

Fig. S1 Schematic diagram of *phn* gene clusters in 128 phycospheric heterotrophic bacterial binnings . Refer to Supplementary Dataset 3 for detailed strain information.



Fig. S1 Continue

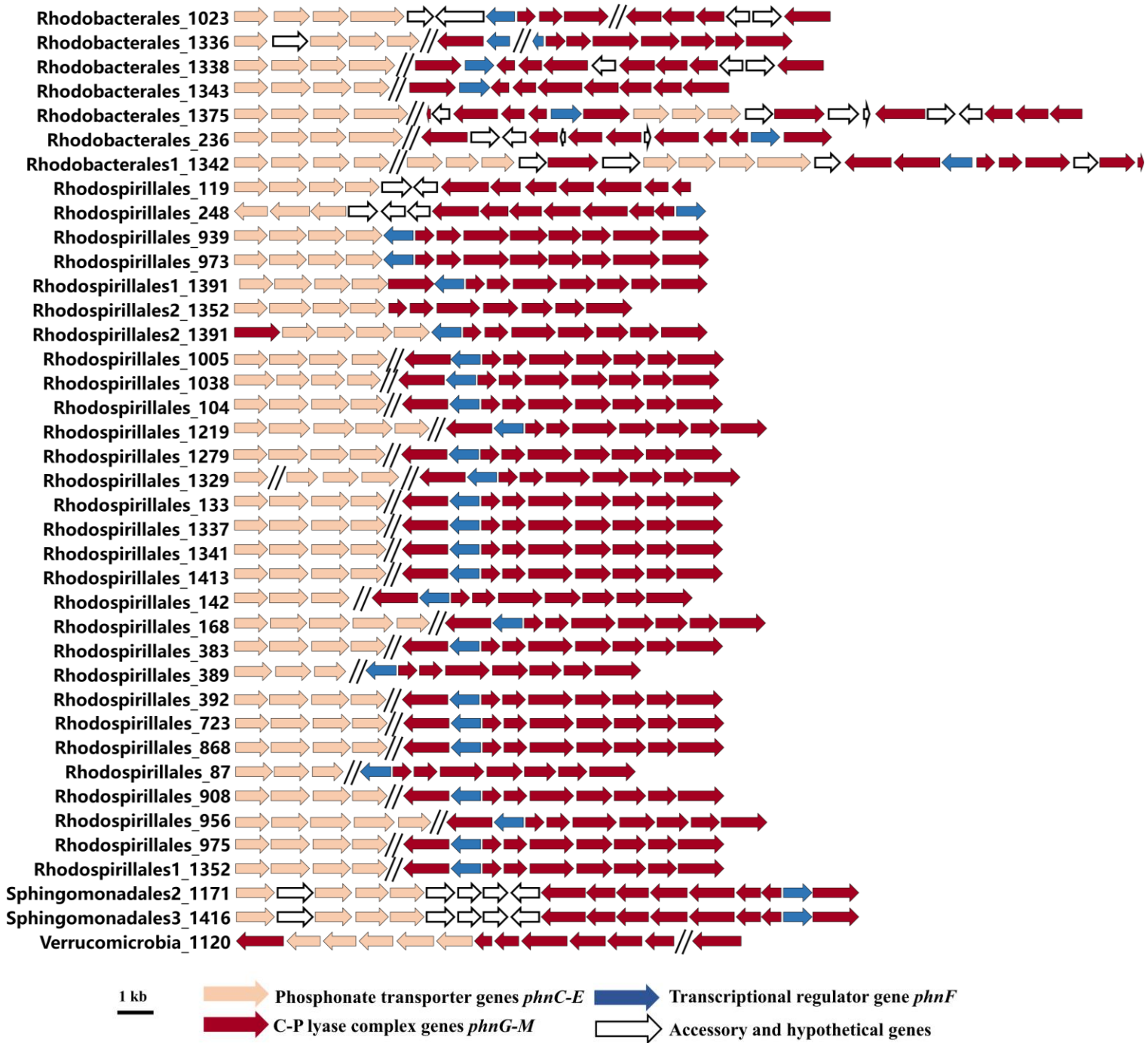
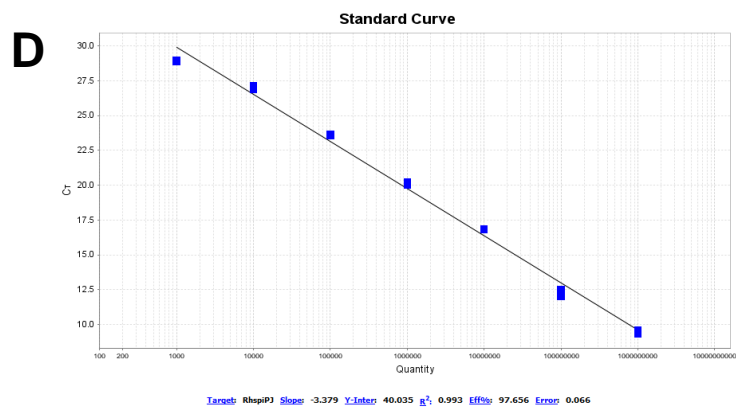
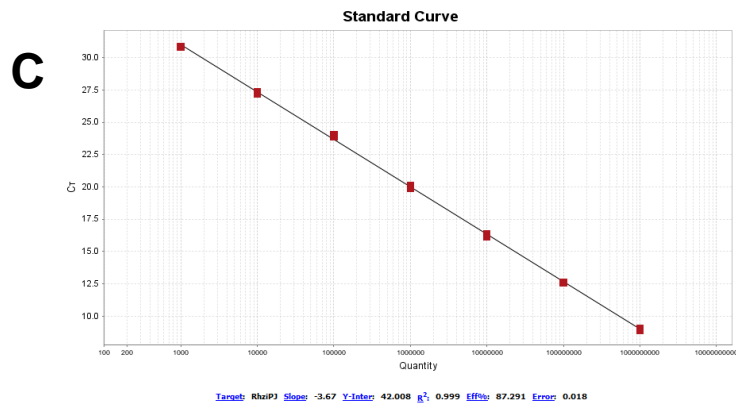
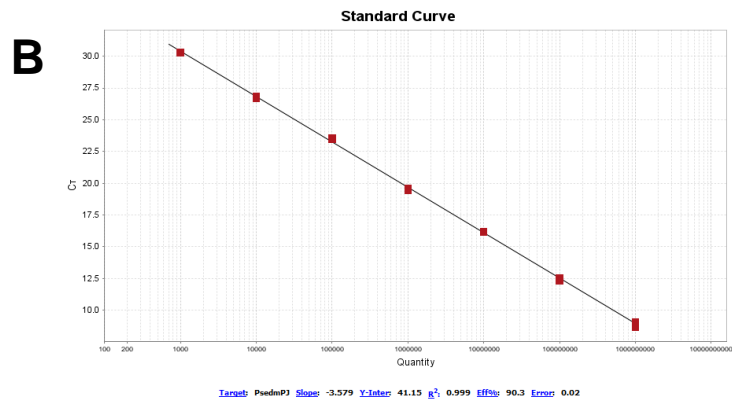
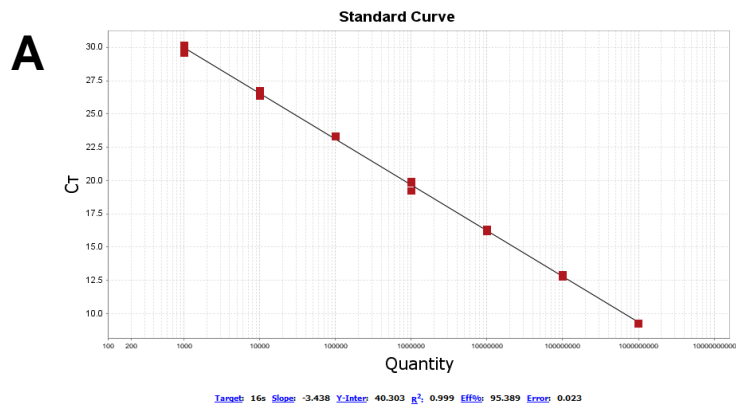
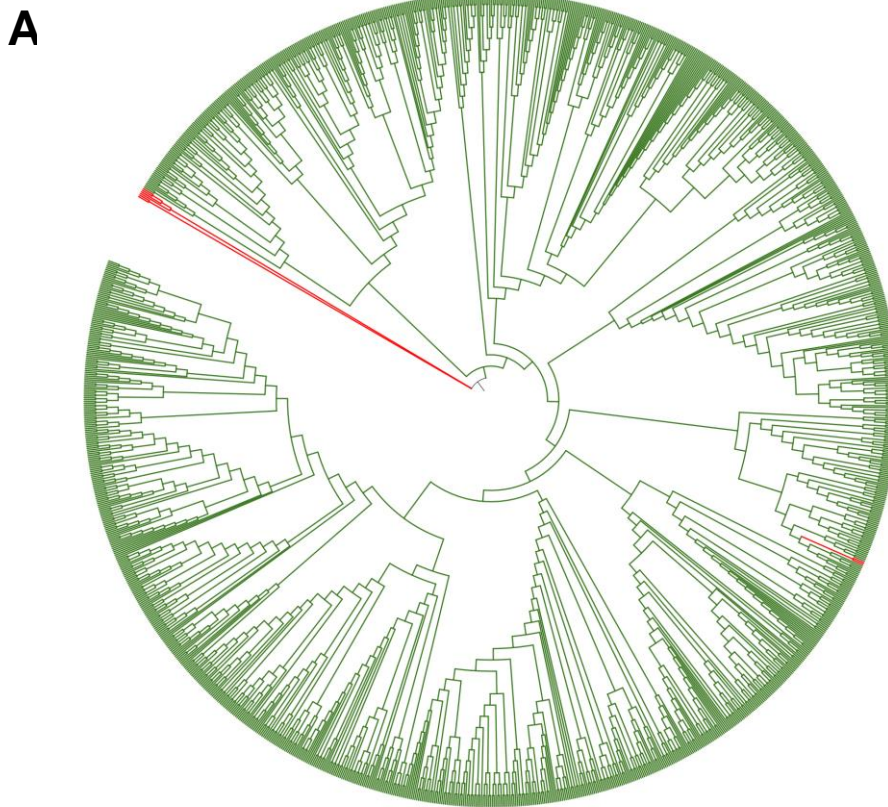


Fig. S1 Continue



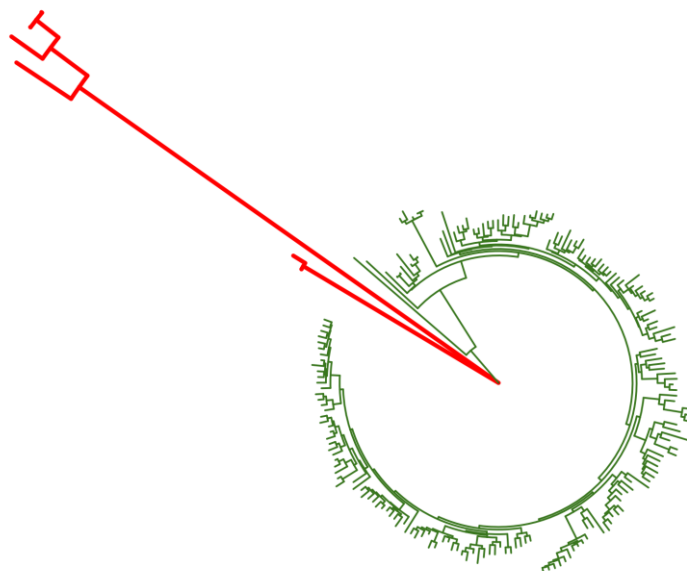
**Fig. S2 Standard curves of the four qPCR primer sets used in this study.**

**A**, 16S rRNA gene; **B**, PsudmoPJ; **C**, RhizobPJ; **D**, RhispiPJ.

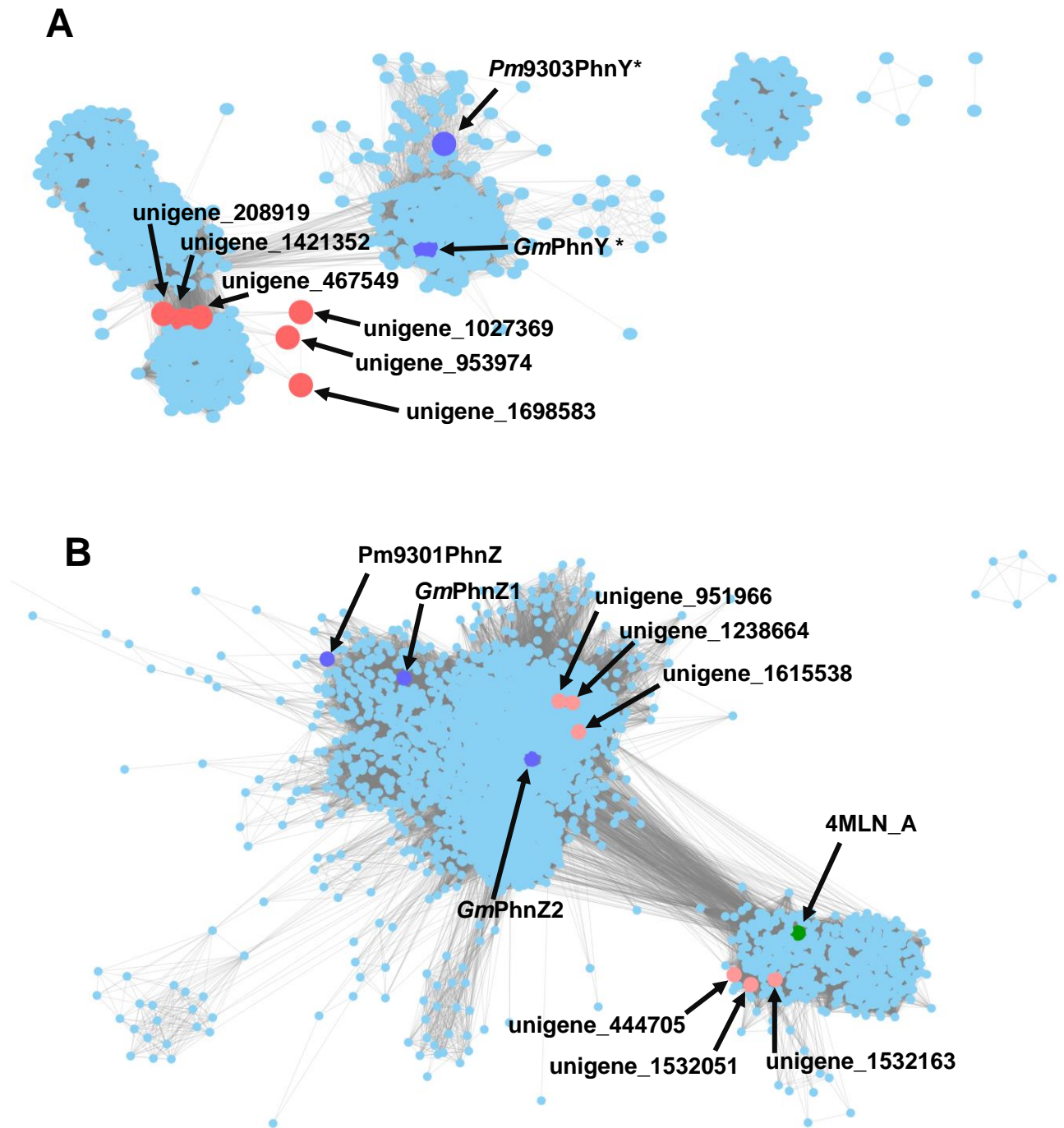


**B**

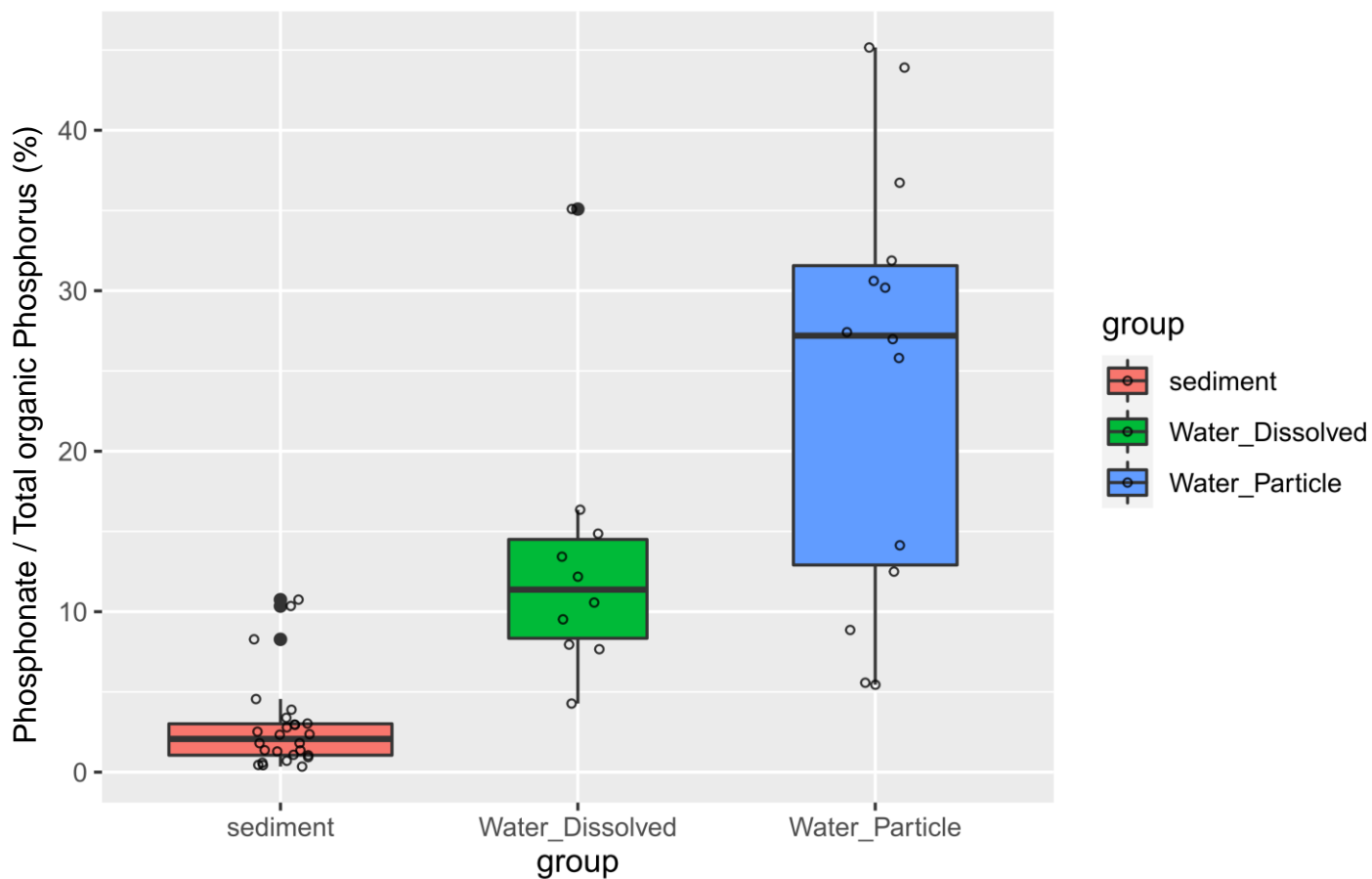
Tree scale: 10



**Fig. S3 PafA (A) and AepX (B) protein function validation results in phylogenetic trees.** Data used in tree construction are from Murphy et al. (2021) and Lidbury et al. (2022), respectively. Trees were constructed using maximum likelihood methods under the LG + Gamma + Invariant model. **A)** Two out of eight potential PafA protein hits (red branches) are located in the main PafA tree (green branches) and their function are confirmed. **B)** All potential AepX protein hits (red branches) are located out of the main AepX tree (green branches), indicates the mis-annotation of this protein.



**Fig. S4 Sequence similarity network for PhnY\* and PhnZ unigenes specificity validation.** The method used in network construction was described in Gama et al., 2019. **A)** The network is based on 1000 unique sequences retrieved from a BLAST analysis of HF130PhnY\*. Individual clusters correspond to sequences with >40% identity (alignment score = 60). **B)** The network is based on 5000 unique sequences retrieved from a BLAST analysis of HF130PhnZ. Individual clusters correspond to sequences with >45% identity (alignment score = 40). Nodes in purple: MPn-specific genes; node in green: 2-AEP-specific genes; nodes in red: unigenes generated with meta-transcriptomic data in this work.



**Fig. S5: Percentage of total organic phosphorous that occurs as phosphonates in different fractions of various sources of freshwater.** Data was obtained through a literature search for quantitative measurements of freshwater phosphorous (including phosphonate) contents. Refer to Supplementary Dataset 6 for detail.