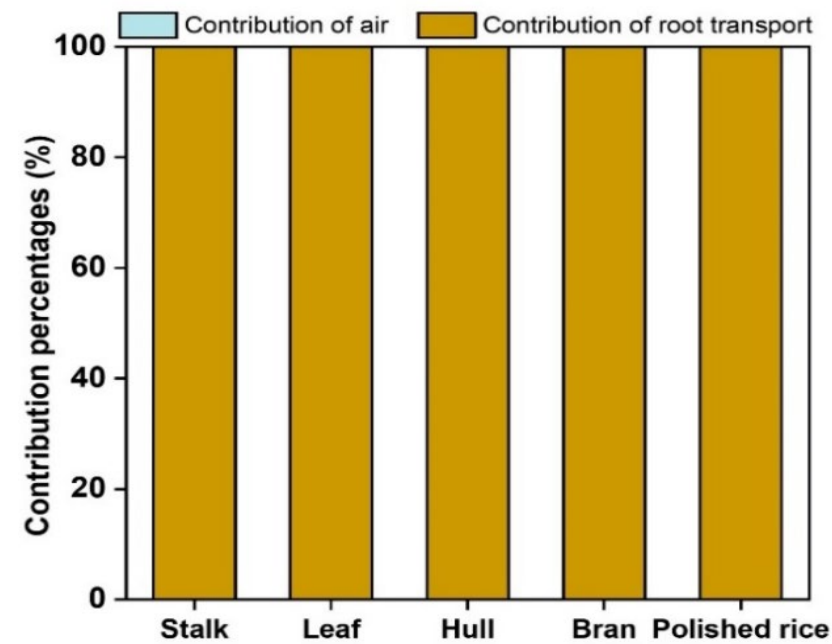
Permissible limit : 20 ng/g (Chinese National Standard Agency)

Source of MeHg in rice

- ◆ Paddy soil is a unique source of MeHg in rice
- ◆ MeHg formed in paddy soil via microbial methylation, then accumulated in rice



MeHg Flux of different vectors to soil MeHg pool



Sources MeHg in tissues of rice plant

Hg methylation in rice paddy ecosystem

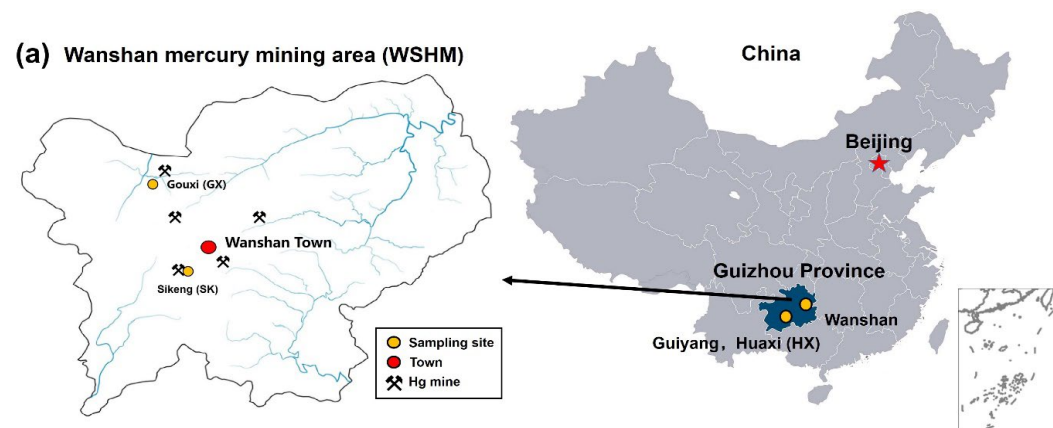
Major knowledge gaps

- (1) Geochemically relevant Hg^{II} species that can be methylated in paddy soils?
- (2) The relative importance of microbial groups on MeHg production and degradation in paddy soils
- (3) The key factors in regulating MeHg formation in paddy soils

Prevent the development of remediation measures necessary to ensure safe consumption of this staple food

Study area and sampling sites

Wanshan Hg mining area: large scale and long mining history



(1) Control site (HX)

(2) Artisanal Hg smelting site (GX)

(3) Abandoned Hg mining site (SK)



Artisanal Hg smelting site (2007)



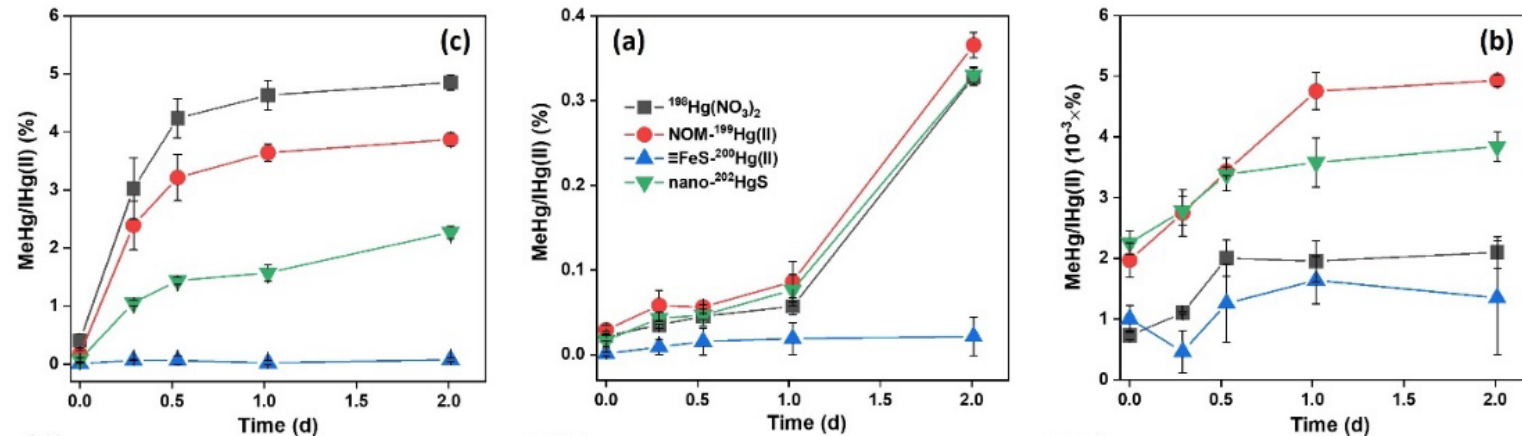
Abandoned Hg mining site (2008)



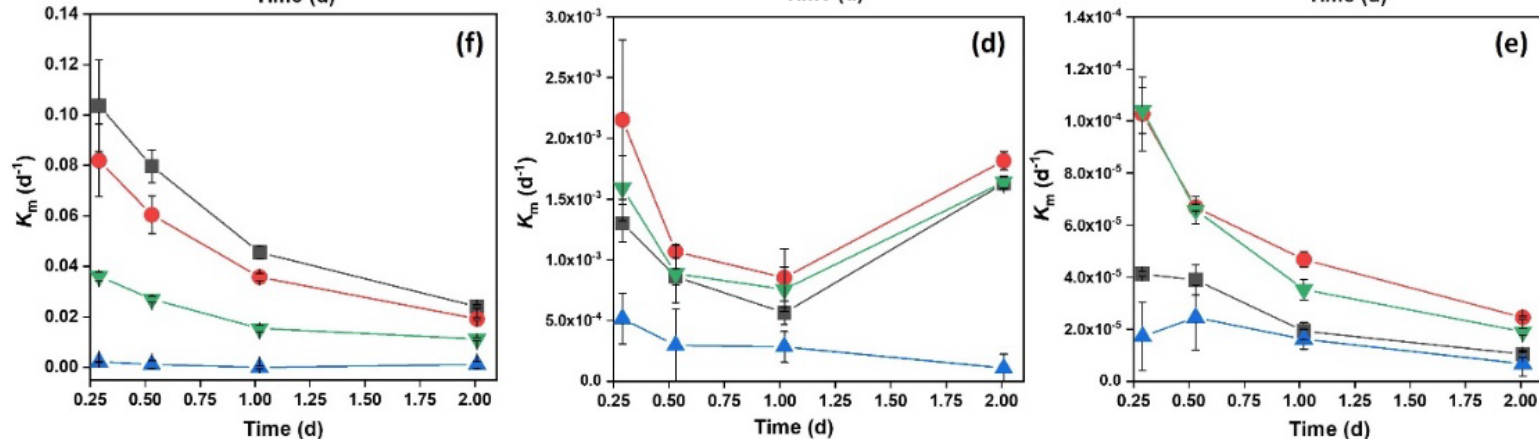
(1) Geochemically relevant Hg^{II} species that can be methylated in paddy soils?

- Methylation rates of NOM-¹⁹⁹Hg^{II} and nano-²⁰²HgS as substrates were similar or greater than rate obtained using the labile ¹⁹⁸Hg(NO₃)₂ substrate
- Highlight the role of NOM-Hg^{II} and nano-HgS as substrates for methylation in paddy soils

MeHg production Rate
(MeHg/Hg(II))



Methylation rate
(K_m)



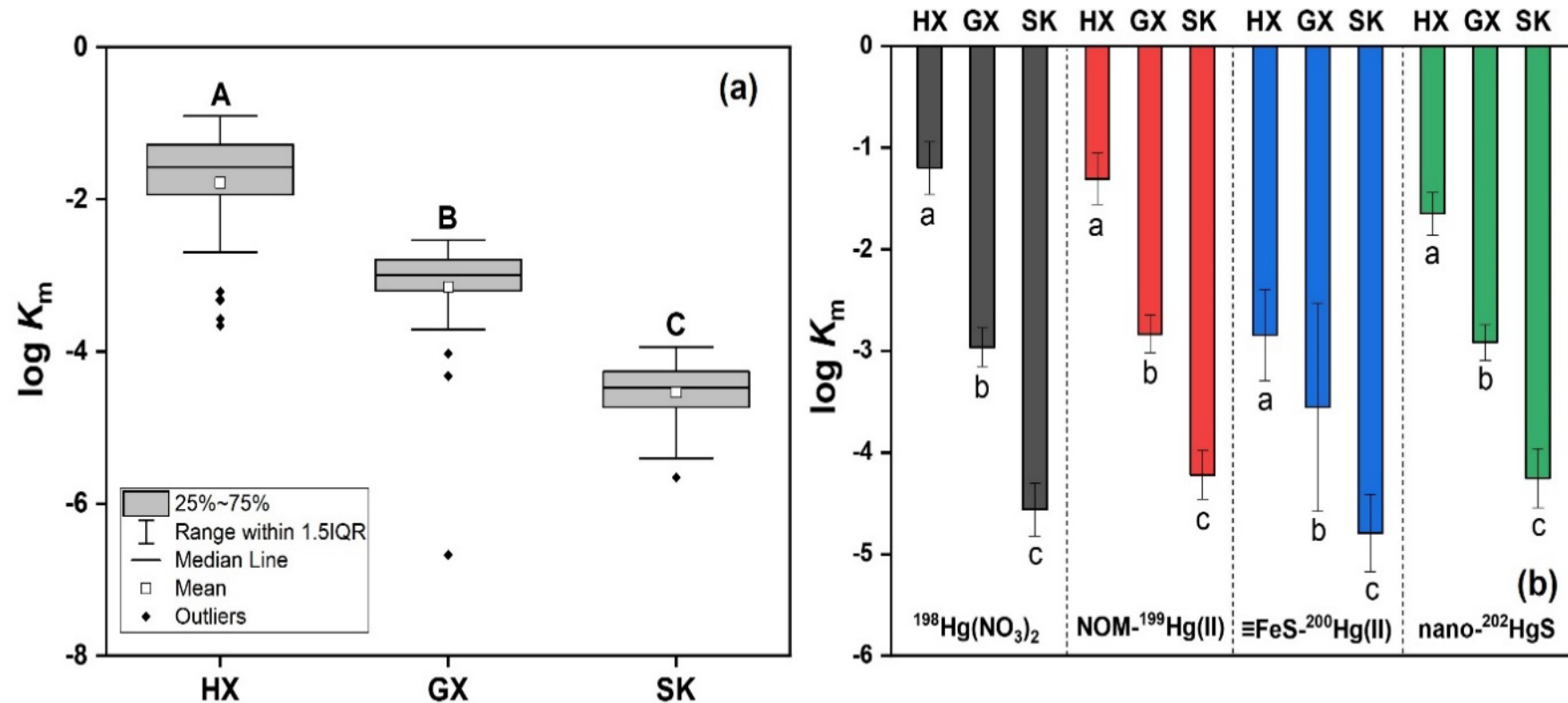
Control site

Artisanal Hg mining site

Abandoned Hg mining site

(1) Geochemically relevant Hg^{II} species that can be methylated in paddy soils?

- The variability in the methylation data for a given site was smaller than what was observed across the Hg concentration gradient

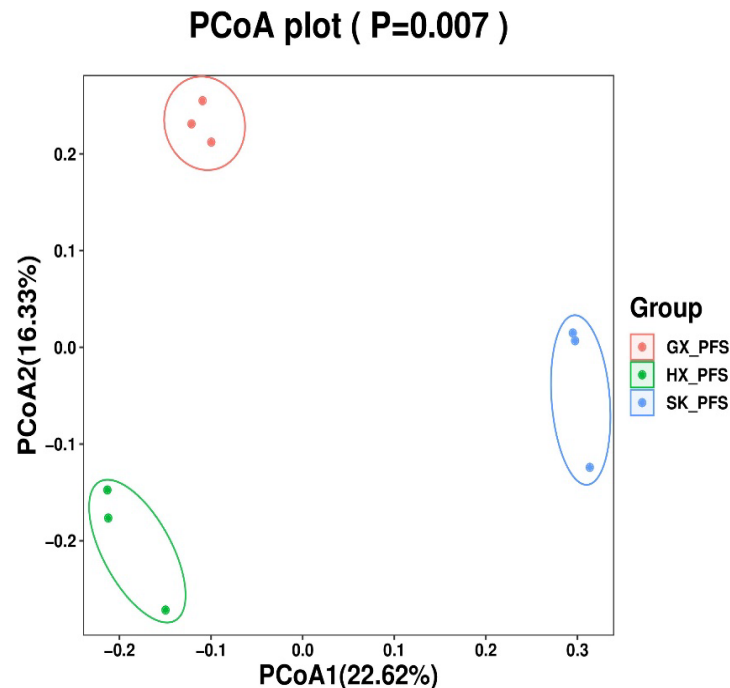


Log-transformed methylation rate constants

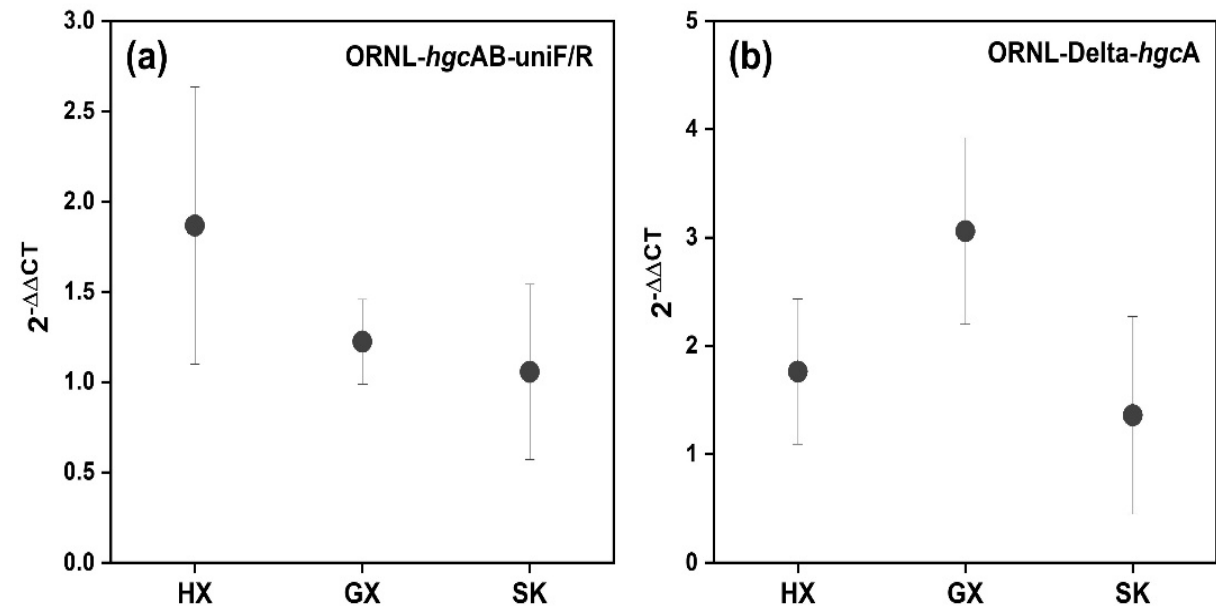
(1) Geochemically relevant Hg^{II} species that can be methylated in paddy soils?

Implications:

At broad spatial scales, site-specific characteristics, such as microbial community structure, need to be taken into consideration, alongside the nature of the Hg substrate available for methylation, to determine net MeHg production



PCoA for 16S rRNA gene sequencing



Relative gene expression for *hgcAB* (a) and *hgcA* in Deltaproteobacteria (b)

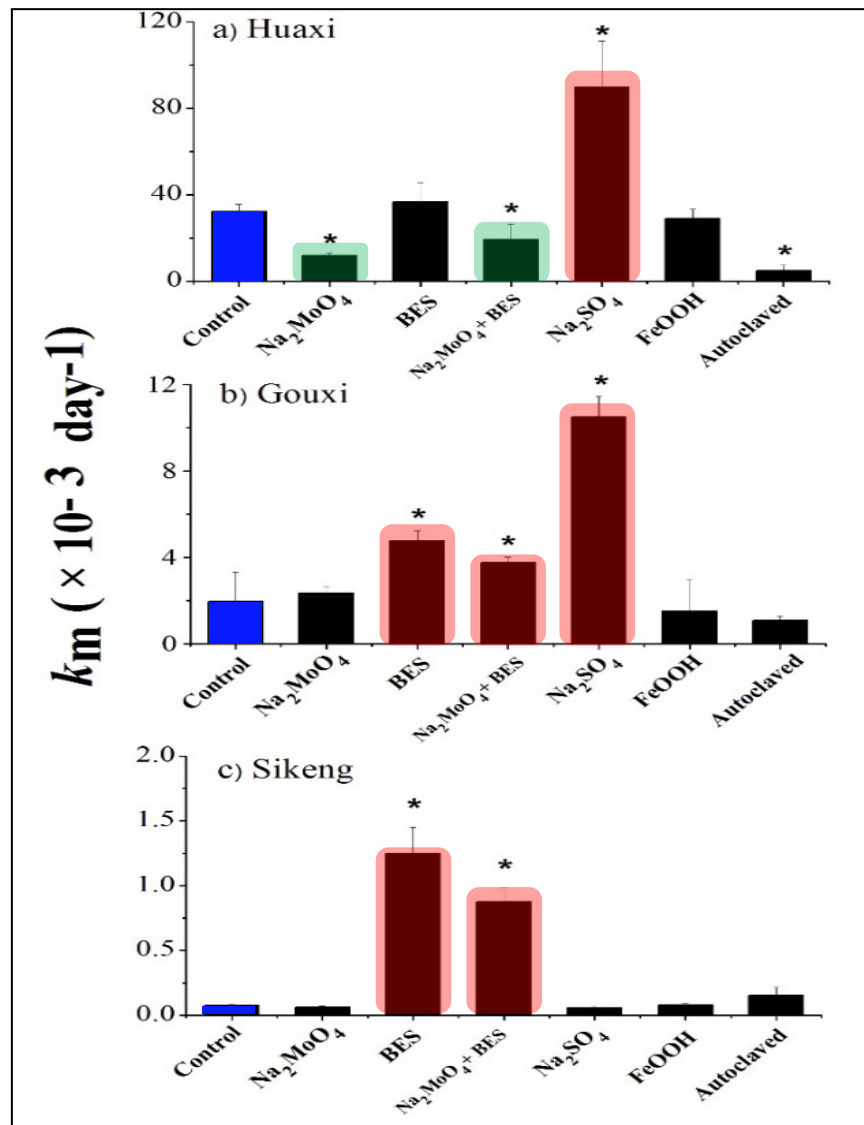
Hg methylation in rice paddy ecosystem

Major knowledge gaps

- (1) Geochemically relevant HgII species that can be methylated in paddy soils?
- (2) The relative importance of microbial groups on MeHg production and degradation in paddy soils**
- (3) The key factors in regulating MeHg formation in paddy soils

Prevent the development of remediation measures necessary to ensure safe consumption of this staple food

(2) Microbial groups on MeHg production and degradation in paddy soils



Using Specific Inhibitors/Stimulants

Hg methylation

- **Control site :**

Sulfate reduction was the potential driver of MeHg formation (SRB)

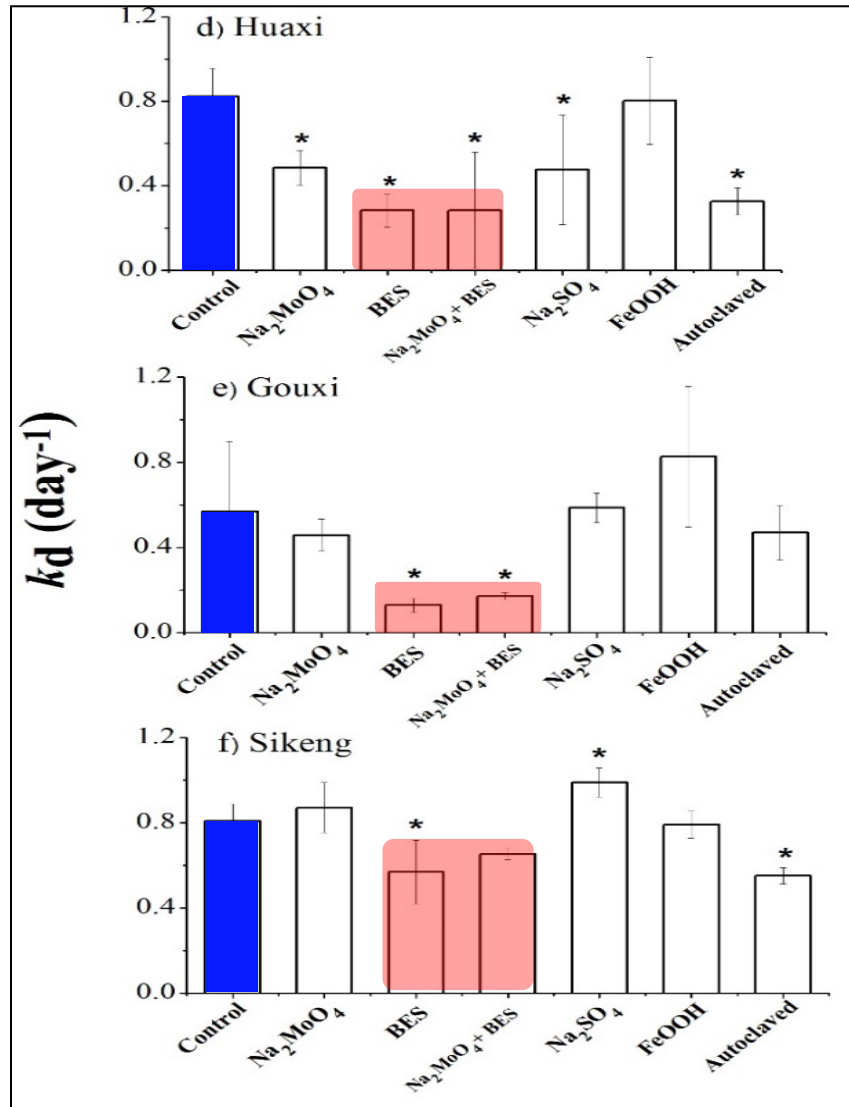
- **Artisanal Hg mining site**

In addition to SRB, some unclassified microbial groups still functional in Hg methylation

- **Abandoned Hg mining site**

Microbials that compete with methanogens participate in the Hg methylation

(2) Microbial groups on MeHg production and degradation in paddy soils



Using Specific Inhibitors/Stimulants

MeHg Demethylation

- Methanogenesis is associated with MeHg degradation

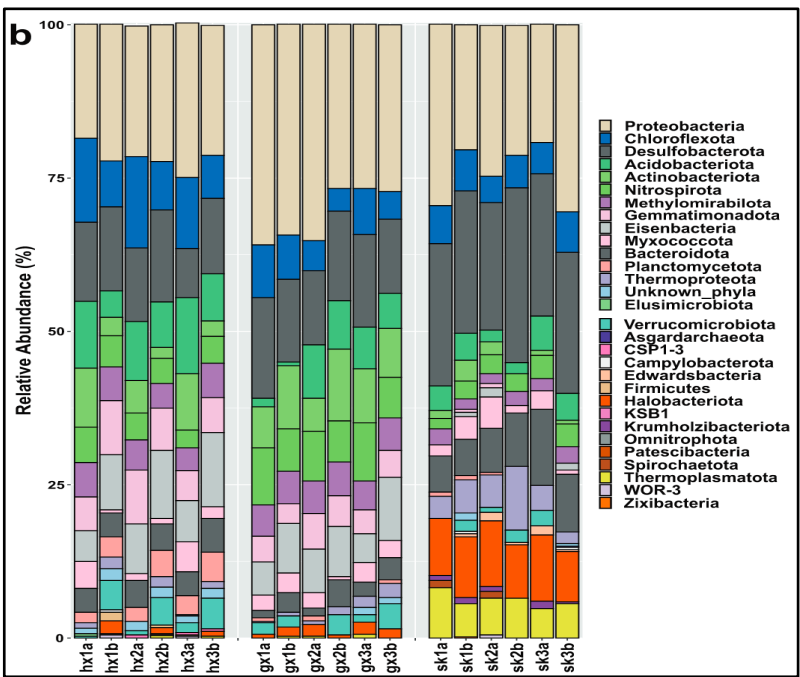
With implication for managing MeHg contamination in paddy soils especially in Hg mining area

Wu et al., 2020, ES&T

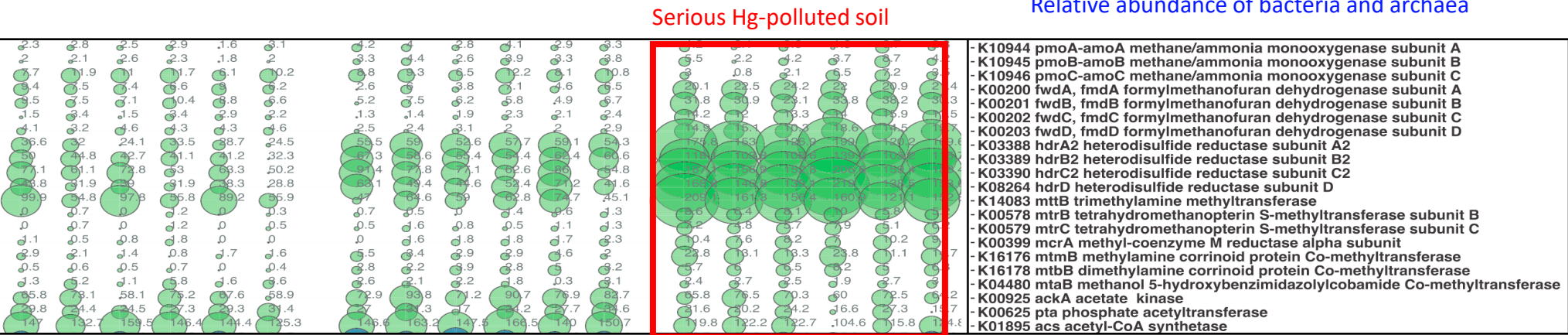
(2) Microbial groups on MeHg production and degradation in paddy soils

- Using metagenomics, we identified Methanogens played very important role in MeHg degradation and competition with other Hg-methylating guilds

Zhang et al., 2023, ISME Commun.



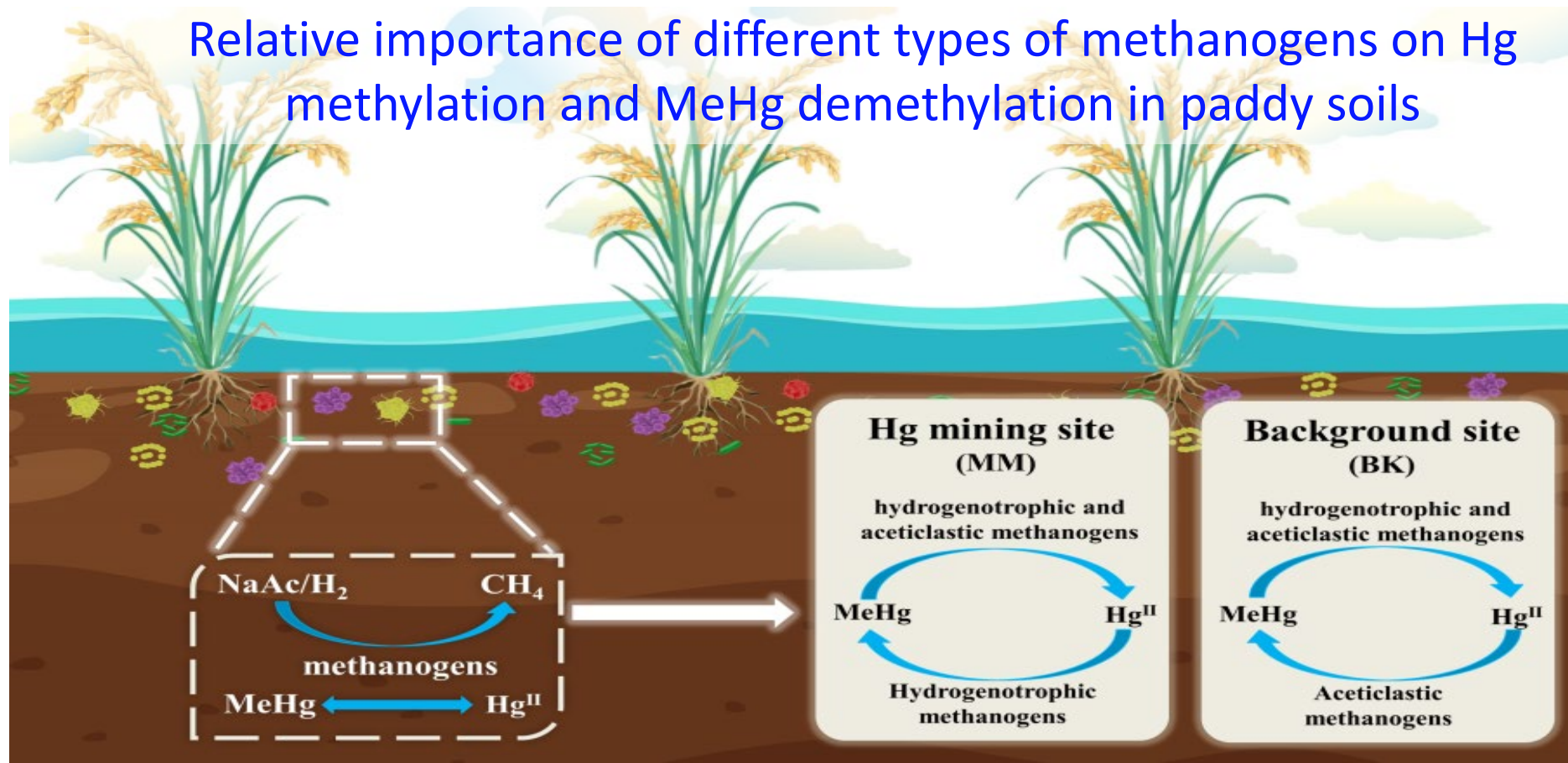
Relative abundance of bacteria and archaea



Normalized abundance of functional genes related to various metabolic pathways

(2) Microbial groups on MeHg production and degradation in paddy soils

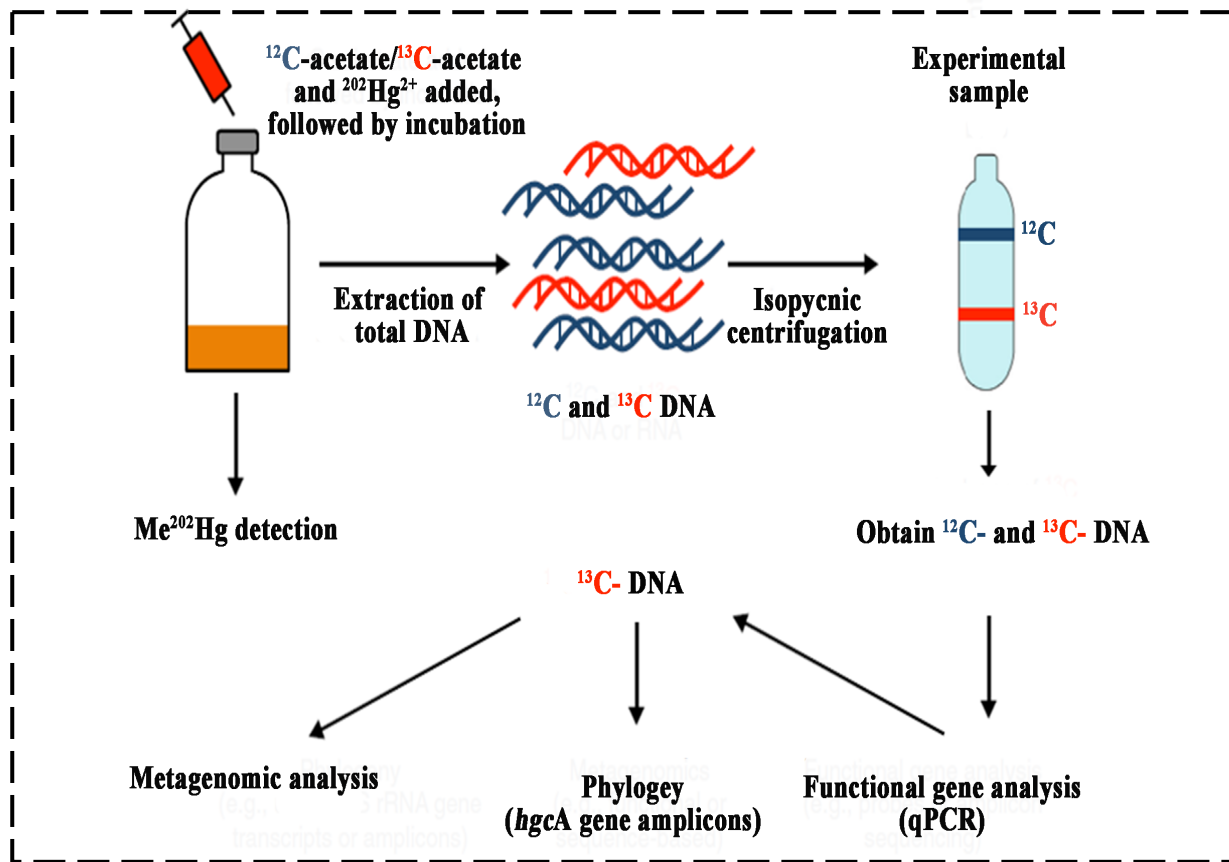
- Aceticlastic methanogens are the potential Hg methylation communities at control site
- Hydrogenotrophic methanogens involved in MeHg production as Hg level increased
- Both aceticlastic and hydrogenotrophic methanogens facilitate MeHg degradation in paddy soils



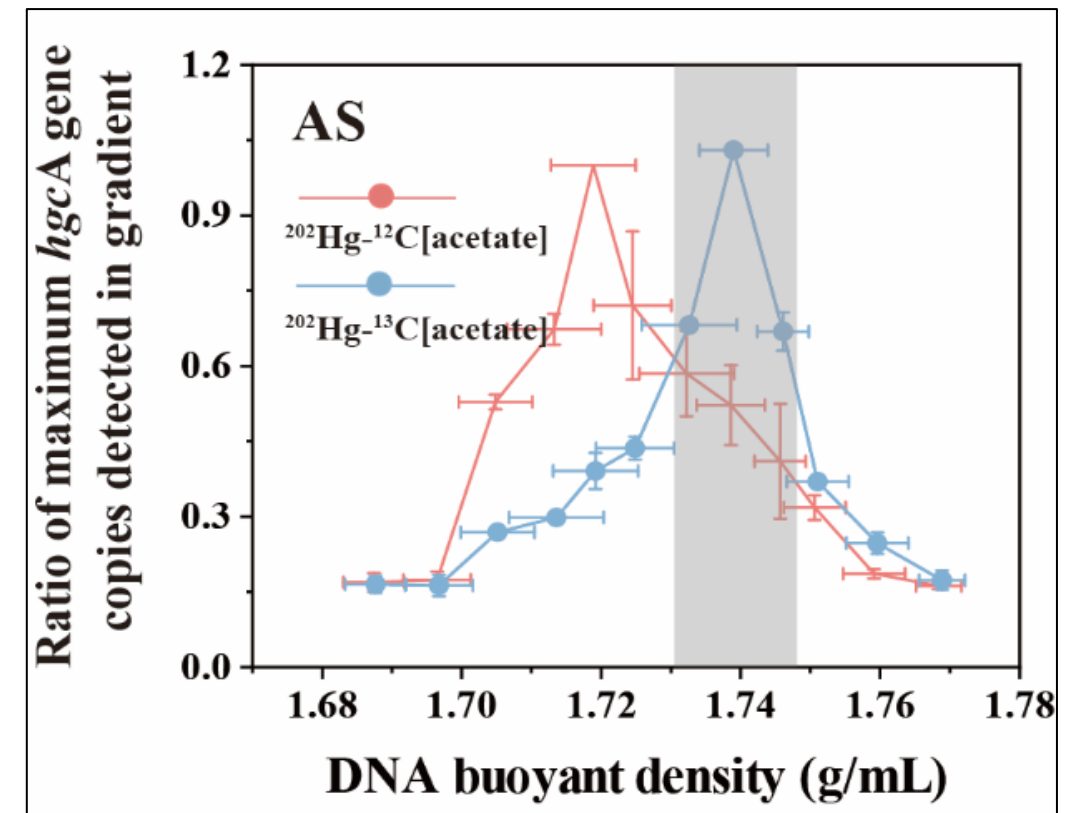
Hao et al., 2023, STOTEN

(2) Microbial groups on MeHg production and degradation in paddy soils

- Identify actively key Hg methylation communities in paddy soils



Methodology: DNA-SIP

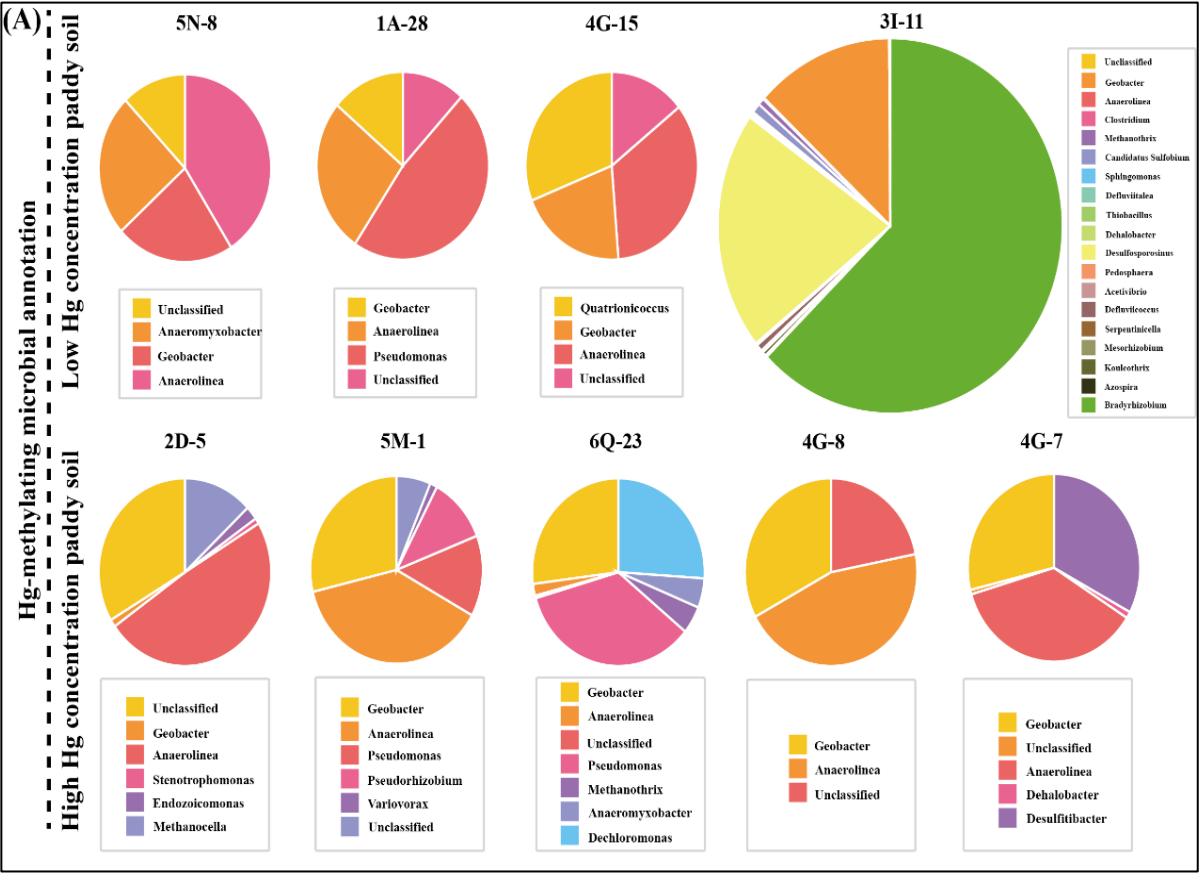
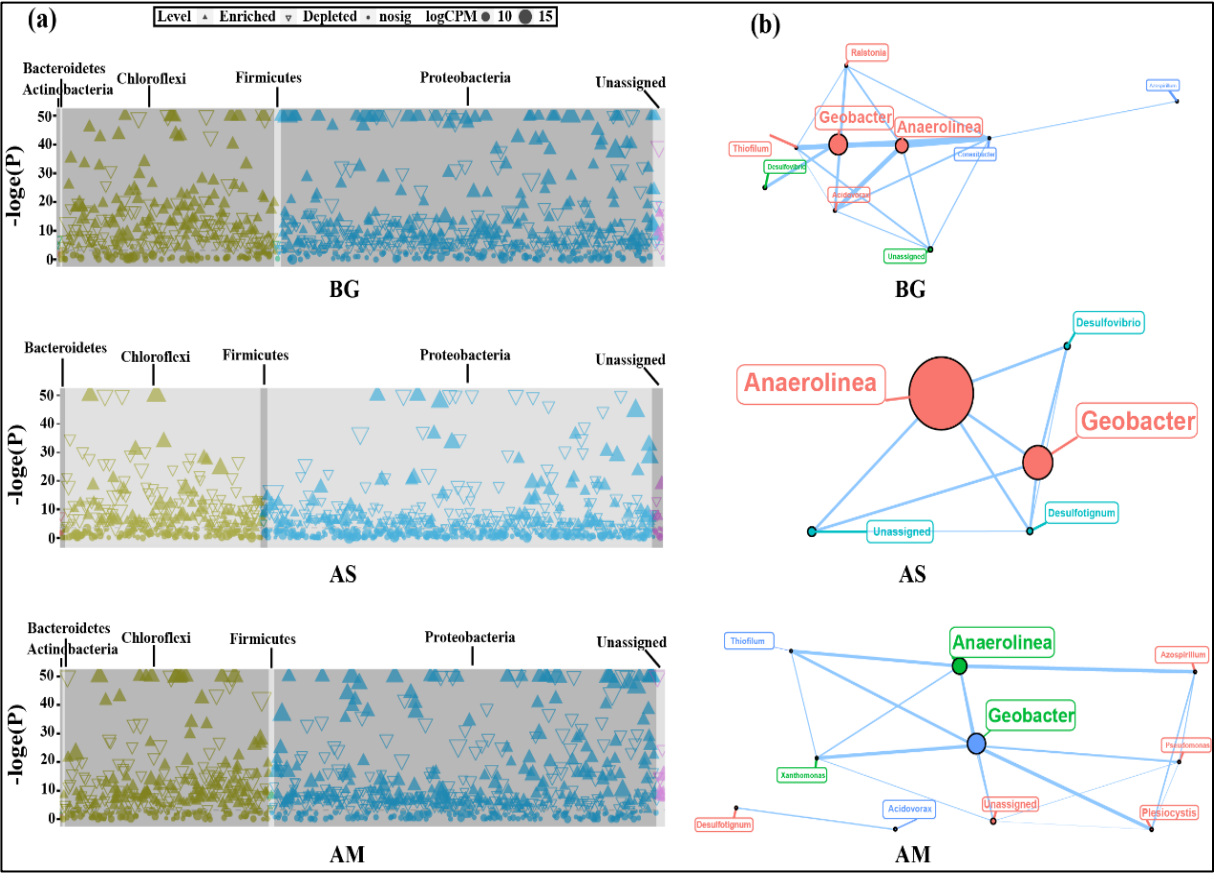


Quantification of *hgcA* gene in SIP incubation cultures

(2) Microbial groups on MeHg production and degradation in paddy soils

● *Geobacter* and *Anaerolinea* are active Hg methylators in paddy soils

Pu et al., unpublished data



Key Hg-methylating microorganisms in paddy soils

Transcriptomic of Hg-methylating microorganisms in paddy soils

Hg methylation in rice paddy ecosystem

Major knowledge gaps

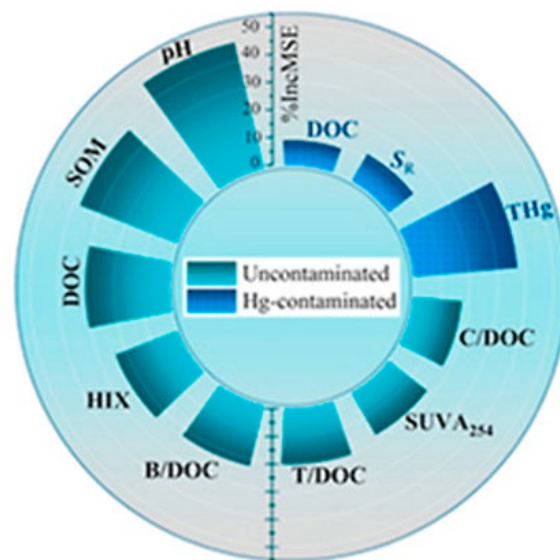
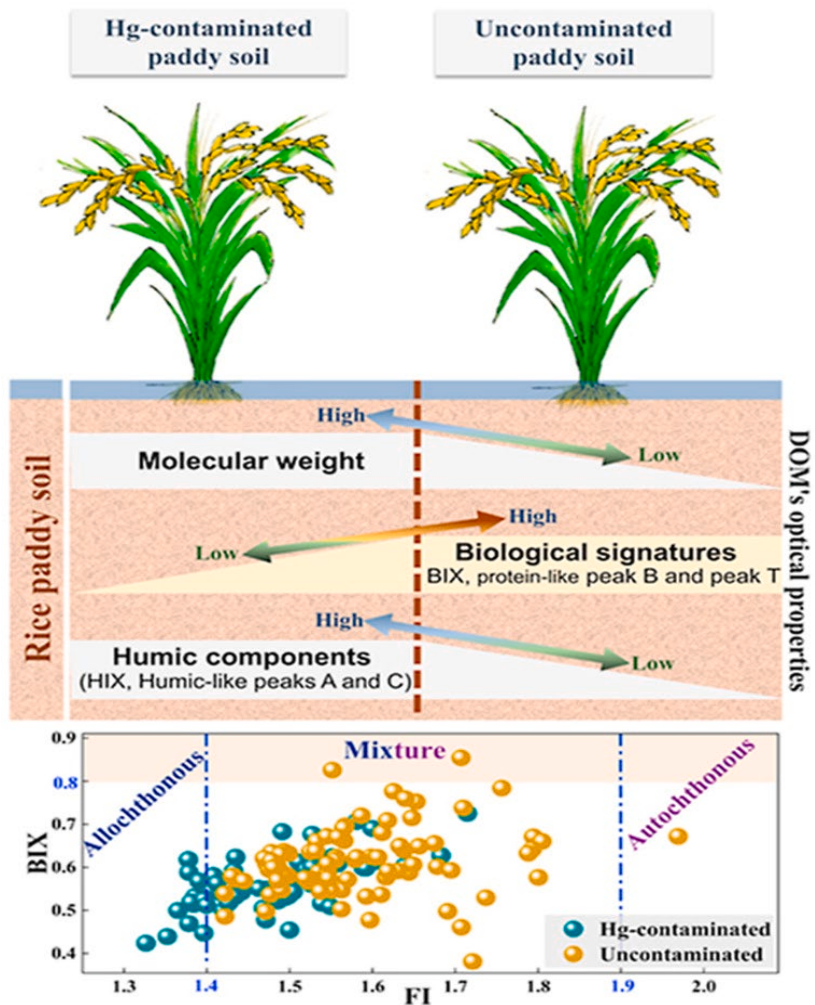
- (1) Geochemically relevant HgII species that can be methylated in paddy soils?
- (2) The relative importance of microbial groups on MeHg production and degradation in paddy soils
- (3) The key factors in regulating MeHg formation in paddy soils**

Prevent the development of remediation measures necessary to ensure safe consumption of this staple food

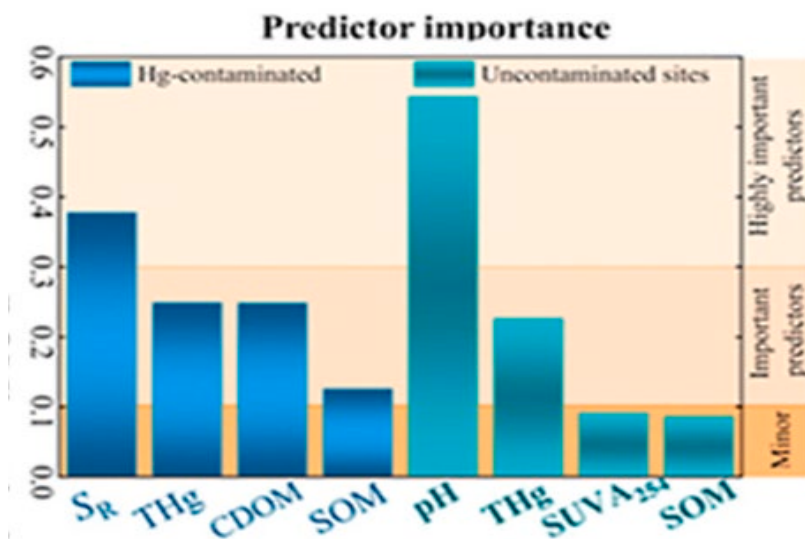
(3) The key factors in regulating MeHg formation in paddy soils

- Low molecular weight DOM promotes Hg methylation
- DOM with higher molecular weight reduced MeHg production

Abdelhafiz et al., 2023, EP



Random Forest Modelling

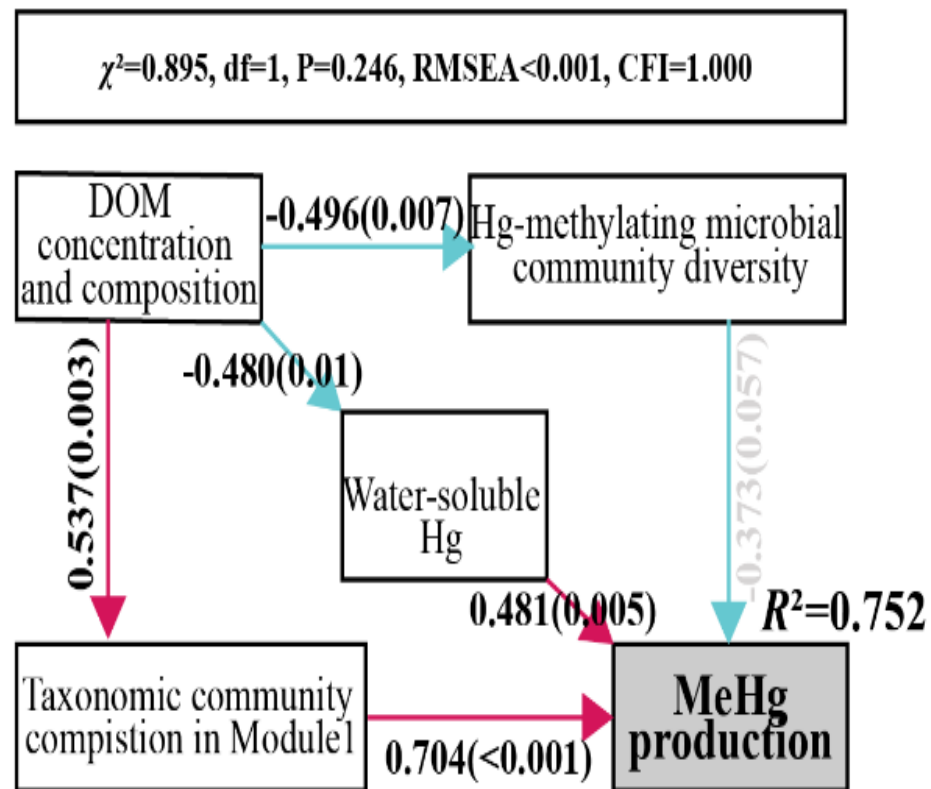


DOM influences Hg methylation in paddy soils across a Hg contamination gradient

(3) The key factors in regulating MeHg formation in paddy soils

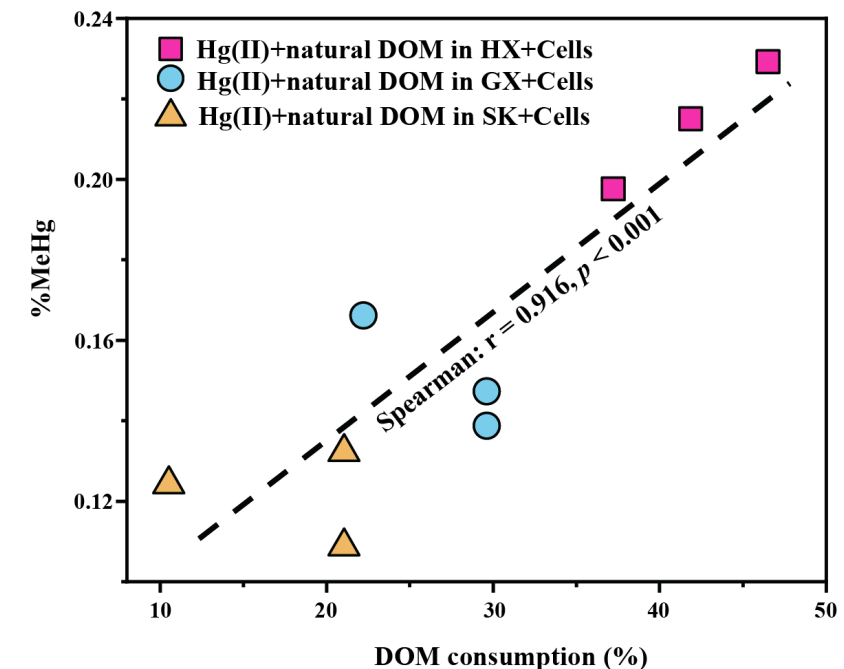
- DOM promote microbial Hg methylation by regulating core Hg methylating microbial composition

Pu et al., unpublished data



Structural equation models reveal the effect of DOM concentration and composition on MeHg production

Direct evidence: DOMs indeed stimulate core Hg methylator (*Geobacter* sp.) for MeHg production



Correlation between MeHg production and DOC

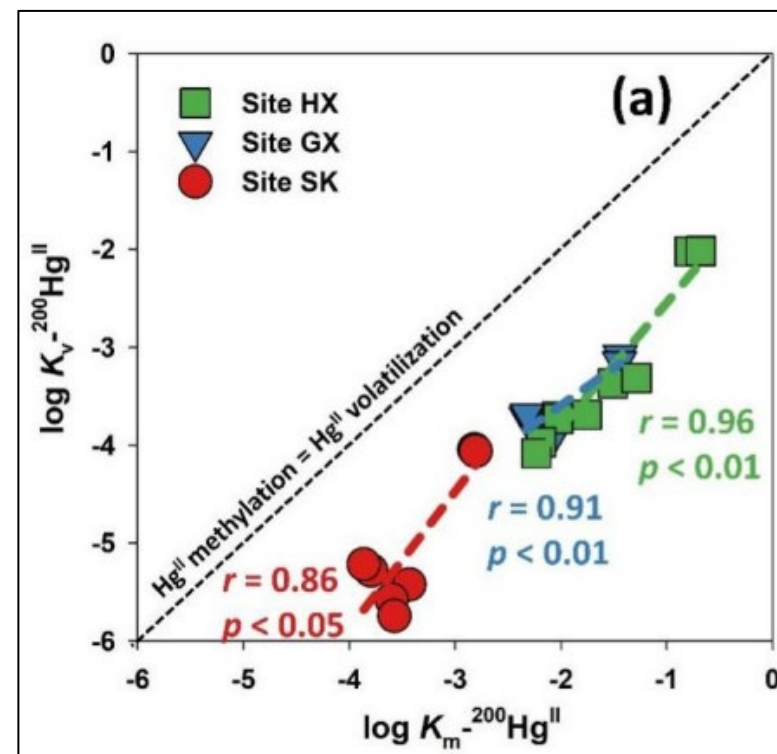
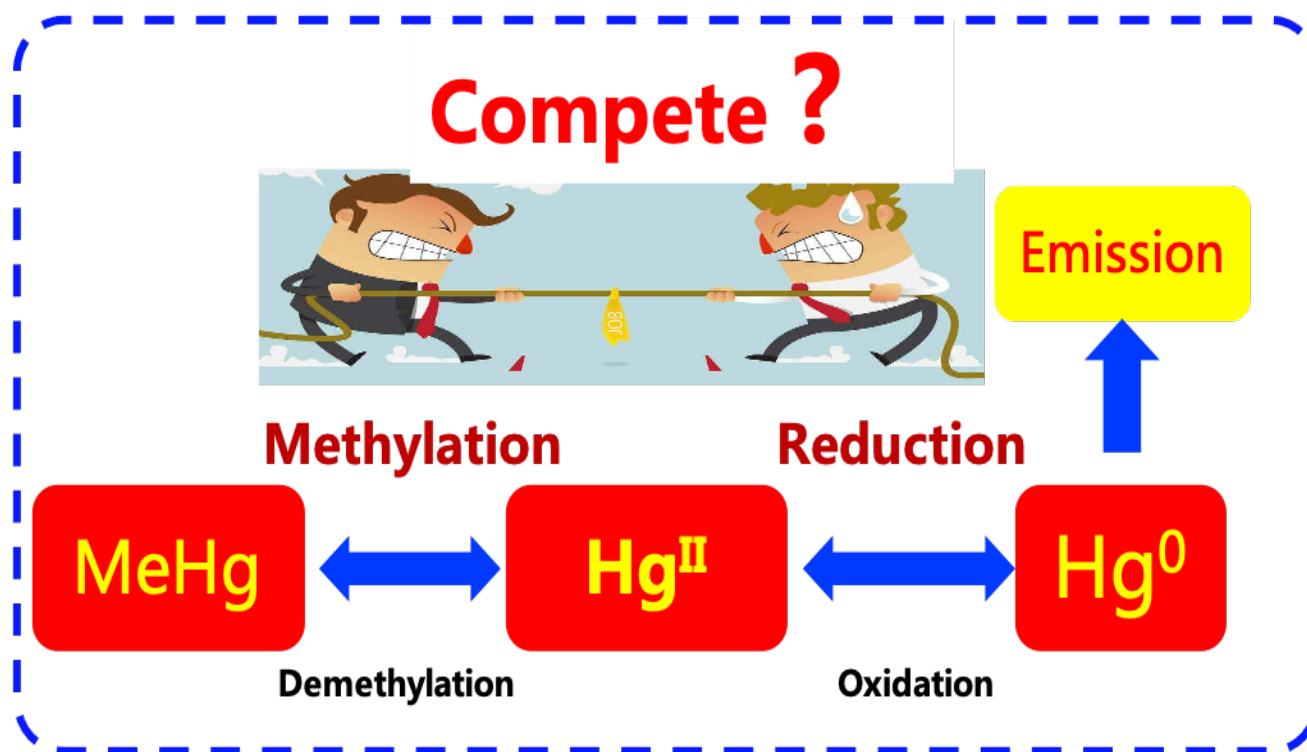
consumption by *Geobacter* sp. WWW.MERCURYCAPETOWN.COM

(3) The key factors in regulating MeHg formation in paddy soils

- Rapid redox recycling of Hg species contributed to Hg speciation resetting, which promoted the transformation between Hg^0 and MeHg by generating bioavailable Hg^{II} for fuel methylation

The role of Hg reduction/oxidation on MeHg production

Liu et al., 2023, EST

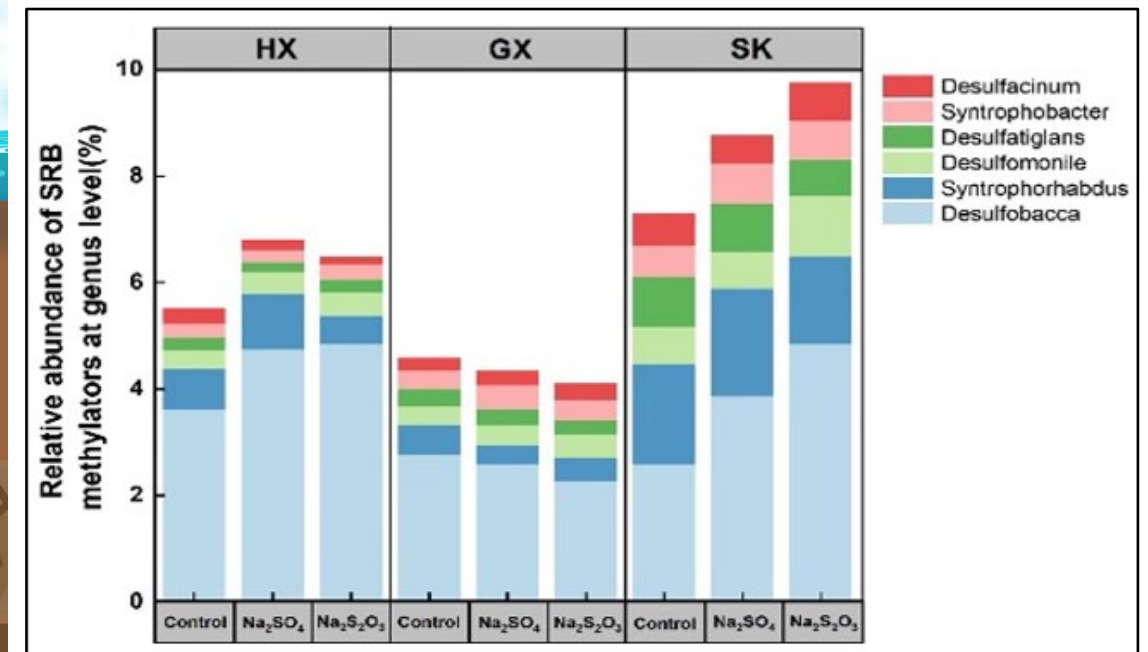
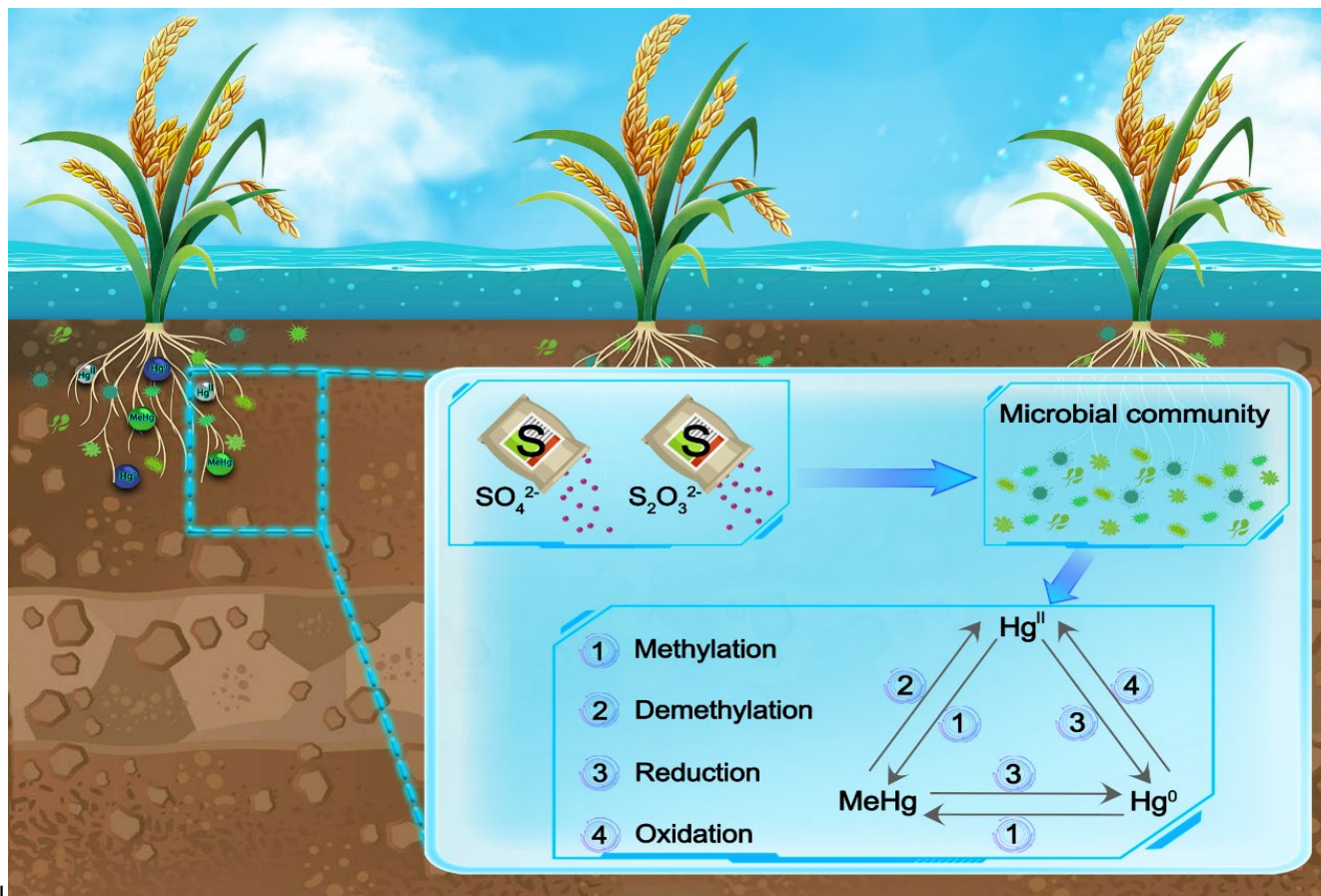


Relationship between $^{200}\text{Hg}^{\text{II}}$ reduction and $^{200}\text{Hg}^{\text{II}}$ methylation

(3) The key factors in regulating MeHg formation in paddy soils

- S-input promoted Hg^{II} methylation by stimulating the Hg methylators
- Evidenced by the more abundance of Hg methylators in sulfur added treatment

Liu et al., 2023, ES&T

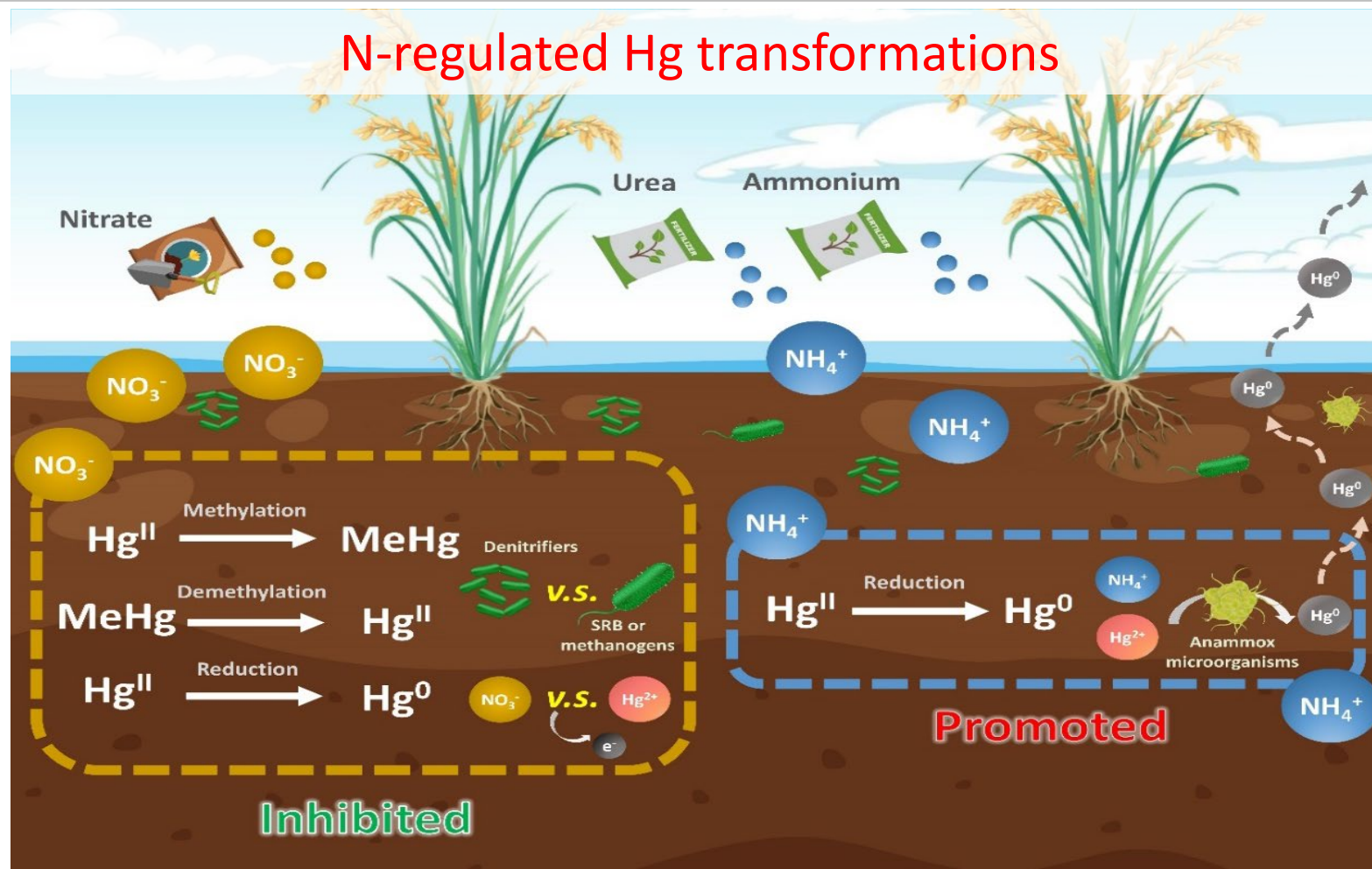


Relative abundance of SRB methylators under S-input

(3) The key factors in regulating MeHg formation in paddy soils

- Nitrate addition inhibits Hg^{II} methylation and MeHg demethylation
- Yielded higher net MeHg accumulation in the incubated paddy soils

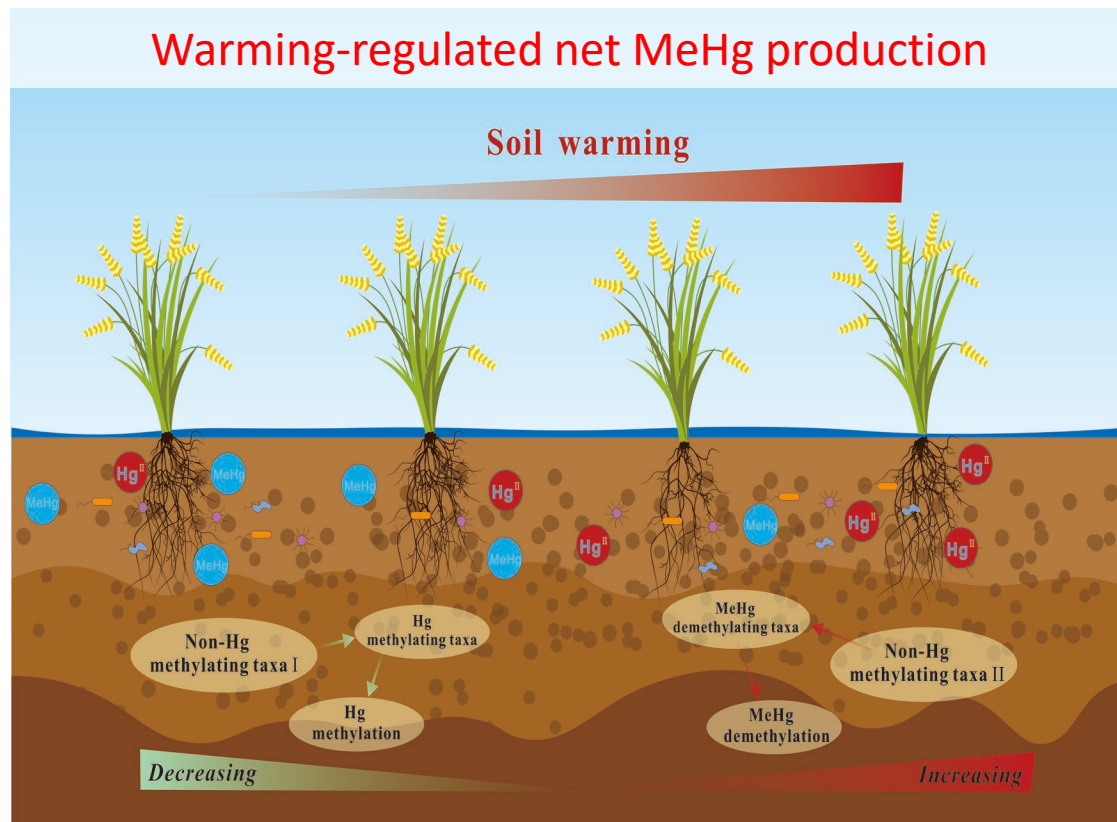
Chen et al., 2023, JHM



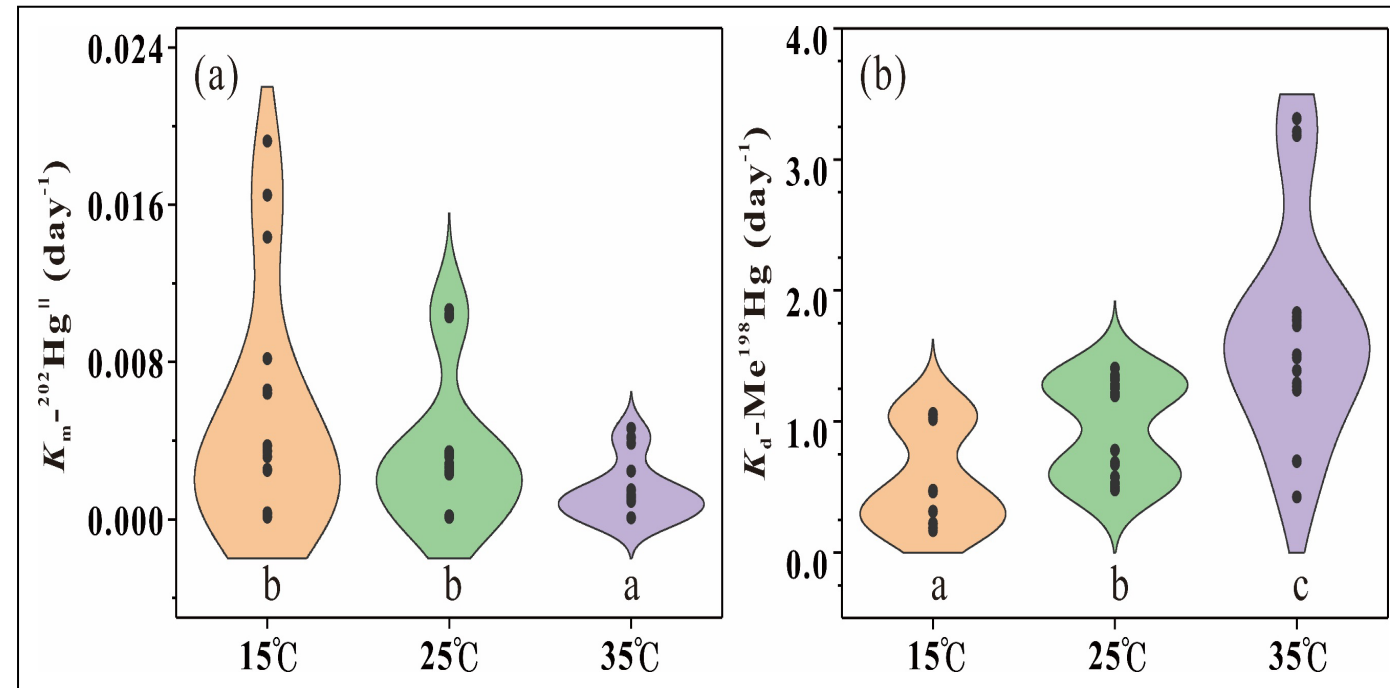
(3) The key factors in regulating MeHg formation in paddy soils

- Warming inhibits Hg^{II} methylation but stimulates MeHg demethylation in paddy soils
- Net MeHg production in paddy soils was largely inhibited under the increasing temperature

Warming-regulated net MeHg production



Zhang et al., 2024, STOTEN



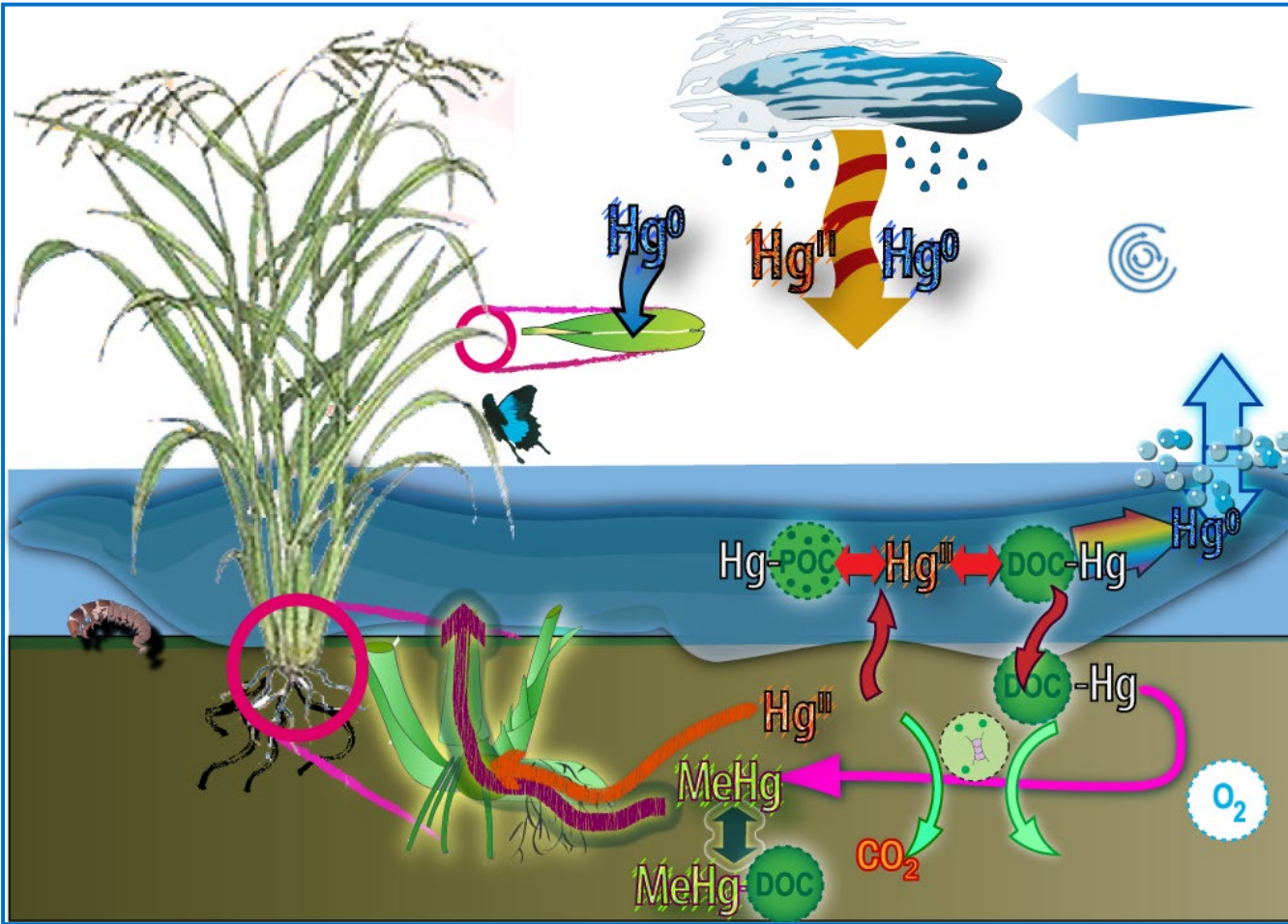
Hg methylation/demethylation rate constants at different temperature

In summary: Hg methylation in rice paddy ecosystem

- (1) Geochemically relevant Hg^{II} species that can be methylated in paddy soils?
 - Bioavailable Hg^{II}, NOM-Hg, and nano-HgS (underappreciated)
- (2) The relative importance of microbial groups on MeHg production and degradation in paddy soils
 - Methylators: site dependent; Demethylators: methanogens
- (3) The key factors in regulating MeHg formation in paddy soils
 - DOM, S/ N redox cycling, and warming

- **Broaden the understanding of biogeochemical cycling of Hg in rice paddy ecosystem**
- **Provide a theoretical basis for remediation of Hg polluted paddy soil**

Challenges and future research



Hg cycling in rice paddy ecosystem

- ◆ Factors in controlling MeHg production in paddy soils is far to be fully understood
- ◆ The absorption pathways of MeHg through rice root?
- ◆ The gene associated with MeHg up-taken in rice plant is still unknown
- ◆ Remediation measures are urgently needed to reduce Hg exposure to local residents

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谢谢！

Thanks for your attention!

