**Supporting Text**

**Taxa with no obvious ecological functions**

Besides the taxonomic groups described in the main text, we could also identify further positive linear relationships of bacterial sequences with decreasing pH, which could not directly be linked to ecological functions. This includes the phylum of Actinobacteria (9% of all bacterial sequences) with the major order of Actinomycetales (R2 = 0.25, *P =* 0.096, Table S4), which represented 34% of all Actinobacteria associated sequences and is known to contain ecologically, morphologically, and chemotaxonomically very divergent range of species (Gao and Gupta, 2012). Among the Deltaproteobacteria we found another positive linear relationship between pH decrease and sequence occurrence for the order Bdellovibrionaceae (R2 = 0.41, *P =* 0.025, representing 2% of all Deltaproteobacteria sequences, Table S4). Being widespread in marine environments, they typically predate on other bacteria using an unusual mechanism to stick to cell surfaces of their prey (Madigan et al., 1997; Starr and Baigent, 1966).

**Quadratic relationships of microbial MPTS sequence occurrence with pH**

Among the archaeal community we found negative quadratic relationships with pH for sequences of three OTUs, which were closely related to the Thaumarchaeota *Nitrosopumilus* *koreensis* (with nt identities of 91, 97 and 91%, respectively), which together represented 9.4% of all archaeal sequences in the dataset. *N. koreensis* have previously been isolated from 78 m-deep marine sediment off Svalbard (Park et al., 2012). In our dataset peaks of sequence occurrence for the three OTUs were found at sites with pH 8.3 and 8.1 (R2 = 0.80, *P =* 0.026; Table S7), 8.2 and 7.8 (R2 = 0.48, *P =* 0.091; Table S7) and pH 7.8 (R2 = 0.64, *P =* 0.032; Table S7), respectively. Despite the fact that we found a highly significant linear increase with decreasing pH for sequences of the Thaumarchaeota *Nitrosopumilus* *maritimus*, only a low number of *N. koreensis* related sequences were detected at pH < 7.8. Another negative quadratic relationship with pH was identified for an OTU with sequences closely related to Thaumarchaeota *Nitrososphaera gargensis* (92% nt identity) with a peak at sites ranging between pH 8.0-7.8 (R2 = 0.90, *P =* 0.001; Table S7). *N. gargensis* related sequences represented less than 0.1% of all archaeal sequences in our dataset. The species has previously been enriched from a runoff basin of the Garga spring (Spang et al., 2012), at a water pH in the similar range as the one in our study (pH 7.9). Despite the fact that *N. gargensis* seems able to use various alternative ammonia sources for ammonia oxidation and harbors genes with functions not found in other known Thaumarchaeota species, little is known about its possible role in marine environments (Spang et al., 2012).

Sequences related to the Euryarchaeota genus *Methanoplanus* (representing 0.3% of all archaeal sequences), showed a negative quadratic relationship with pH, peaking at pH 8.0 (R2 = 0.89, *P =* 0.002; Table S7). While we have found no increased numbers of *Methanoplanus* sequences below pH 8.0, methanogenic species of this archaeal family have been described to grow optimally at pH 7 in a temperature range between 17-41°C under the strict requirement of an acetate source (Wildgruber et al., 1982).

All other archaeal sequences with significant quadratic relationships to pH were related to the Crenarchaeota group Thermoprotei as well as the Euryarchaeota groups Methanopyrus and Thermoplasmatales, and fell into the category of thermophiles, but could not be further identified. Sequences of two OTUs related to the Thermoprotei genus *Aeropyrum* represented 0.4% of all archaeal sequences and showed a negative quadratic relationship with pH at sites with a seawater pH between 7.8-6.9 (R2 = 0.52, *P =* 0.091 and R2 = 0.46, *P =* 0.095, respectively; Table S7). *Aeropyrum* species have been isolated before from a coastal solfataric vent at Kodakara-Jima Island, Japan and were described as strictly aerobic, heterotrophic, thermophiles which grow between a temperature range from 70-100°C with a pH optimum at 7.0 (Sako et al., 1996). Sequences related to *Methanopyrus*, which are another group of extreme thermophilic methanogens, being able to survive temperatures up to 110°C, have been previously described from thermally heated deep sea sediment at Guaymas Basin, Gulf of California (Kurr et al., 1991), made up 1.2% of all archaeal sequences in our dataset and showed a negative quadratic relationship with pH and a peak at sites with a pH range from 8.0-7.8 (R2 = 0.81, *P =* 0.008; Table S7). While sequences of one group of the thermophilic and extremely acidophilic Thermoplasmatales were found to significantly increase in relative abundance with pH, we found another subgroup, which represented 0.4% of all archaeal sequences in our dataset with a negative quadratic relationship with pH and the highest sequence occurrence at a site with pH 7.8 (R2 = 0.81, *P =* 0.008; Table S7). Altogether most significant quadratic relationships of extremophile archaeal members occurred with highest sequenced occurrences at sites between pH 8.0-7.8. As has been speculated in the main text of this study, we rather expect them to be transported through the vents from deeper sub-surface layers with suspected higher temperatures, since there are no further indications for such extreme conditions at the surface of the studied sites.

The same is probably true for the thermophilic Chloroflexi members of the bacterial community, which showed a likewise negative quadratic relationship with highest sequence occurrences at sites in the pH range 8.0 to 7.8. Nevertheless, sequences of the Chloroflexi class Thermomicrobia, which are a group of thermophilic green non-sulphur bacteria and represented 0.3% of all bacterial sequences in our dataset showed the highest sequence occurrence at sites with pH levels between 7.8 and 7.2 (R2 = 0.33, *P =* 0.096; Table S6). While sequences related to the Chloroflexi genus Bellilinea have been described as strictly anaerobic organisms with no photosynthetic growth (Yamada et al., 2007). They represented 0.5% of all bacterial sequences in our data (R2 = 0.39, *P =* 0.044; Table S6).

Sequences assigned to one deltaproteobacterial OTU, which represented 1% of all bacterial sequences, showed a positive quadratic relationship with pH, with peaks of occurrence at sites with pH 8.2 and 6.9 (R2 = 0.31, *P =* 0.076; Table S6). The representative sequence of this OTU was closely related to *Acidobacterium capsulatum* strain ATCC 51196 (90% nt identity), which has been described from soil environments as a heterotrophic, facultative anaerobic and acidophilic organism with a preferred pH range of 6.0-3.0 (Ward et al., 2009). *A. capsulatum* members are also frequently found in iron-rich environments such as abandoned mines (Ward et al., 2009).

Besides extremophile members of the bacterial community, we found more quadratic relationships for members of the Deltaproteobacteria, Gammaproteobacteria, Bacteroidetes and Planctomycetes. Sequences of the sulphate reducing Deltaproteobacteria members Desulfovibrionaceae with its subgroup Desulfonauticus as well as Syntrophaceae all showed negative quadratic relationships of sequence occurrence with pH, with peaks at sites with a seawater pH range of 8.2-7.8 (R2 = 0.36, *P =* 0.080, R2 = 0.41, *P =* 0.086 and R2 = 0.37, *P =* 0.059, respectively; Table S6).

Another Deltaproteobacteria of the Desulfovibrionaceae was represented by sequences related to Bilophila which lacks the ability for sulphate reduction and is typically found in mammal faeces (Baron, 1997). It represented < 0.1% of all bacterial sequences, showed a negative quadratic relationship with pH and the highest sequence occurrence at pH 7.8 (R2 = 0.52, *P =* 0.027; Table S6). Sequences of the Deltaproteobacteria order Sorangiineae, which predominantly live in soil and feed on insoluble organic substances and are known for their exceptionally large genomes (Schneiker et al., 2007), represented 0.2% of all bacterial sequences in our dataset, and showed a negative quadratic distribution with pH, peaking in occurrence at pH 7.8 (R2 = 0.44, *P =* 0.026; Table S6).

Among the Gammaproteobacteria we found positive quadratic relationships with pH for the orders Oceanospirillales (0.5% of all bacterial sequences) and Chromatiales (1.0% of all bacterial sequences), with peaks of sequence occurrence at sites with pH ranges of 8.3-8.1 and 7.3-6.9 (R2 = 0.40, *P =* 0.079 and R2 = 0.36, *P =* 0.076, respectively; Table S6). While Oceanospirillales are strictly aerobic and mostly marine species (Garrity et al., 2005), Chromatiaceae are so called phototrophic purple sulphur bacteria and typically grow under anoxic conditions (Ehrenreich and Widdel, 1994).

For sequences of the Planctomycetes family Phycisphaeraceae, which represented 0.6% of all bacterial sequences, we found a negative quadratic relationship with pH, peaking at sites with levels between pH 8.3-8.1 (R2 = 0.48, *P =* 0.062; Table S6). Phycisphaeraceae are motile and facultative anaerobic organisms, which optimally grow at a temperature range between 25-30°C. As all members of the Planctomycetes they are Gram-negative and ubiquitously found in diverse aquatic and terrestrial environments, yet mostly uncultured (Fukunaga et al., 2009).

While we generally found a highly significant linear increase of all Bacteroidetes sequences with pH, including those of the dominant class Sphingobacteriales, we found a negative quadratic relationship with pH for sequences related to the unclassified Sphingobacteriales clone BD2-2 (0.2% of all bacterial and 2.2% of all Bacteroidetes sequences) with the highest sequence occurrence at sites with pH 7.7 (R2 = 0.44, *P =* 0.026; Table S6). Also sequences of the Bacteroidetes family Reichenbachiella, which represented less than 0.1% of all bacterial and 0.9% of all Bacteroidetes sequences, showed a negative quadratic distribution with pH with the highest occurrence at sites with a seawater pH of 7.9 and 7.2 (R2 = 0.67, *P =* 0.004; Table S6). Species of the motile, Gram-negative and strictly aerobic Reichenbachiella family have been described to optimally grow at 28-30°C at a pH range between 7.0 to 7.5 (Cha et al., 2011).

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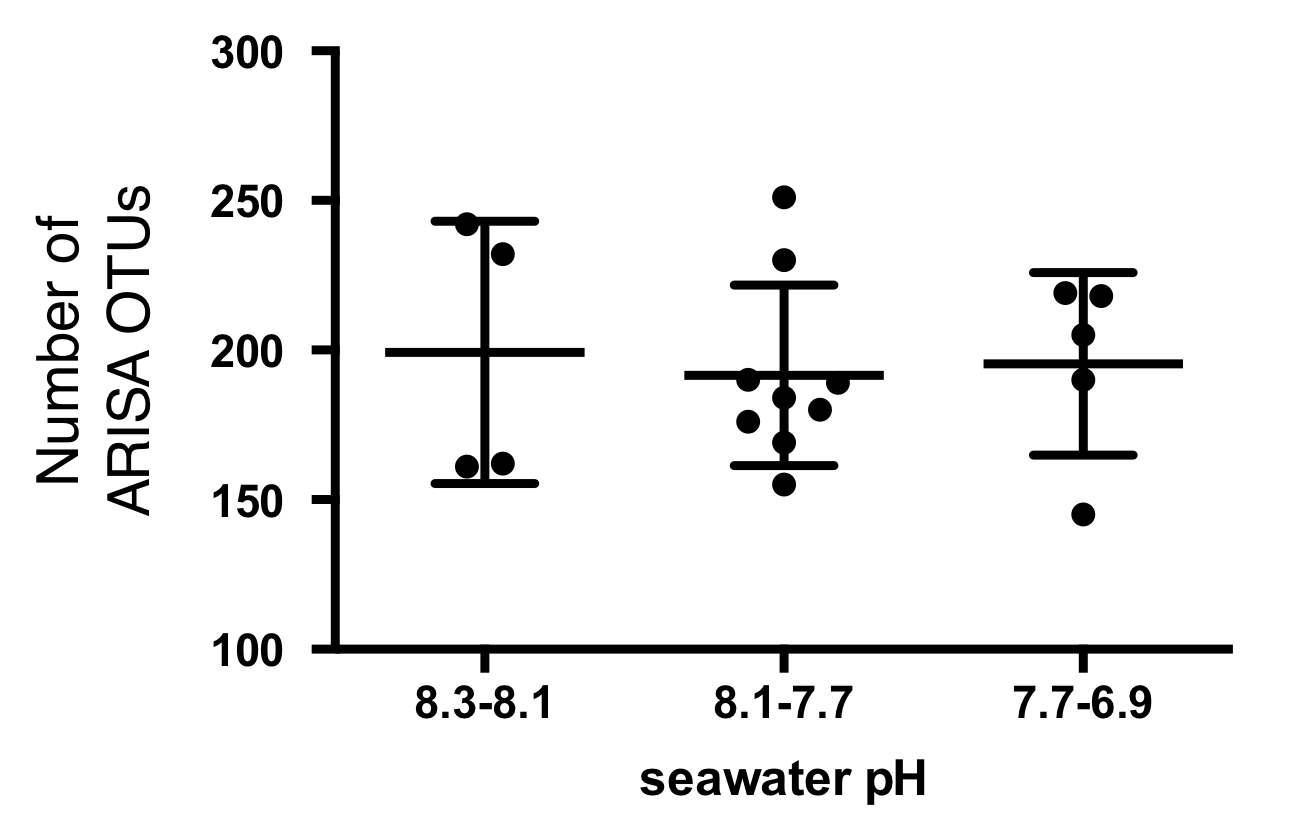
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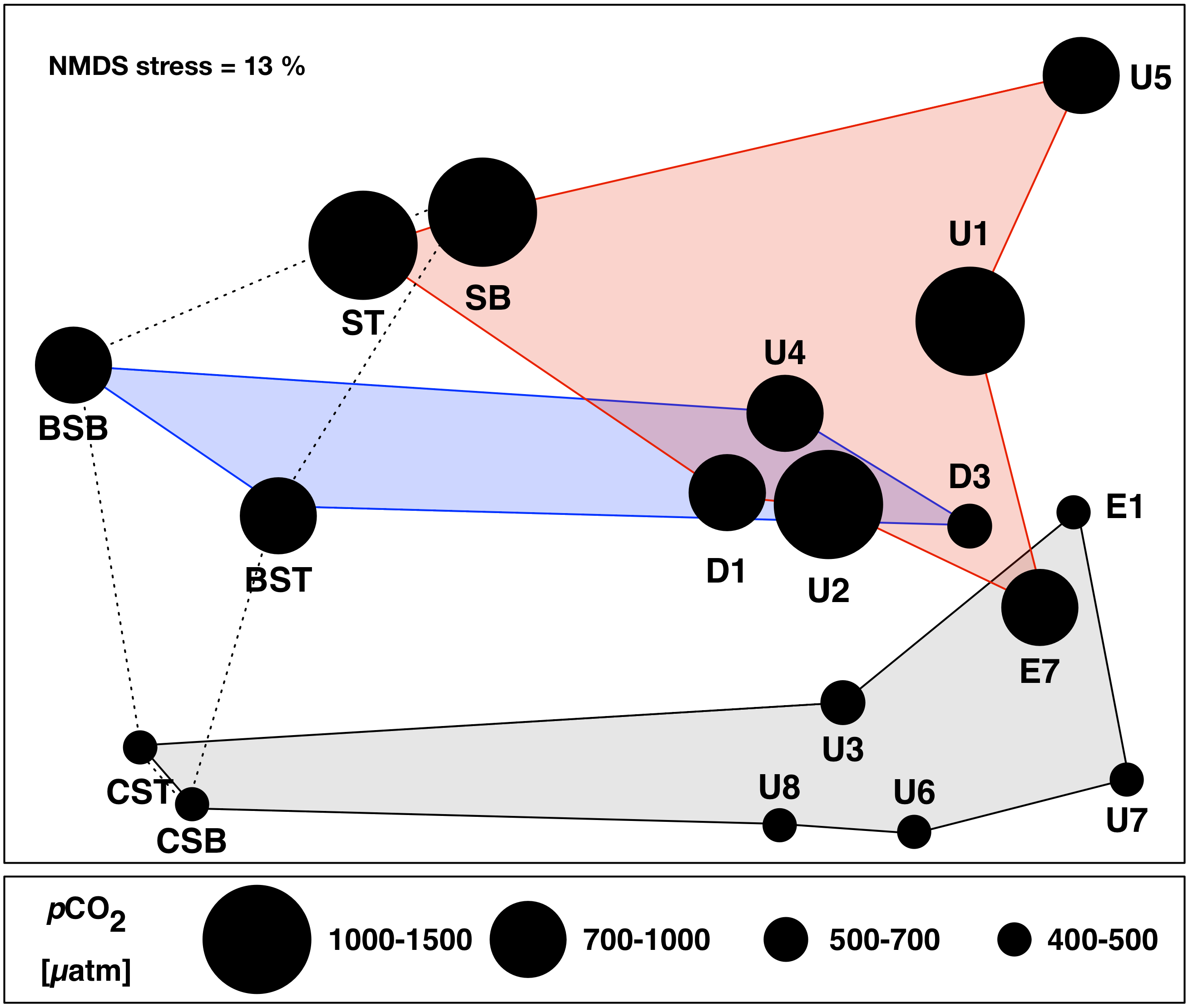
**Supporting Figures**

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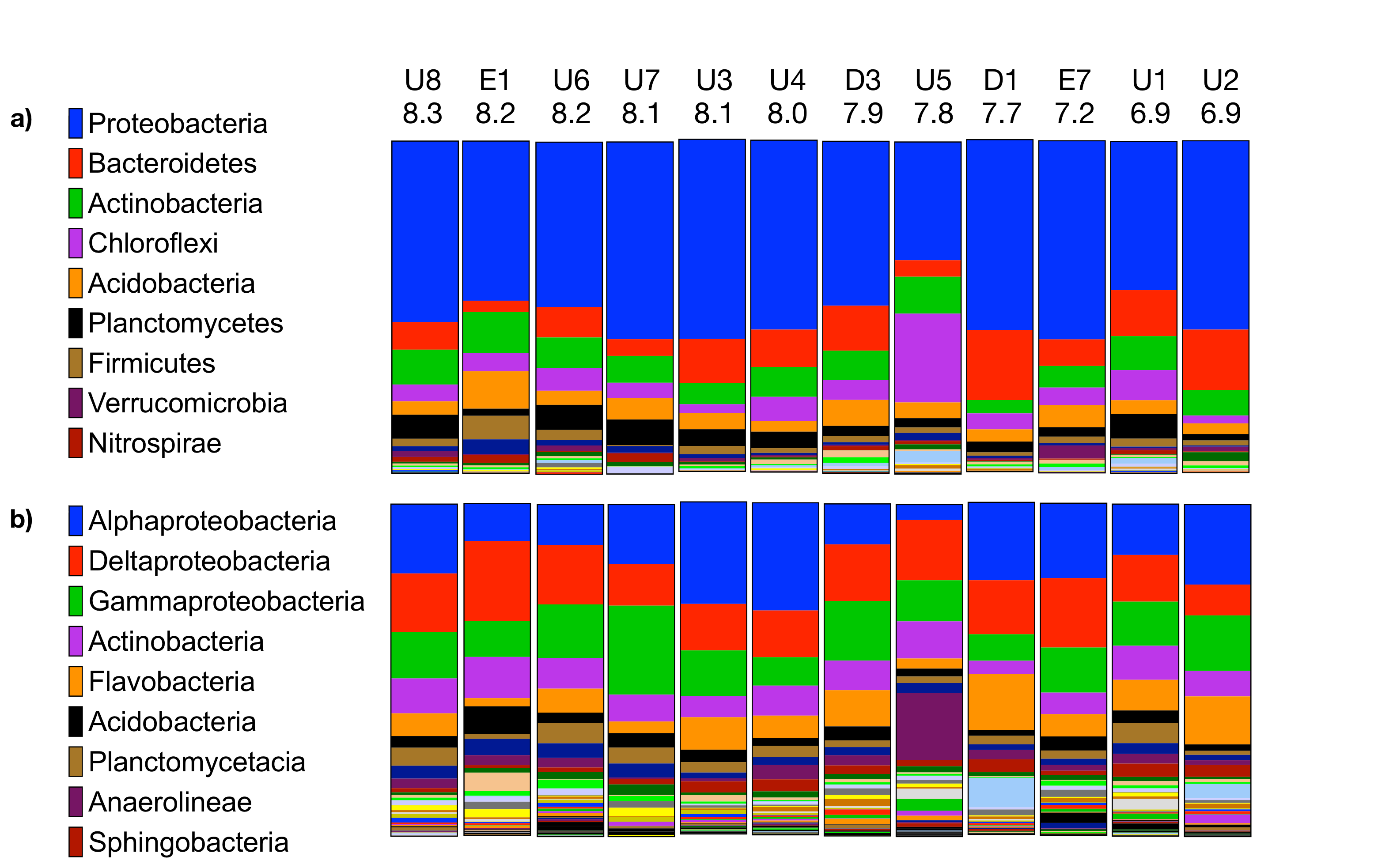
**Figure S1**: Map of sampling area (map from Google 2014), with the seeping sites Upa-Upasina (A), Esa´Ala (B) and Dobu (C). The coordinates for each sample can be found in Table S1.

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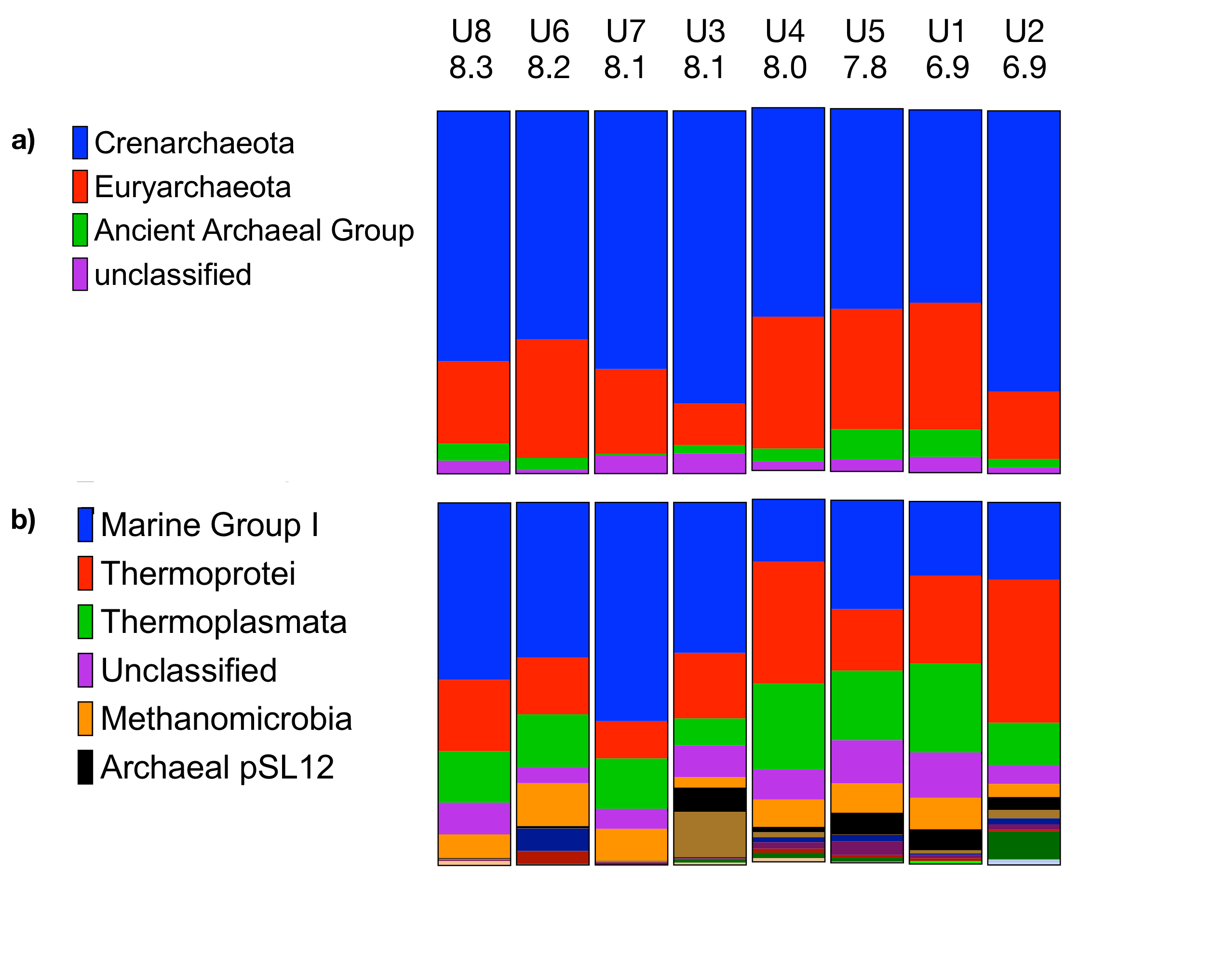
**Figure S2**: Numbers of bacterial ARISA OTU per seawater pH category, i.e. control (pH 8.3-8.1), medium (pH 8.1-7.7) and high (pH 7.7-6.9) *p*CO2 sites.



**Figure S3**: Non-metric multidimensional scaling ordination of Bray-Curtis dissimilarity matrices based on bacterial community data (MPTS OTU3%) from samples of 2010 and 2011 (dotted line). The size of the symbol dots is a (non-linear) representation of differences in *p*CO2 exposure at the respective sites. Colored shapes highlight the three CO2 impact groups “control” (grey), “medium *p*CO2” (blue) and “high *p*CO2” (red).



**Figure S4**: Dominant bacterial phyla (a) and classes (b) at the studied sites. The taxonomic assignments are based on MPTS sequences using the SILVA 16S rRNA reference database. At the top the site names with the respective seawater pH are indicated.



**Figure S5**: Dominant archaeal phyla (a) and classes (b) at the studied sites. The taxonomic assignment is based on MPTS sequences using the SILVA 16S rRNA reference database. At the top the site names with the respective seawater pH are indicated.

**Supporting Tables (Supporting tables.ods [open Document Format])**

**Table S1:** Overview of all 24 samples with time of sampling, sampling location, sampling depth, type of molecular analysis and measurements of environmental parameters.

**Table S2:** Summary of bacterial and archaeal sample diversity, based on ARISA and MPTS and abundance of rare bacterial and archaeal MPTS OTUs.

**Table S3:** Number of bacterial and archaeal OTUs and taxa, characterised by MPTS sequences which either uniquely occur at samples from the categories “high”, “medium” and “control *p*CO2” or show linear or quadratic relationships with pH.

**Table S4:** Bacterial taxa and MPTS OTUs, which were found to significantly increase in terms of relative sequence abundances with increasing seawater *p*CO2.

**Table S5:** Archaeal taxa and MPTS OTUs, which were found to significantly increase in terms of relative sequence abundances with increasing seawater *p*CO2.

**Table S6:** Bacterial taxa and MPTS OTUs, which were found to have a significant quadratic relationship with increasing *p*CO2.

**Table S7:** Archaeal taxa and MPTS OTUs, which were found to have a significant quadratic relationship with increasing *p*CO2.