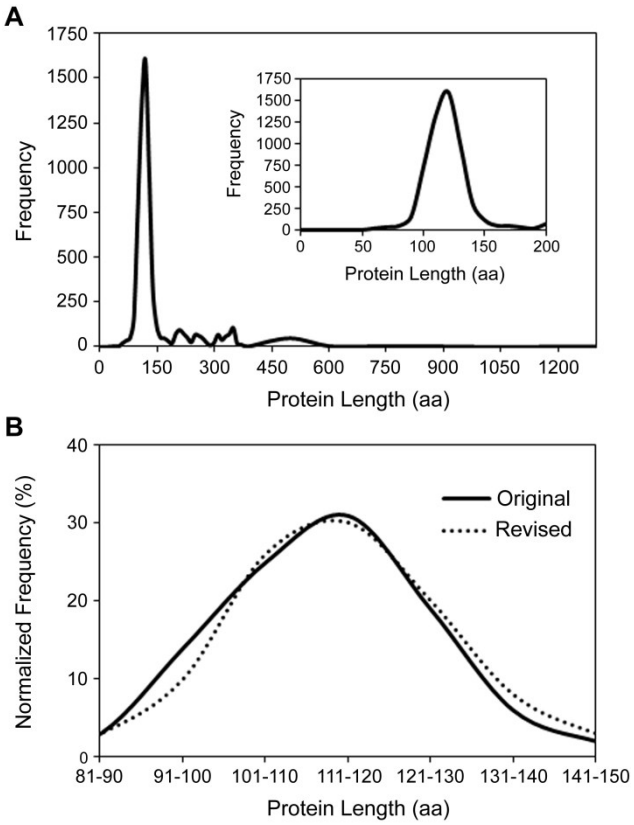
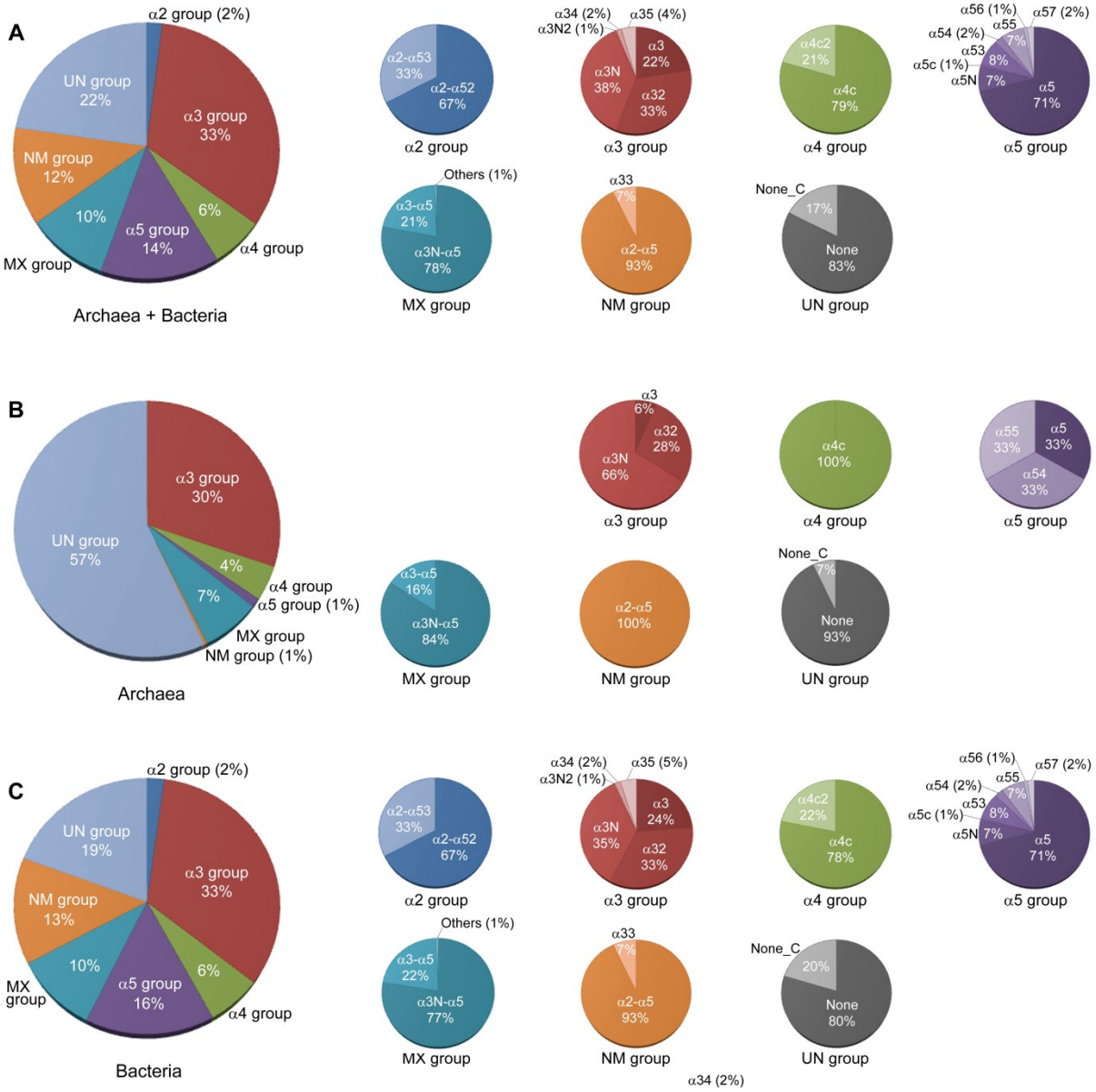
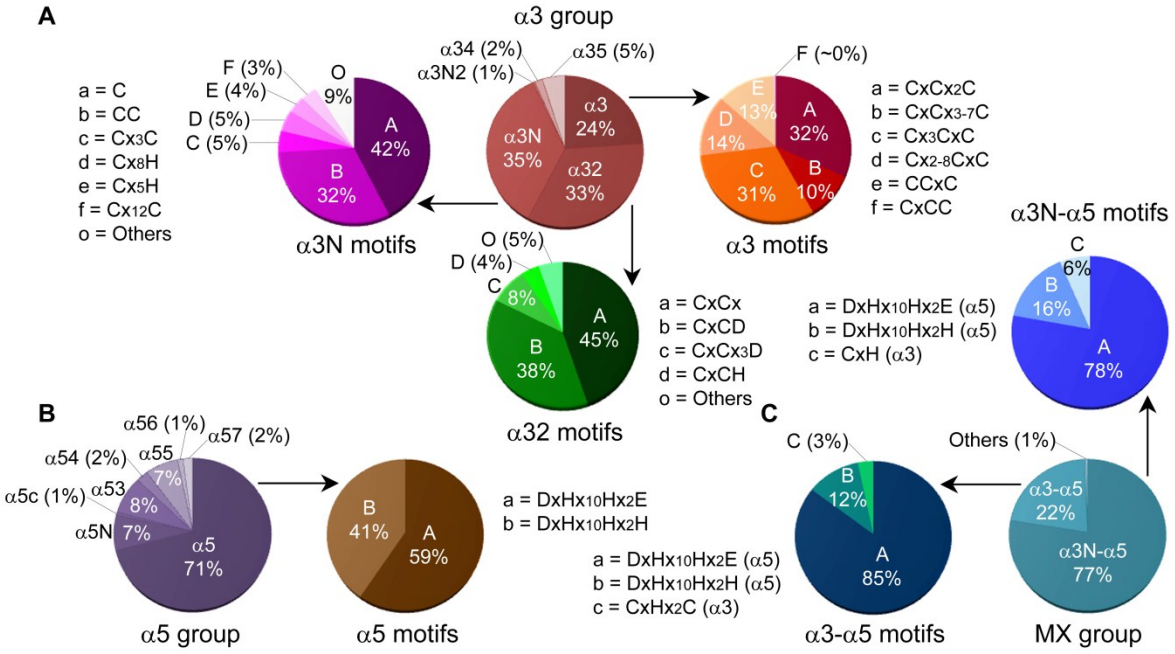
Electronic Supplementary Material (ESI) for Metallomics.



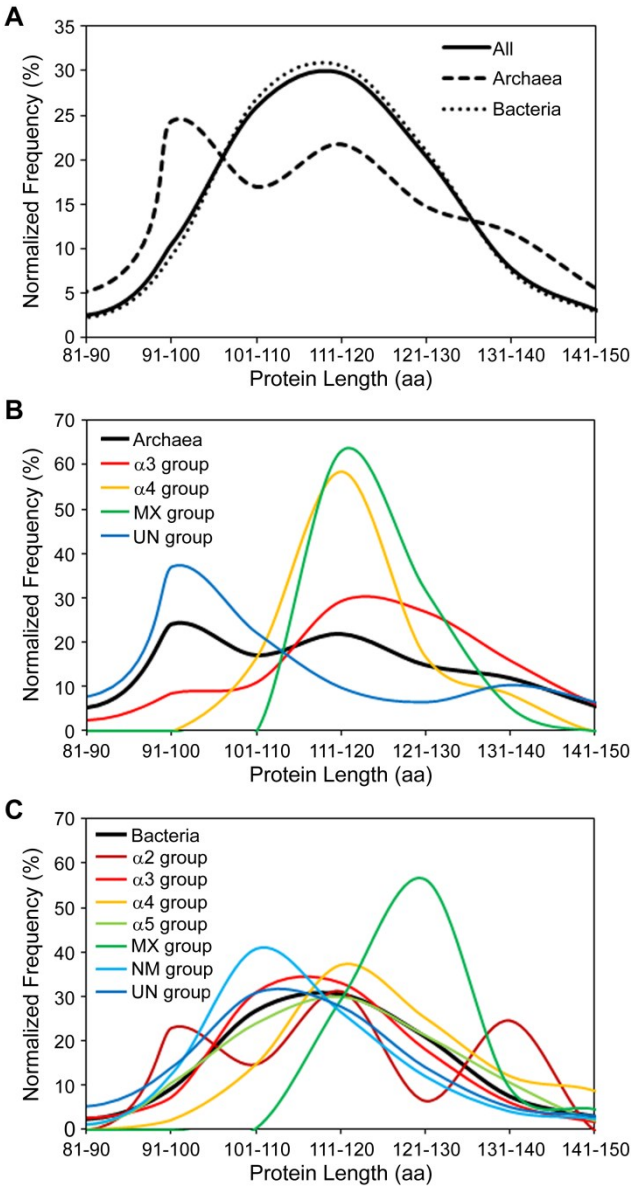
Supplementary Figure S1. (A) Distribution of the ArsR-SmtB family protein lengths (in aa) in bacteria and archaea. This protein length distribution is based on 6478 sequences obtained from the Pfam database. Sequences range from 28 to 1210 amino acids. In the inset, the distribution of sequences range from 81 to 150 is highlighted. (B) Two sets of protein sequences (ranges from 81 to 150 aa) - the original set of 5218 sequences and the revised set of 3104 sequences (removing duplicates and near-identical sequences) are overlaid to show the similarities. Smoothed line option was used to generate the graph plots in excel.



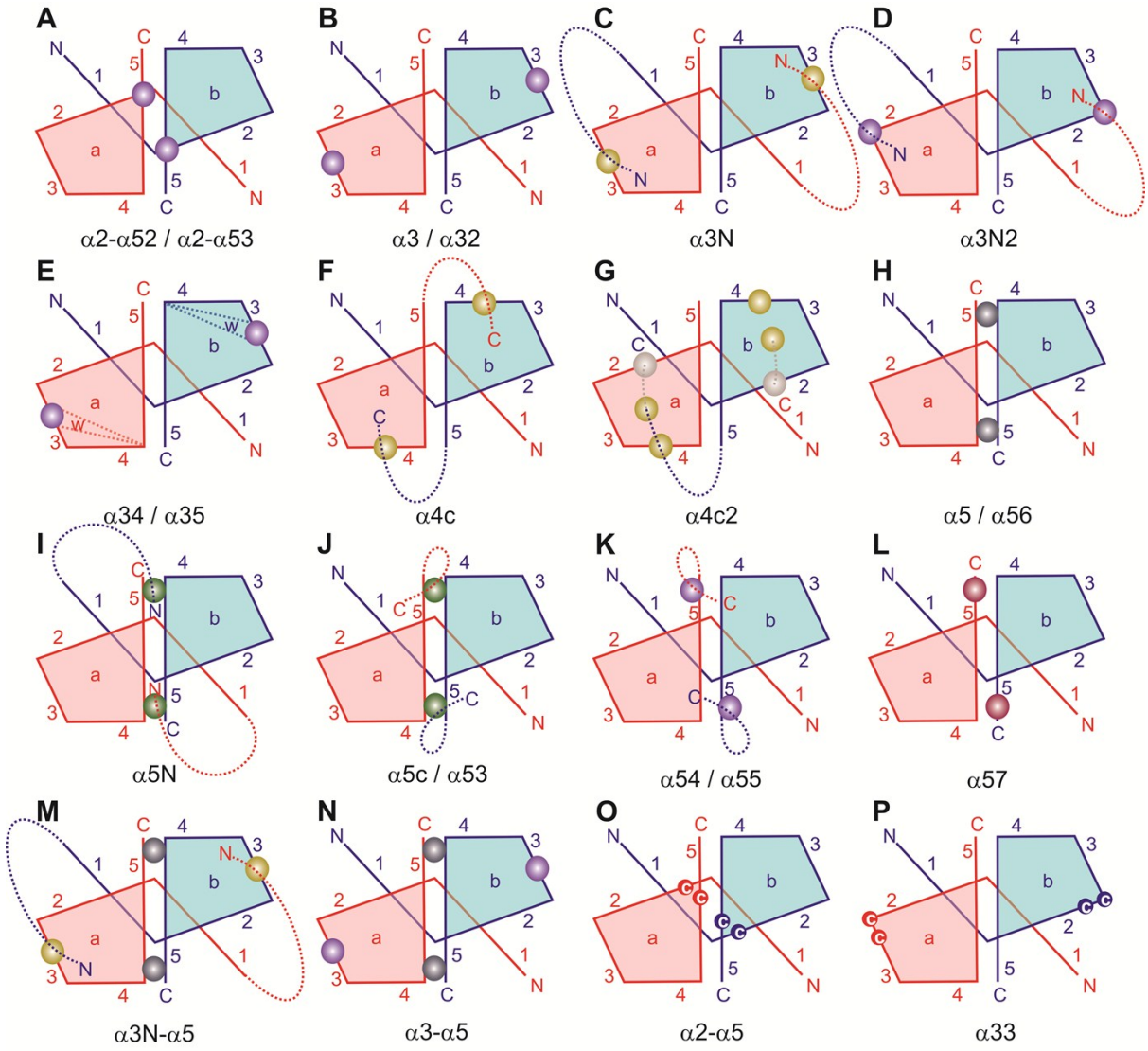
Supplementary Figure S2. Distribution (in percentage) of different motifs in seven groups of metal-/nonmetal-binding sites (α2, α3, α4, α5, MX, NM, and UN) in (A) all 3104 sequences (archaea and bacteria), (B) 271 archaeal sequences, and (C) 2833 bacterial sequences. This is an expanded version of Figure 1 showing the distribution of motifs (class) in each group. The α2 group motifs are not found in archaeal sequences, hence not shown.



Supplementary Figure S3. Distribution (in percentage) of major motif sub-classes found in (A) α3, (B) α5 and (C) α3N-α5 class of bacterial sequences shown in Supplementary Figure S2. (A) The detailed distribution of different α3, α32 and α3N motifs found in the bacterial α3 group. (B) The detailed distribution of different α5 motifs found in the bacterial α5 group. (C) The detailed distribution of different α3N-α5 and α3-α5 motifs found in the bacterial MX group. Different sequence motif sub-classes (denoted in small letters) are shown adjacent to the pie chart of the respective motif figures.



Supplementary Figure S4. (A) Three sets of protein sequences (ranges from 81 to 150 aa) - all 3104 sequences of archaea and bacteria, 271 archaeal sequences and 2833 bacterial sequences are overlaid to show relationships. (B) 271 archaeal sequences are overlaid with different group sequences (α3, α4, MX, and UN) from archaea. The α5 and NM group sequences from archaea are not shown due to a low number of occurrences (<5). The α2 group sequences were not found in archaea, hence not shown. (C) 2833 bacterial sequences are overlaid with different group sequences (α2, α3, α4, α5, MX, NM, and UN) from bacteria. Distribution of the protein lengths in seven groups, α2, α3, α4, α5, MX, NM, and UN, are shown in maroon, red, orange, light green, dark green, sky blue and blue colors, respectively. Smoothed line option was used to generate the graph plots in excel.



Supplementary Figure S5. (A) Cartoon representation of α2-α52 or α2-α53 metal binding site is shown. Cartoon representation of (B) α3 or α32, (C) α3N, (D) α3N2, and (E) α34 or α35 metal binding sites are shown. (E) The dotted triangle with 'w' letter represents the wing comprising β1–β2 strands in between α4 and α5 helices. Cartoon representation of (F) α4c and (G) α4c2 metal binding sites are shown. (G) The cartoon representation of α4c2 site shown represents two different models. One model has two metal binding sites (two orange spheres in the α4 helix and the C-terminal, respectively), and the other model has one metal binding site in the α4 (orange sphere) and one in the α2 (white sphere) helices. Cartoon representation of (H) α5 or α56, (I) α5N, (J) α5c or α53, (K) α54 or α55 and (L) α57 metal binding sites are shown. Cartoon representation of two MX group sites (M) α3N-α5 and (N) α3-α5 are shown. Cartoon representation of (O) α2-α5 and (P) α33 redox-responsive sites are shown. (O-P) Cysteine residues are marked as filled circles. Metal ions are denoted as spheres (purple for As, orange for Cd, grey for Zn, green for Ni, and violet for MAs). Two subunits (a and b) of the protein are indicated in red and blue colors, respectively. N- and C-terminal ends of each subunit are indicated and for each subunit, α-helices are numbered from 1 to 5. The dotted line represents N- or C-terminal extensions.

Supplementary Table S1. The final sequence dataset (3104 sequences) of the ArsR-SmtB family proteins. UniProt IDs of all sequences are mentioned.

B1X005	C3HA99	F4GEA8	G1WWD0	Q6KHJ8	F9VBV0	C9P424	F7XN77	D5SWJ4	A6TP79	C2FX99	C0EHA2	Q9V2K4
F7YSG6	D5TZB9	B7RJ69	A0ZJ51	E6U372	A4YDA4	F7YSG5	D1BFA9	C6WIQ0	E1YF30	G8QZA6	A3TK05	A1U6T8
F2KJ04	I0GL86	A1S1J3	E5WEU8	D9RAP1	D7E7U1	B7VMC1	F3FIP3	F6DQ66	D5NYH3	Q0JWR4	G7ZF47	I0ANJ4
A4YEP0	F0S2N4	C5BEF3	G2RVP2	A9NG94	I0FX86	E8M070	H0QN88	Q84FJ1	D5UHR4	C8XIV0	C5BNB8	A5UTZ6
B4RQF8	E8T2J1	B9MEE7	D5EIW7	F9VEZ2	C6D8J2	A4SFL9	E1UTK6	C2EHL8	C0XR44	H1ZAZ3	D0P6F1	D5UHW1
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C3IA11	E3GM80	D4F868	D4ZDA7	C5USP8	H0UFG7	C7CBJ3	B8H964	A7GR47	E8LK40	E6TPX7	E7FM99	D1C9C0
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B5Y614	E3GHX4	C6RMA4	D0SWJ9	B1HNK9	Q12TH0	B6R8M4	H0JIH0	G2HA70	A4EAP8	H6WCN2	A3HX12	D7CE76
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F2L5A2	C8PXF7	H0RP28	E1SUM6	C4FKQ5	F6B9U7	A3X8H6	G4T298	D7EAJ0	H1LH50	D6Z0R0	C2GKA3	E8N2I0
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O50591	G4CPQ6	D0ZBM2	E6WNM8	C6A3X2	B7K1E3	A3K9Y0	C3PI09	G4L4Z1	A0AWY2	Q31EG1	G0IVR4	C0CUL7
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H6RQ71	F1T776	E8RRE3	D4XQ33	B4U802	D8P7P2	E4L9G9	Q24NC4	H0DZR8	C0ZIU2	H0QWX5	D5P4G0	A1WW26
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C5RAM6	F5S7Z3	Q6D0L8	Q3ELP7	F1YGZ2	Q1GUI7	C1AUI1	H8I4F7	E8PKL8	E4WCD7	F0LM33	C7QDQ0	B7KB27
C7XX99	Q9KEA8	Q5QWI3	C2QKV3	D0S363	A1SKU4	A0R0S0	E0RSV0	F6B329	G5JAE0	G0J0E6	G2GBS6	C8WSS9
Q9KI48	G8S699	E5BEL2	G7GA79	F5I814	H3SF27	E0QIS7	D5CDS3	F7KRE3	D9TSX9	D5BUU6	D7W9Q3	D2QV67
E7R043	D4GVR8	F2G541	D8IAW8	F8DHS5	D5NYY8	C8NFU2	P74986	E4LSQ2	D9RZR8	B2A7P3	G2PE84	C7M1C2
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G0HTB9	G0JQF8	H0TRB5	A8UZ97	A6QA25	Q0RV07	F9W1R5	H2J991	E3PW79	E7NCZ4	A1RD96	A8F6V6	O57801
C7P0D1	C2PGY2	C6RM09	F7V4I9	F5LFL5	C5F8A7	D0WU28	Q5FMZ6	G5FE59	E1QYL4	A0AW45	C0ZBD3	H2CEJ9
D4GTN0	C6WB18	E3I670	F8WKB2	C2TMP8	G0IY78	F3KH79	C0WYD0	D7CLJ4	Q2EFX9	D0L7Y5	H5SV04	F5XHC6
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D1AVJ4	B0SSJ3	B1C8D3	E8U312	G2PN52	A3IFS2	B7WWV6	F5ZAG7	E0RUZ8	C8W806	C2JTI6	Q24P53	G7URV6
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C3I9P8	E4RYN2	F6BC31	C1D3X8	G8R7S6	E8YSX0	D0J7R0	A3Y4W8	G5GIP9	H0QN11	G8QY10	Q212C4	Q46W66
E4RNQ3	D8HAK9	Q04PN2	F8WQU1	A5FFL5	D3PUZ7	A3UEV6	F3NDW1	F4A006	Q9K5Q1	C0VHC1	E2CQ24	D3EQW3
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D5VUC6	D2ZQ55	B5UJY3	Q0ZKJ9	G0L9K7	E6H7E6	D1BPW7	D9PXD4	C8VX22	P30339	D6TIN5	Q3B575	A4U004
D4W829	D0KRQ1	I0L9Q3	E5CIF8	D5BF42	C9R9P0	G4L720	Q9L1V5	C7LTU1	E6TZM5	F0M231	B4S6P9	A5FBK2
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F6D594	C8PP27	B3E731	F7R1V7	I0WQD9	H1D098	H0RMA9	D6SSP3	C7NGD0	B3EE97	I0GTZ4	B5EF53	F7VCD4
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E7GUF5	D9T9G2	F0GY99	Q0G015	G2KV87	D5XB12	C9QHE9	D4H288	F0S2A1	F1TF91	E4Q8N8	E9UVL8	F4LSL9
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H1KYW9	C7RE18	G5FA53	Q7P1X8	E0NJ96	Q8YXV6	E6W701	A3XH18	A6TPQ8	C5VR61	C2YLW4	E1L235	D6U1U0
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A6UVE7	E6TWH1	F8EZJ6	Q89I66	C5D131	F5L7P2	F6ADP2	B1C085	E3IU02	A8MGT7	D3P408	A9KR37	B4TGV5
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Q57682	B8KK49	B0TJA5	C1A7V1	Q21E43	F4GD99	B5I9N2	D2PQ16	C6J9H1	D1PP09	I1D0V4	I0H695	D5QGL9
F6D3H7	A7MXK4	F7YNX5	Q46KP0	F8E9Q0	F2IH40	F4B6Y3	H8IU02	A6P0A4	F9VPL0	E8SGJ1	C7NHY5	D5U235
C9RDU5	A9DRB5	A4Q808	G1V619	G7WP58	F4FWM0	A9VF49	B1MMC4	H7CZ09	C7NDS2	C3WSJ6	I1D4J4	C4SZ57
F0T8G7	A4A7K9	B1L436	F3ALF3	A4CVC7	C3MIL0	F6FW97	Q3A848	Q727R7	F4CYQ8	E3DMI9	D6E6V8	F6ERT6
D3DFU1	B0TXV7	D3EFQ0	D4TFE1	A9DNR4	E6S9T2	E2SE47	G7G5H8	C4L2F1	F4GLG8	C9KL94	B6GE45	D1BG40
A1TA18	A3Q8U9	Q469V6	E4TXK8	A1HUC1	G7ZA59	C3N321	E6RQN0	F7YYQ7	C8PMD4	F9DTU2	H5TTU3	B0JM43
A6UVD2	F4QTE9	G5I9X1	F3MAW5	A6ET09	E4KN11	Q47MI8	B7SKG9	C0CI95	A8ZL51	C8WVN6	A9B0J4	A3DKH1
A8MGV9	A8SW86	E1RDF2	H8XU33	E6X9H2	F4CK97	E2NSB1	Q9EUU5	F1TE71	B1G2L9	H8YZF6	I0RVC6	A5W4V2
D9TSX4	D9UWM4	C2Y3Z8	A6GZY9	E8MXM9	F9MTT5	A6FCH5	E1R6K9	C6JFS1	B9CL93	C7M2N3	D9Q498	Q8PUT9
A9A9M1	Q6L1W0	C3FV37	D2EFM7	F6GBY4	Q0W7Z6	D5RS82	E2S694	B9E869	E6MIE4	H8KTJ9	H8IQ72	A1T2F1
D2JC95	F4Y282	Q027E6	F9VCH5	Q26H69	G8PAX5	C7R057	D1C3J3	Q314U8	G4T1C9	F5LFX5	A2VGC7	C7MB55
C2GK70	Q9YFE8	E3DVB7	A6CKU4	F0RFY8	F3Z432	Q83N40	Q7MCM3	D6XWM8	D2S4W1	E7GBH9	G0CXE6	H0G8Q1
C2EPH0	B5UYW5	A7Z1D6	H0IQL1	F4AYP2	D0D4P8	F9DY38	I0IA13	A1HUB0	G7CIH0	E1QPX0	B7ASA4	B4WC40
C0VYB1	E1YJ72	C4FI52	G4D4P7	H7FVV9	Q0FXC9	A1RDA6	B8G166	A9NHM8	E4NIS9	C5E7X5	Q55940	F5SIH1
H8E8T4	B5IQ09	C1DXK7	D8MHY7	G0J119	A6Q8D5	F3NQG8	D5P4G3	C0GI40	E3B9B0	E2CP83	G5IDD7	Q6AQ20
Q1NUN9	H1Z2T6	E8T4E5	G4EV27	F2IJY4	F5UIZ8	C6D099	A2VJA6	Q12UN1	A4AKQ4	D9SPH8	E5WQI9	D3R0Q4
F8GBT7	G8TJU2	D9QVF4	E9RYX4	Q7NPQ3	G8M128	F0LHX7	I0V8D8	F6DKN4	B2HH01	D6Y3P6	F0T6W0	C1BD59
Q1Q908	F4BA10	E0DC63	A5GR23	D0MF84	Q7CKC2	Q46EY1	H2C1S3	A4J4Q4	Q8ETI4	C7NYS3	C0R0B4	Q8VML1
F5YPZ5	A1RYF7	E0U2G1	G1UTW6	I0I4H0	E6U3W0	Q8TI13	E9UVN2	D0CYI6	F8F350	F4L7N7	F8BVW5	F4LS23
D4W3K5	B9DIE6	E8WN99	B6WXY3	F4GZB5	Q0AYF3	F4HJT4	F5YZ77	I1DUX3	H7GG38	G8RMV4	B3EHR3	E5Y8S7
O29604	E6LDB2	A4FK14	A7NAY9	Q8FLG0	B4S7W1	D5E837	D9UW00	D5VF12	Q5L1Z6	A1TA38	I0A034	D9UQ96
F0T9S4	C2Q5T7	A5GCN6	B9MHG8	G7H576	B3EM94	F8AJA2	F9MNR1	B8HU58	F5YP55	A1UCZ4	A1S0B6	A7HGH8
E0MZB3	C2Z7U7	A5PCL7	C5B5S7	B6W742	C7RBY0	Q8PY24	A5Z5U3	A0LBW7	F7JS14	I0L2L7	D0L844	A1RCV0
O30267	C6WTQ8	D2ZAA4	F3KCI7	G6FQ76	E2SBV7	H3ZMT0	B9MG21	A8TRC0	Q8KE78	D9T082	A3MTF6	B2HN90
B9LWR3	A2CDU1	B1XPW9	A6FPQ9	H0A789	Q8PUU2	B0TA75	H5TWY2	C9LYU2	B8D0P6	A5D5Q8	D2R1C2	D3CRX1
E2S6I0	A8TC72	C6JQW6	A4SQ15	Q38YK0	F2GD97	D8A0T7	D7N823	A1RCC0	G8QWI2	D5UE52	D5E6K8	D3R0R0
C2QZY3	G8S5J2	C4L6C6	G9A790	H1AKZ9	D5EAJ6	Q92ZB9	H1LW73	H5X0M4	A6BJ31	D5UH98	F8A3K8	D1ACS3
D7N913	A3Z6L4	D2J731	D0S8L9	C5ZVB5	Q4LAB6	A4ENM3	B3QFR2	E3GLB8	D7WXP9	C5BXM7	G0Q086	A7IJE0
Q8KBX8	B6G272	G7ZPG3	Q0BZG7	Q65IM9	D2B1R0	A3WJA1	F0SH46	F2L6E5	Q72KG0	Q1J3B6	F4HL10	G7Z2R0
F8ANI1	F5YCA9	Q5HRI2	G0HMP2	E1UV78	B9M9J5	D6V9F2	A3ZS15	D7BR57	B6G2A8	B1MME5	A3TRP7	G5FIG5
F8JP62	E3HCD4	Q4LAB3	H3NGV8	F8KDK5	B2A001	F9QDM2	F2NKW7	Q72ZZ8	H9ZSD1	G2G9U3	D9W8V2	A8KZ20
C0QIC7	B4UHR8	A6TLY0	A1AQC0	G4I113	D3P8T9	C5D8Z8	Q9V061	B0CB29	D1YYC1	E3B863	F6D5Y2	F6D4L2
D5AVK5	D3DK69	F4FXT8	A8TKF7	F8GKN1	Q39YA9	Q82KP1	D4J0W0	B9DJ16	F8FDI6	A1AMC2	E5CIP5	B8J090
G2FN66	Q9K8L0	B9E7M3	H8IKQ2	Q0AT54	Q748P9	I0KVR8	C1A7Z3	F7UHK1	F4FXR3	B5EK86	Q4ZWB8	Q8PUT5
B7GF65	F2RBN7	P45949	C4L214	C8N7V7	B9XS30	C2PDK6	F7NIX9	B5I456	A0R802	A6W5A1	C5ASH6	B4WTL5
E4TF07	O67394	B4X1H5	A1UIY4	H8FSR9	F8G8T2	D9WEW6	F0SUY1	E4RMX1	Q82RZ7	H1Z1W2	Q6SKA1	O58828
F0GTH1	G3GCB1	C4IGI7	F0SHF8	Q0F968	A5GCL2	C4RLZ6	F1TBW1	F4CXD0	Q0G1I3	A1TBB9	F1YGL1	Q0RKE6
G4L907	E3PWS8	D3APB4	D2ZYN4	A6U9X0	B3E6Q1	Q28UY3	E7RK22	Q2JKB0	Q212C1	F4CVQ3	F0HN83	A2SRN6
H6RD88	B8HZS4	F5XI37	G4Q775	G2DVJ0	E4T5F0	D9T9R8	E6VU65	E7NVR8	Q9A4H0	D0LEW7	G8RKQ7	F5XIL8
G5IS31	G4KU60	F0SUS4	Q1QW69	Q8RK00	E6J5U9	C4WFK6	F2JJ26	G1WQA1	Q1IKY6	E6S8J7	H8IT10	G2GDQ6
B1WQ14	A8SDF9	H2J993	F9USR6	F3L399	D5NYJ1	D1FA97	A7VW86	C7UHP0	E1VT73	E2SED3	A0AW51	Q2S6B6
B6AX86	C6LD57	Q0VL20	O53478	A3VJR7	H0QN72	B2J709	H2J987	C7WQN0	Q9HJ03	D5XFN0	C6WRG2	H5X6G0
Q47E55	D5V235	A6TML9	A1TGH2	D8IV76	F9VNU6	C6E9I5	F0JHX5	C6PRG9	A4QFY6	G8RJ93	E9UV56	H5TIV2
Q6LUL6	A4YC59	H2J8S0	E8WLR5	G1Y2T0	D3PXG5	D4H371	I0JLI3	G1WWD1	C0E9N0	C2FAS1	D7H7R8	H3SIG6
A7N569	H0JPY6	C1DUJ4	H1Y355	C6XMS4	Q67NY2	A1T0Q5	F8AE73	C2TR45	G2LEK5	H0JXU5	B0A7H9	H1KWN2
F0T6P7	A6Q0Y9	H1PP18	A3CV60	H1G5K6	C3XL94	G9XVM7	D4LEB1	A3IS01	E7N9T8	C2MP64	D3MQK2	Q13RV9
Q8ZYK0	G2EAK5	B1YFW2	D5EKW5	H5WXR1	C7H1I4	D9QT23	B0P994	C9YZ35	H6R272	C1A1D2	F0RQI7	Q9LCV3
D3FDH2	F5XIM9	Q5WJT4	A6D305	D3PVH2	C5EYS7	C5BHL4	D9SQ59	C6W7Y9	F6B2Q1	F3NQG1	D6S9C8	H1KXC2
Q7NSR9	E2SEQ4	B8G134	E6VTE1	C7NRQ0	G1WFF1	D8G3D4	B2A185	G9EDS3	A4J200	D0LBV7	A5WDR5	F6BAI0
F0PLV0	H1HWP0	Q3AIJ0	D2RJ16	F7PF83	H7EIP9	Q9I1J7	E3HA11	A7NLK2	F3AL88	Q6SK40	A0LMA9	D1YW18
A0K0V1	A6CHI2	H1PYT1	H5U2B6	H1LIC7	A7HVZ7	G8QQ79	F7K243	C7UHP7	D3SAQ0	D3E3S3	D6TQ13	Q9L209
D3STM5	F3B2Z3	H7SG46	B8CIH1	G8P2Z6	C9Z006	F9N6H8	H8YY10	I0V8A8	I0PX92	D9QQ45	D9QBQ8	E9T6W7
D2RWW7	G8N3A8	B6G0W3	A3IQU1	G0FXC4	D7UW72	D5DPM7	F2J5A5	B0CCI7	D9X1V2	Q1J281	H1S1S7	D6TER6
G4RAV9	Q5L2G5	H0JYC6	B4VGZ8	E9UPE6	C8P5I1	Q9K8K6	D7A6S9	G4F4X6	E4TV11	D9R8C0	G2I516	H1D334
D4GS91	E6GYI6	H6R966	D8G9M9	Q38ZB9	E8PLM7	D3FW72	C7CBN2	A8U9A2	D7VT52	F3MIW0	B4WTA9	F8ALP3
F0LH06	C6J3V4	B5Y6Z2	Q8YTF1	A4ETW3	E6L3R5	D3PXF7	A4WZG2	E1QKA7	I1CWF7	D3AG13	F5L322	
A8USM9	D0WY73	G1WQA2	F4XUW9	A3IA91	G0M5U9	C4RLC0	H7EK30	H6LC76	C7Q9D1	H1C6M7	Q82GY9	
C2WGT6	Q6FB66	D4L6Z3	B2IZA5	A3TYD7	D7VD45	Q47DR3	B6IRB0	F8CPF0	A5FFB5	E0E1E2	G7WP78	

Supplementary Table S2. Distribution of different motifs (class and sub-class) in seven metal-/nonmetal-binding groups of the ArsR-SmtB proteins in archaea and bacteria. Archaeal and bacterial phyla are indicated against each motif. Numbers in the bracket indicate the total number of sequences.

Domain	Group	Class	Sub-class¥	Phylum
Archaea (271)	α3 group (82)	α3 (5)	a = $CxCx_2C$ (1)	Euryarchaeota (5)
			b = $CxCx_{3\text{-}7}C$ (2)	
			d = $Cx_{2\text{-}8}CxC$ (1)	
			e = CCxC (1)	
		α3N (54)	a = C (19)	Euryarchaeota (54)
			b = CC (7)	
			c = Cx_3C (13)	
			f = Cx_5H (2)	
			g = Cx_3H (4)	
			o = Others (9)	
		α32 (23)	a = CxCx (11)	Euryarchaeota (23)
			b = CxCD (3)	
			c = $CxCx_3D$ (9)	
	α4 group (12)	α4c (12)	NA£	Crenarchaeota (9) Thaumarchaeota (2) Parvarchaeota (1)
	α5 group (3)	α5 (1)	NA£	Euryarchaeota (2)
		α54 (1)		
		α55 (1)		Thaumarchaeota (1)
	MX group (19)	α3N-α5 (16)	a = $DxHx_{10}Hx_2E$ (α5) (13)	Euryarchaeota (12) Crenarchaeota (1)
			b = $DxHx_{10}Hx_2H$ (α5) (3)	Euryarchaeota (3)
		α3-α5 (3)	a = $DxHx_{10}Hx_2E$ (α5) (2)	Euryarchaeota (3)
			b = $DxHx_{10}Hx_2H$ (α5) (1)	
	NM group (1)	α2-α5 (1)	NA£	Euryarchaeota (1)
	UN group (154)	None (143)	NA£	Euryarchaeota (76) Crenarchaeota (59) Thaumarchaeota (6) Korarchaeota (1) Parvarchaeota (1)
		None_C (11)		Euryarchaeota (7) Crenarchaeota (3) Thaumarchaeota (1)
Bacteria (2833)	α2 group (61)	α2-α52 (41)	NA£	Proteobacteria (24) Cyanobacteria (15) Bacteroidetes (2)
		α2-α53 (20)		Proteobacteria (17) Firmicutes (2) Cyanobacteria (1)
	α3 group (947)	α3 (226)	a = $CxCx_2C$ (72)	Proteobacteria (28) Cyanobacteria (22) Firmicutes (16) Chloroflexi (3) Spirochaetes (1) Synergistetes (1) Thermotogae (1)
			b = $CxCx_{3\text{-}7}C$ (23)	Proteobacteria (15) Firmicutes (4) Actinobacteria (1) Cyanobacteria (1) Fusobacteria (1) Tenericutes (1)
			c = Cx_3CxC (70)	Firmicutes (65) Actinobacteria (1) Aquificae (1) Cyanobacteria (1) Spirochaetes (1) Tenericutes (1)
			d = $Cx_{2\text{-}8}CxC$ (31)	Firmicutes (20) Proteobacteria (7) Actinobacteria (3) Deinococcus-Thermus (1)
			e = CCxC (29)	Firmicutes (21) Proteobacteria (3)

				Chloroflexi (2) Actinobacteria (1) Nitrospirae (1) Thermodesulfobacteria (1)
			f = CxCC (1)	Firmicutes (1)
		α3N (336)	a = C (143)	Firmicutes (55) Proteobacteria (46) Actinobacteria (22) Spirochaetes (5) Thermotogae (5) Bacteroidetes (4) Fusobacteria (2) Aquificae (1) Chloroflexi (1) Cyanobacteria (1) Verrucomicrobia (1)
			b = CC (106)	Actinobacteria (97) Chloroflexi (4) Bacteroidetes (2) Acidobacteria (1) Cyanobacteria (1) Proteobacteria (1)
			c = Cx_3C (16)	Firmicutes (7) Cyanobacteria (4) Actinobacteria (3) Deinococcus-Thermus (1) Proteobacteria (1)
			d = Cx_8H (16)	Actinobacteria (13) Chloroflexi (1) Firmicutes (1) Proteobacteria (1)
			e = $Cx_{12}C$ (13)	Bacteroidetes (13)
			f = Cx_5H (12)	Firmicutes (9) Actinobacteria (1) Cyanobacteria (1) Deinococcus-Thermus (1)
			o = Others (30)	Proteobacteria (11) Actinobacteria (6) Bacteroidetes (4) Firmicutes (3) Cyanobacteria (2) Spirochaetes (2) Chloroflexi (1) Verrucomicrobia (1)
		α3N2 (4)	NA[£]	Actinobacteria (3) Spirochaetes (1)
		α32 (319)	a = CxCx (143)	Proteobacteria (58) Firmicutes (56) Thermotogae (6) Deferribacteres (5) Spirochaetes (3) Acidobacteria (2) Actinobacteria (2) Bacteroidetes (2) Fusobacteria (2) Synergistetes (2) Aquificae (1) Chlorobi (1) Chloroflexi (1) Nitrospirae (1) Thermodesulfobacteria (1)
			b = CxCD (120)	Proteobacteria (44) Firmicutes (32) Actinobacteria (25) Cyanobacteria (9) Deinococcus-Thermus (4) Fusobacteria (2) Gemmatimonadetes (2) Bacteroidetes (1) Thermotogae (1)
			c = $CxCx_3D$ (26)	Firmicutes (16) Chloroflexi (2) Proteobacteria (2) Verrucomicrobia (2)

				Aquificae (1) Fusobacteria (1) Ignavibacteriae (1) Thermotogae (1)
			d = CxCH (13)	Proteobacteria (5) Actinobacteria (3) Firmicutes (3) Planctomycetes (1) Verrucomicrobia (1)
			o = Others (17)	Firmicutes (7) Proteobacteria (5) Cyanobacteria (2) Aquificae (1) Deferribacteres (1) Fusobacteria (1)
		α34 (16)	NA[£]	Deferribacteres (4) Chlorobi (4) Actinobacteria (3) Aquificae (2) Bacteroidetes (2) Firmicutes (1)
		α35 (46)		Bacteroidetes (35) Firmicutes (4) Planctomycetes (3) Proteobacteria (2) Cyanobacteria (1) Chloroflexi (1)
	α4 group (183)	α4c (143)	NA[£]	Actinobacteria (111) Firmicutes (17) Proteobacteria (7) Chloroflexi (5) Acidobacteria (1) Bacteriodetes (1) Cyanobacteria (1)
		α4c2 (40)		Actinobacteria (40)
	α5 group (441)	α5 (313)	a = $DxHx_{10}Hx_2E$ (186)	Firmicutes (72) Actinobacteria (58) Proteobacteria (29) Spirochaetes (10) Cyanobacteria (8) Fusobacteria (3) Thermotogae (2) Cloacimonetes (1) Deferribacteres (1) Deinococcus-Thermus (1) Synergistetes (1)
			b = $DxHx_{10}Hx_2H$ (127)	Firmicutes (78) Actinobacteria (34) Chloroflexi (5) Synergistetes (3) Proteobacteria (2) Acidobacteria (1) Deferribacteres (1) Deinococcus-Thermus (1) Fibrobacteres (1) Thermotogae (1)
		α5N (33)	NA[£]	Actinobacteria (30) Proteobacteria (2) Firmicutes (1)
		α5c (3)		Firmicutes (2) Chloroflexi (1)
		α53 (36)		Actinobacteria (36)
		α54 (11)		Actinobacteria (5) Cyanobacteria (2) Firmicutes (2) Proteobacteria (1) Verrucomicrobia (1)
		α55 (31)		Proteobacteria (14) Actinobacteria (13) Planctomycetes (2) Firmicutes (1) Verrucomicrobia (1)
		α56 (5)		Proteobacteria (3) Planctomycetes (1)

				Cloacimonetes (1)
		α57 (9)		Proteobacteria (3) Cyanobacteria (2) Actinobacteria (2) Planctomycetes (1) Nitrospirae (1)
	MX group (280)	α3N-α5 (217)	a = $DxHx_{10}Hx_2E$ (α5) (169)	Firmicutes (109) Deinococcus-Thermus (15) Proteobacteria (15) Cyanobacteria (11) Chlorobi (4) Spirochaetes (4) Fusobacteria (3) Chloroflexi (2) Thermotogae (2) Synergistetes (1) Acetothermia (1) Deferribacteres (1) Gemmatimonadetes (1)
			b = $DxHx_{10}Hx_2H$ (α5) (34)	Firmicutes (21) Actinobacteria (8) Proteobacteria (2) Acidobacteria (1) Cyanobacteria (1) Fusobacteria (1)
			c = CxH (α3) (14)	Actinobacteria (7) Firmicutes (3) Spirochaetes (2) Bacteroidetes (1) Chloroflexi (1)
		α3N-α5c (1)	NA[£]	Firmicutes (1)
		α3N-α4c (1)		Firmicutes (1)
		α3-α5 (61)	a = $DxHx_{10}Hx_2E$ (α5) (52)	Firmicutes (45) Chlorobi (2) Proteobacteria (2) Spirochaetes (1) Synergistetes (1) Thermotogae (1)
			b = $DxHx_{10}Hx_2H$ (α5) (7)	Firmicutes (5) Acidobacteria (1) Tenericutes (1)
			C = $CxHx_2C$ (α3) (2)	Firmicutes (2)
	NM group (381)	α2-α5 (354)	NA[£]	Proteobacteria (327) Firmicutes (9) Chlorobi (7) Cyanobacteria (5) Actinobacteria (1) Bacteroidetes (2) Fusobacteria (1) Spirochaetes (1) Verrucomicrobia (1)
		α33 (27)		Firmicutes (17) Proteobacteria (4) Spirochaetes (2) Tenericutes (2) Actinobacteria (1) Synergistetes (1)
	UN group (540)	None (430)	NA[£]	Firmicutes (151) Actinobacteria (113) Proteobacteria (102) Bacteroidetes (15) Spirochaetes (13) Planctomycetes (8) Chloroflexi (6) Acidobacteria (5) Cyanobacteria (4) Deferribacteres (3) Aquificae (2) Nitrospirae (2) Acetothermia (1) Dictyoglomi (1) Fibrobacteres (1) Fusobacteria (1) Thermotogae (1)

				Verrucomicrobia (1)
		None_C (110)		Firmicutes (37) Proteobacteria (19) Aquificae (14) Actinobacteria (10) Bacteroidetes (4) Chlorobi (4) Cyanobacteria (4) Caldiserica (2) Chloroflexi (2) Chrysiogenetes (2) Deferribacteres (2) Fusobacteria (2) Spirochaetes (2) Acidobacteria (1) Dictyoglomi (1) Synergistetes (1) Tenericutes (1) Thermotogae (1) Verrucomicrobia (1)

¥ 'x' denotes any amino acid, C = cysteine, D = aspartic acid, H = histidine, E = glutamic acid; small letters (a, b, c, ..., o) denotes different motif types

£ NA, not applicable

Supplementary Table S3. Distribution of different archaeal and bacterial phyla in seven metal-/nonmetal-binding groups (α2, α3, α4, α5, MX, NM and UN) of the ArsR-SmtB family.

Archaea (271)						
α2 group (0)	**α3 group (82)**	**α4 group (12)**	**α5 group (3)**	**MX group (19)**	**NM group (1)**	**UN group (154)**
	Euryarchaeota (82)	Crenarchaeota (9) Thaumarchaeota (2) Parvarchaeota (1)	Euryarchaeota (2) Thaumarchaeota (1)	Euryarchaeota (18) Crenarchaeota (1)	Euryarchaeota (1)	Euryarchaeota (83) Crenarchaeota (62) Thaumarchaeota (7) Korarchaeota (1) Parvarchaeota (1)
Bacteria (2833)						
α2 group (61)	**α3 group (947)**	**α4 group (183)**	**α5 group (441)**	**MX group (280)**	**NM group (381)**	**UN group (540)**
Proteobacteria (41) Cyanobacteria (16) Bacteroidetes (2) Firmicutes (2)	Firmicutes (321) Proteobacteria (229) Actinobacteria (184) Bacteroidetes (63) Cyanobacteria (45) Chloroflexi (16) Spirochaetes (13) Thermotogae (14) Deferribacteres (10) Fusobacteria (9) Aquificae (7) Deinococcus-Thermus (7) Chlorobi (5) Verrucomicrobia (5) Planctomycetes (4) Acidobacteria (3) Synergistetes (3) Gemmatimonadetes (2) Nitrospirae (2) Tenericutes (2) Thermodesulfobacteria (2) Ignavibacteriae (1)	Actinobacteria (151) Firmicutes (17) Proteobacteria (7) Chloroflexi (5) Acidobacteria (1) Bacteroidetes (1) Cyanobacteria (1)	Actinobacteria (178) Firmicutes (156) Proteobacteria (54) Cyanobacteria (12) Spirochaetes (10) Chloroflexi (6) Planctomycetes (4) Synergistetes (4) Fusobacteria (3) Thermotogae (3) Cloacimonetes (2) Deferribacteres (2) Deinococcus-Thermus (2) Verrucomicrobia (2) Acidobacteria (1) Fibrobacteres (1) Nitrospirae (1)	Firmicutes (187) Proteobacteria (19) Actinobacteria (15) Deinococcus-Thermus (15) Cyanobacteria (12) Spirochaetes (7) Chlorobi (6) Fusobacteria (4) Thermotogae (3) Chloroflexi (3) Synergistetes (2) Acidobacteria (2) Acetothermia (1) Deferribacteres (1) Gemmatimonadetes (1) Tenericutes (1) Bacteroidetes (1)	Proteobacteria (331) Firmicutes (26) Chlorobi (7) Cyanobacteria (5) Spirochaetes (3) Actinobacteria (2) Bacteroidetes (2) Tenericutes (2) Fusobacteria (1) Verrucomicrobia (1) Synergistetes (1)	Firmicutes (188) Actinobacteria (123) Proteobacteria (121) Bacteroidetes (19) Aquificae (16) Spirochaetes (15) Chloroflexi (8) Cyanobacteria (8) Planctomycetes (8) Acidobacteria (6) Deferribacteres (5) Chlorobi (4) Fusobacteria (3) Caldiserica (2) Chrysiogenetes (2) Dictyoglomi (2) Nitrospirae (2) Thermotogae (2) Verrucomicrobia (2) Acetothermia (1) Fibrobacteres (1) Synergistetes (1) Tenericutes (1)

Supplementary Table S4. Propensities of 20 amino acid residues in the ArsR-SmtB family. Propensities of residues in major phyla of archaea and bacteria are indicated.

Amino Acid	Archaea + Bacteria	Archaea			Bacteria			
		All	Euryarchaeota	Crenarchaeota	All	Firmicutes	Proteobacteria	Actinobacteria
Ala	8.5	5.3	5.5	5.0	8.9	5.5	9.6	13.3
Arg	7.0	6.8	6.7	7.2	7.0	5.1	7.9	8.6
Asn	3.1	4.2	4.0	4.6	3.0	4.1	2.9	1.4
Asp	5.4	5.2	5.8	3.6	5.4	5.8	4.9	5.9
Cys	2.7	1.7	2.1	0.7	2.8	2.9	2.7	2.4
Gln	4.3	2.5	2.5	2.2	4.4	4.5	5.4	3.1
Glu	7.4	9.3	9.7	8.6	7.2	8.3	7.0	6.0
Gly	4.8	5.0	4.7	5.6	4.8	3.9	4.6	6.6
His	3.1	2.3	2.4	2.1	3.2	3.7	2.5	3.5
Ile	5.6	7.8	7.6	8.1	5.4	7.3	4.8	3.1
Leu	13.2	13.1	12.6	14.6	13.2	12.2	14.4	13.2
Lys	5.7	8.5	8.4	8.4	5.4	8.2	4.6	2.0
Met	2.9	2.9	3.0	2.7	2.9	3.3	3.1	2.2
Phe	2.2	2.5	2.4	2.4	2.2	2.8	1.9	1.3
Pro	3.1	2.3	2.5	1.9	3.2	1.8	3.3	5.3
Ser	6.7	6.7	6.3	7.7	6.7	6.7	7.0	6.5
Thr	4.6	3.9	3.9	3.7	4.7	4.4	4.4	5.5
Trp	0.6	0.7	0.7	0.6	0.6	0.4	0.7	0.5
Tyr	2.5	3.5	3.3	4.1	2.4	3.0	2.3	1.6
Val	6.6	5.9	5.8	6.3	6.7	6.2	6.0	8.2

Supplementary Table S5. Propensities (in percentage) of 20 amino acids in the ArsR-SmtB family. Propensities of residues in different groups and motifs are shown.

Amino Acid	α2 group			α3 group							α4 group			α5 group									MX group			NM group			UN group		
	All	α2-α52	α2-α53	All	α3	α3N	α3N2	α32	α34	α35	All	α4c	α4c2	All	α5	α5N	α5c	α53	α54	α55	α56	α57	All	α3N-α5	α3-α5	All	α2-α5	α33	All	None	None_C
Ala	9.4	9.0	10.3	7.8	6.2	9.5	6.2	7.0	6.7	7.4	12.5	12.5	12.9	9.2	8.3	13.7	6.7	12.5	11.4	9.3	7.4	9.9	7.7	7.9	7.1	9.7	9.9	7.2	7.7	7.9	6.7
Arg	6.9	6.9	6.9	6.1	5.6	6.6	5.8	6.5	5.2	2.6	8.6	8.2	10.2	7.3	6.8	9.0	6.4	9.7	6.8	7.5	6.3	7.7	6.2	6.5	5.4	7.5	7.6	5.8	7.5	7.8	6.1
Asn	3.5	3.5	3.6	3.3	3.7	2.6	1.1	3.6	4.9	4.7	2.3	2.5	1.9	2.4	2.5	1.2	2.2	2.5	2.4	2.2	2.5	2.8	2.9	2.7	3.4	3.1	3.0	4.5	3.7	3.7	3.8
Asp	3.5	3.6	3.3	5.1	5.4	5.2	4.3	5.0	4.2	4.2	6.1	5.8	7.4	6.2	6.4	5.4	5.3	6.3	5.4	5.5	6.3	4.2	6.8	6.8	6.9	4.6	4.6	4.9	5.0	5.0	4.6
Cys	3.5	3.4	3.7	3.9	5.0	3.9	4.1	3.0	4.1	5.2	3.8	3.4	5.2	1.4	1.0	0.8	0.8	0.4	4.1	4.6	4.8	3.9	3.2	3.0	4.0	2.4	2.4	3.6	1.1	0.8	2.4
Gln	6.3	6.6	5.7	4.1	4.5	3.7	6.0	4.2	3.6	5.4	2.7	3.0	1.9	4.4	4.4	4.0	4.7	4.1	4.1	5.2	4.8	3.7	3.9	4.0	3.7	5.8	5.9	4.2	4.0	3.9	4.5
Glu	7.0	7.1	6.9	7.3	7.3	7.1	5.8	7.6	8.1	7.2	5.7	5.8	4.9	7.1	7.6	5.7	7.5	6.5	7.2	5.4	5.0	5.7	8.4	8.4	8.2	7.4	7.5	6.9	7.9	7.9	8.0
Gly	4.4	4.4	4.4	4.6	3.8	5.0	5.6	4.3	4.5	6.0	7.0	7.1	6.9	4.9	4.4	7.8	4.7	5.3	4.5	4.8	4.8	6.5	3.8	3.8	3.6	4.7	4.8	3.8	5.1	5.2	4.6
His	2.0	1.6	2.8	2.6	3.0	2.5	2.8	2.5	2.7	2.5	2.5	2.6	2.3	4.9	5.2	5.5	8.3	5.5	2.6	2.7	3.0	3.1	4.2	4.2	4.3	2.3	2.2	4.3	2.8	2.8	2.8
Ile	5.3	5.2	5.3	6.1	6.1	5.2	4.7	6.8	6.6	8.8	3.4	3.3	3.8	5.0	5.8	3.2	7.5	2.9	3.7	3.4	4.6	3.5	6.0	5.7	6.9	4.8	4.7	6.6	6.4	6.3	6.6
Leu	12.8	12.3	14.0	13.4	13.4	13.5	13.9	13.8	11.6	10.8	13.7	13.6	14.0	12.9	12.9	14.5	11.4	10.0	13.9	14.0	13.0	15.0	11.7	12.1	10.3	14.4	14.6	11.7	13.1	13.1	13.2
Lys	4.8	4.9	4.4	6.6	6.9	5.6	4.5	7.4	7.1	8.4	2.0	2.4	0.6	4.4	4.9	0.7	4.2	2.5	4.9	4.3	8.6	4.3	6.7	6.4	7.9	4.7	4.5	6.6	6.5	6.2	8.0
Met	3.0	2.8	3.6	2.7	3.0	2.5	2.8	2.8	2.8	2.4	2.0	2.0	2.0	2.8	2.9	2.0	3.1	2.8	2.8	3.0	4.6	2.8	3.2	3.1	3.2	3.5	3.4	3.8	2.8	2.9	2.6
Phe	2.2	2.5	1.5	2.3	2.6	1.9	2.8	2.4	2.5	3.3	1.4	1.6	0.7	2.2	2.4	0.3	1.9	2.0	1.8	2.0	2.7	2.1	2.7	2.6	3.0	1.6	1.5	2.9	2.5	2.5	2.7
Pro	3.7	3.5	3.9	3.5	3.0	4.1	4.9	3.1	3.2	2.9	4.2	3.9	5.2	3.2	2.9	4.2	3.9	4.0	3.4	4.7	1.3	3.8	1.6	1.8	1.0	2.8	2.8	3.1	3.1	3.2	2.8
Ser	7.5	7.6	7.2	6.7	7.0	6.7	10.9	6.5	6.0	5.3	6.6	6.8	5.7	7.1	7.3	6.4	5.0	6.2	6.8	7.5	6.1	7.0	6.1	6.1	5.9	7.3	7.3	7.6	6.5	6.5	6.6
Thr	4.9	4.6	5.8	4.5	4.5	4.9	7.1	4.0	6.4	4.9	5.8	5.9	5.4	4.5	4.4	5.3	2.8	5.0	6.3	3.4	4.4	5.3	4.1	4.0	4.4	4.4	4.5	3.4	4.6	4.8	4.0
Trp	0.2	0.2	0.1	1.0	1.2	0.9	0.2	1.0	0.8	0.7	0.1	0.1	0.0	0.2	0.2	0.0	0.3	0.5	0.6	0.9	0.2	0.1	0.1	0.1	0.0	0.2	0.2	0.7	0.7	0.7	0.5
Tyr	2.2	2.2	2.3	2.4	2.6	2.2	0.6	2.6	2.2	2.0	2.0	2.0	1.9	2.2	2.3	2.1	3.6	1.4	1.7	1.7	3.0	1.3	2.6	2.5	2.7	2.6	2.6	2.8	2.7	2.7	2.8
Val	7.0	8.2	4.3	5.8	5.1	6.3	6.0	5.7	7.0	5.4	7.5	7.6	7.1	7.7	7.4	8.2	9.7	10.0	5.5	7.9	6.9	7.4	8.2	8.3	8.0	6.0	6.0	5.5	6.3	6.2	6.7

Supplementary Table S6. Protein names and the corresponding UniProt IDs of 10 different metal-binding protein families (MerR, CsoR, CopY, NikR, Fur, DtxR, LysR, GntR, TetR, and MarR) are shown. The sequences of these proteins were used for the PCA and the result is shown in Figure 3D.

MerR		CsoR		CopY		NikR		Fur		DtxR		LysR		GntR		TetR		MarR	
Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID	Protein Name	UniProt ID
MerR	P0A183	CsoR	P9WP49	CopR	Q9CHA6	NikR	P0A6Z6	Fur	V6F4Q0	IdeR	P9WMH1	OxyR	P0ACQ4	GntR	P10585	ComR	P75952	AdcR	Q04I02
ZntR	C3SC52	CsoR	O32222	CopY	Q47839	NikR	O25896	PerR	A0A0E0UWH0	MntR	Q99VY1	ModE	P0A9G8	CGL2915†	Q8NLM6	TetR	A0A081PX32	HucR	Q9RV71
CueR	C3TL37	CsoR	A0A151FPC8	CopY	Q9F683	NikR	D0ZAF0	Fur	Q03456	ScaR	Q9RFN3	FkbR1	Q9KIF0	FadR	P0A8V6	TetR	P0ACT4	SlyA	Q82ZP8
PbrR	Q58AJ5	RcnR	P64530	CopY	A0A081PVW3	NikR	C0Q138	Irr	O85206	TroR	F7IW50	MopA	A0A0M3FG26	CitO	Q2KKB8	HrtR	Q9CHR1	OhrR	O34777
SoxR	P0ACS2	DmeR	NA§	CopY	D0R6D0			Mur	Q1MMB4	SirR	Q5HRA0	OxyR	Q5ZZ38	TM0439†	Q9WYS0	XCC2027†	Q8P940	MarR	P27245
MerR	P22874	RicR	O07434					Nur	Q9K4F8	IdeR	P9WMH1	MopB	Q08386	GfcR	A0QUD9	MSMEG_3765†	A0QYS0	ZitR	A2RNS2
CadR	Q93TP7	InrS	Q55554					Zur	Q65HA6	DtxR	WP_010935052‡	LysR	P03030	Rv0494†	P9WMG7	Rv1685c†	O33187	MexR	P52003
TipA	P0A4T9	CopU	G2FXW0					Fur	Q65HT6	SloR	I6L923			LldR	I1Y872	SczA	Q9F8C4	MgrA	P0C1S0
NolA	P22537	FrmR	P0AAP3					FurR	Q8DVA3	MtsR	B4U486			PS5454†	Q87U60	SczA	A0A150NGQ9	SarZ	Q5HDG9
BmrR	P39075	CstR	NA§					FurA	P9WN87					GntR	P46833	SczA	Q8DNK2	Rv2887†	P9WME9
NimR	NP_439766.1‡															PsaR	I6L8Z6		
GolS	Q8ZRG6																		
CoaR	NA§																		
AdhC	P44557																		
NmlR	A0A0U1RJ76																		

§NA = Not Available

‡GenBank ID

†Gene Name

Supplementary Table S7. (A) Sequence length (aa) distribution of the ArsR-SmtB family proteins. (B) Sequence length (aa) distribution in different archaeal and bacterial metal-/nonmetal-binding groups (α2, α3, α4, α5, MX, NM and UN) of the ArsR-SmtB family.

(A)

Domain	Average	Std. Dev.§
Archaea + Bacteria	114.6	12.7
Archaea	112.8	16.6
Bacteria	114.8	12.3

§Standard Deviation

(B)

Group	Archaea		Bacteria	
	Average	Std. Dev.§	Average	Std. Dev.§
α2 Group	-	-	115.4	13.8
α3 Group	120.0	13.7	113.8	11.4
α4 Group	117.1	7.1	121.2	11.7
α5 Group	116.3	20.5	115.3	12.2
MX Group	120.1	5.7	124.2	6.8
NM Group	107.0	-	111.1	11.8
UN Group	107.8	17.6	110.6	14.3

§Standard Deviation

Supplementary Table S8. Sequence lengths (aa) distribution in different archaeal and bacterial groups (α2, α3, α4, α5, MX, NM, and UN) in the ArsR-SmtB family. Each group has multiple classes of motifs and some classes comprised of several sub-classes. Class-specific averages with standard deviations and individual sub-class averages with standard deviations are mentioned, where applicable.

Domain	Group	Class	Sub-class[¥]	Average	Std. Dev.[§]
Archaea	α2 Group	α2-α52	NA[£]	-	-
		α2-α53		-	-
	α3 Group	α3	NA[£]	123.6	10.5
		α3N		124.5	10.5
		α3N2		-	-
		α32		108.5	14.6
		α34		-	-
		α35		-	-
	α4 Group	α4c	NA[£]	117.1	7.1
		α4c2		-	-
	α5 Group	α5	NA[£]	111.0	-
		α5N		-	-
		α5c		-	-
		α53		-	-
		α54		139.0	-
		α55		99.0	-
		α56		-	-
	MX Group	α3N-α5	NA[£]	119.1	5.6
		α3N-α5c		-	-
		α3N-α4c		-	-
		α3-α5		125.3	1.5
	NM Group	α2-α5	NA[£]	107.0	-
		α33		-	-
	UN Group	None	NA[£]	106.3	16.2
		None_C		127.5	23.6
Bacteria	α2 Group	α2-α52	NA[£]	120.9	12.2
		α2-α53		104.1	9.4
	α3 Group	α3	a = $CxCx_2C$	113.3	10.9
			b = $CxCx_{3-7}C$	116.2	8.2
			c = Cx_3CxC	108.9	6.3
			d = $Cx_{2-8}CxC$	112.5	10.4
			e = CCxC	113.3	7
			f = CxCC	99.0	-
			Overall	112.0	9.1
		α3N	a = C	112.5	12.7
			b = CC	123.6	7.6
			c = Cx_3C	124.4	8.2
			d = Cx_8H	126.4	8.7
			e = $Cx_{12}C$	126.5	4.4
			f = Cx_5H	119.7	6.6
			o = Others	115.9	8.3
			Overall	118.3	11.5
		α3N2	NA[£]	116.8	1.5
		α32	a = CxCx	113.6	12.8

			b = CxCD	109.0	10.1
			c = $CxCx_3D$	107.7	10.9
			d = CxCH	108.9	9.5
			o = Others	108.9	10.0
			Overall	111.0	11.6
		α34	NA[£]	108.7	11.6
		α35		111.2	8.8
	α4 Group	α4c	NA[£]	122.6	11.8
		α4c2		116.2	9.8
	α5 Group	α5	a = $DxHx_{10}Hx_2E$	115.0	12.1
			b = $DxHx_{10}Hx_2H$	111.8	11.2
			Overall	113.7	11.8
		α5N	NA[£]	122.5	7.8
		α5c		120.0	12.1
		α53		125.3	10.5
		α54		114.8	17.4
		α55		116.7	9.8
		α56		105.0	7.1
		α57		106.0	11.6
	MX Group	α3N-α5	a = $DxHx_{10}Hx_2E$ (α5)	124.3	6.2
			b = $DxHx_{10}Hx_2H$ (α5)	124.4	7.1
			c = CxH (α3)	133.4	11.5
			Overall	124.9	7.1
		α3N-α5c		148.0	-
		α3N-α4c		103.0	-
		α3-α5	a = $DxHx_{10}Hx_2E$ (α5)	121.7	2.9
			b = $DxHx_{10}Hx_2H$ (α5)	121.1	2.4
			c = $CxHx_2C$ (α3)	122.0	-
			Overall	121.6	2.8
	NM Group	α2-α5	NA[£]	110.7	11.6
		α33		115.7	13.1
	UN Group	None	NA[£]	110.3	14.3
		None_C		112.1	14.6

[§]Standard Deviation

[¥] 'x' denotes any amino acid, C = cysteine, D = aspartic acid, H = histidine, E = glutamic acid; small letters (a, b, c, ..., o) denotes different motif types

[£] NA, not applicable

Supplementary Table S9. Spearman's rank correlation coefficient (ρ) values obtained by comparing (A) archaea and bacteria, (B) archaea and archaeal classes, and (C) bacteria and bacterial classes. In archaea, α2, α5 and NM classes were not used for calculation as the number of sequences in those groups was below five. The analysis was performed using the data shown in Supplementary Figure S4.

(A)

	All	**Bacteria**	**Archaea**
All	1.00	0.98	0.77
Bacteria		1.00	0.79
Archaea			1.00

(B)

	Archaea	**α3**	**α4**	**MX**	**UN**
Archaea	1.00	0.54	0.29	-0.11	0.64
α3		1.00	0.86	0.75	-0.07
α4			1.00	0.69	-0.18
MX				1.00	-0.79
UN					1.00

(C)

	Bacteria	**α2**	**α3**	**α4**	**α5**	**MX**	**NM**	**UN**
Bacteria	1.00	0.66	0.96	0.86	1.00	0.16	0.93	0.89
α2		1.00	0.66	0.52	0.66	0.07	0.63	0.48
α3			1.00	0.79	0.96	0.05	0.89	0.93
α4				1.00	0.86	0.63	0.64	0.61
α5					1.00	0.16	0.93	0.89
MX						1.00	-0.23	-0.30
NM							1.00	0.93
UN								1.00

Supplementary Table S10. Analysis of sequence similarity networks (SSNs) of the ArsR-SmtB family proteins. In total, 62 amino acid sequences (obtained from Table 2) of the ArsR-SmtB family proteins were used to construct networks based on sequence similarities (SSNs). Nodes represent the ArsR-SmtB family protein sequences and the edge lengths represent the sequence similarities. Detailed sequence information with the group, motif, UniProt ID, gene or protein name, organism name, domain, phylum, node number (as shown in Figure 9) and the number of edges, depending on the alignment score stringency (10^{-5}, 10^{-10}, 10^{-15} and 10^{-20}), are given.

Group	No.	Motif	UniProt ID	Protein / Gene Name	Organism	Domain	Phylum	Node No.†	Edges‡			
									10^{-5}	10^{-10}	10^{-15}	10^{-20}
α2 group	1-3	α2-α53	Q8Z4F5	STY2917	*Salmonella typhi*	Bacteria	Proteobacteria	26	26	7	0	0
		α2-α5	P52695	HlyU	*Vibrio cholerae*	Bacteria	Proteobacteria					
		None	Q83TD2	NolR	*Rhizobium fredii*	Bacteria	Proteobacteria					
	4	α2-α52	D6CMI1	AioF	*Thiomonas arsenitoxydans*	Bacteria	Proteobacteria	38	14	1	0	0
α3 group	5	α3	Q5KUX7	ArsR1	*Geobacillus kaustophilus*	Bacteria	Firmicutes	1	37	27	3	0
	6	α3	C7NYS3	Hmuk_2504	*Halomicrobium mukohataei*	Archaea	Euryarchaeota	2	37	21	1	0
	7-9	α3	P15905	ArsR	*Escherichia coli*	Bacteria	Proteobacteria	17	30	12	2	0
		α55	D7H7R8	VCRC385_01870	*Vibrio cholerae*	Bacteria	Proteobacteria					
		α32	A0A0A7HMJ3	ArsR1	*Pantoea* sp.	Bacteria	Proteobacteria					
	10	α3	S0AS94	ArsR	*Ferroplasma acidarmanus*	Archaea	Euryarchaeota	25	27	10	1	0
	11-12	α32	Q46BU5	Mbar_A1705	*Methanosarcina barkeri*	Archaea	Euryarchaeota	12	32	19	3	0
		α3	D5E6K8	Mmah_1294	*Methanohalophilus mahii*	Archaea	Euryarchaeota					
	13	α3	P30338	ArsR	*Staphylococcus aureus*	Bacteria	Firmicutes	24	27	10	0	0
	14	α32	G7WP58	Mhar_1538	*Methanosaeta harundinacea*	Archaea	Euryarchaeota	8	33	14	0	0
	15	α32	A1XP68	ArsR1	*Ochrobactrum tritici*	Bacteria	Proteobacteria	32	21	6	1	0
	16	α32	A5JSW5	ArsRC	*Leptospirillum ferriphilum*	Bacteria	Nitrospirae	35	18	3	1	0
	17-18	α3N	P71941	Rv2642	*Mycobacterium tuberculosis*	Bacteria	Actinobacteria	3	35	24	3	0
		α3N	Q1HW04	ArsR2	*Streptomyces* sp.	Bacteria	Actinobacteria					
	19	α3N	P20047	CadC	*Staphylococcus aureus*	Bacteria	Firmicutes	20	30	13	3	0
	20	α3N	F0LM33	TERMP_00904	*Thermococcus barophilus*	Archaea	Euryarchaeota	19	30	8	0	0
	21	α3N	O52029	ArsR1	*Halobacterium salinarum*	Archaea	Euryarchaeota	30	22	7	1	1
	22	α3N	O52026	ArsR2	*Halobacterium salinarum*	Archaea	Euryarchaeota	34	19	8	1	1
	23	α3N2	Q8NTP5	ArsR2	*Corynebacterium glutamicum*	Bacteria	Actinobacteria	14	32	8	1	0
	24	α34	Q1HW01	ArsR1	*Streptomyces* sp.	Bacteria	Actinobacteria	15	31	8	0	0
	25	α34	D3PAR7	DEFDS_0179	*Deferribacter desulfuricans*	Bacteria	Deferribacteres	28	24	5	0	0
	26-27	α35	H8XU33	KQS_04180	*Flavobacterium indicum*	Bacteria	Bacteroidetes	36	18	2	1	0
		α55	A6L7W8	ArsR	*Bacteroides vulgatus*	Bacteria	Bacteroidetes					
	28	α32	A0A1B3X802	ArsR	*Campylobacter coli*	Bacteria	Proteobacteria	23	28	12	0	0
α4 group	29	α4c	Q4J865	Saci_1710	*Sulfolobus acidocaldarius*	Archaea	Crenarchaeota	4	35	18	1	0
	30-31	α4c	P9WMI9	CmtR	*Mycobacterium tuberculosis*	Bacteria	Actinobacteria	29	24	2	0	0
		α4c2	Q9RD34	CmtR	*Streptomyces coelicolor*	Bacteria	Actinobacteria					
α5 group	32	α5	O85142	CzrA	*Staphylococcus aureus*	Bacteria	Firmicutes	22	29	14	2	1
	33	α53	O53838	KmtR	*Mycobacterium tuberculosis*	Bacteria	Actinobacteria	21	30	15	2	0
	34	α54	Q8PZ32	MM_0662	*Methanosarcina mazei*	Archaea	Euryarchaeota	16	31	14	0	0

	35	α54	Q9L209	SCO6823	*Streptomyces coelicolor*	Bacteria	Actinobacteria	33	21	1	0	0
	36	α54	H0HHH0	ArsR1	*Agrobacterium tumefaciens*	Bacteria	Proteobacteria	42	5	1	0	0
	37	α55	A9A197	Nmar_0487	*Nitrosopumilus maritimus*	Archaea	Thaumarchaeota	27	25	15	1	0
	38	α56	B6WXY3	DESPIG_02964	*Desulfovibrio piger*	Bacteria	Proteobacteria	6	34	15	1	0
	39-40	α5N	O69711	NmtR	*Mycobacterium tuberculosis*	Bacteria	Actinobacteria	39	11	6	2	0
		α5c	D6TY78	Krac_4094	*Ktedonobacter racemifer*	Bacteria	Chloroflexi					
	41	α57	E6XPL1	ArsR	*Shewanella putrefaciens*	Bacteria	Proteobacteria	31	22	3	0	0
MX group	42-45	α3N	A6URK8	Mevan_1232	*Methanococcus vannielii*	Archaea	Euryarchaeota	13	32	22	9	3
		α3N-α5	D1YYC1	MCP_1371	*Methanocella paludicola*	Archaea	Euryarchaeota					
		α3-α5	G7V7Y6	Tlie_0487	*Thermovirga lienii*	Bacteria	Synergistetes					
		α3-α5	D3E3S3	mru_1334	*Methanobrevibacter ruminantium*	Archaea	Euryarchaeota					
	46	α3N-α5	I0RKP2	MXEN_15450	*Mycobacterium xenopi*	Bacteria	Actinobacteria	10	33	20	1	0
	47-51	α3N	Q8ZS91	AztR	*Nostoc* sp.	Bacteria	Cyanobacteria	18	30	19	4	3
		α5	P30340	SmtB	*Synechococcus elongatus*	Bacteria	Cyanobacteria					
		α3N-α5	Q76L30	BxmR	*Oscillatoria brevis*	Bacteria	Cyanobacteria					
		α3N-α5	Q55940	ZiaR	*Synechocystis* sp.	Bacteria	Cyanobacteria					
		α3N-α5	D9SQ59	Clocel_0346	*Clostridium cellulovorans*	Bacteria	Firmicutes					
	52-55	α5	Q0W2G2	RCIX2336	*Methanocella arvoryzae*	Archaea	Euryarchaeota	5	34	21	7	4
		α3-α5	G5GIP9	HMPREF9333_01439	*Johnsonella ignava*	Bacteria	Firmicutes					
		α3-α5	Q0W191	RRC76	*Methanocella arvoryzae*	Archaea	Euryarchaeota					
		α3-α5	C6D253	Pjdr2_3307	*Paenibacillus* sp.	Bacteria	Firmicutes					
	56	α3N-α5	H3ZMT0	OCC_05651	*Thermococcus litoralis*	Archaea	Euryarchaeota	11	33	22	4	3
NM group	57	α2-α5	G0HMP2	GQS_02890	*Thermococcus* sp.	Archaea	Euryarchaeota	37	15	1	0	0
	58	α33	A9KR37	Cphy_0116	*Lachnoclostridium phytofermentans*	Bacteria	Firmicutes	9	33	16	2	0
	59	α33	A4QI86	CyeR	*Corynebacterium glutamicum*	Bacteria	Actinobacteria	41	6	1	0	0
UN group	60	None_C	D1YW18	MCP_0568	*Methanocella paludicola*	Archaea	Euryarchaeota	7	34	14	0	0
	61	None_C	Q9HW47	PyeR	*Pseudomonas aeruginosa*	Bacteria	Proteobacteria	40	9	1	0	0
	62	None	O58788	PH1061	*Pyrococcus horikoshii*	Archaea	Euryarchaeota	43	1	0	0	0

†Node number (from Figure 9)

‡Number of edges connected to a particular node (alignment score stringency, from 10^{-5} to 10^{-20}, see Figure 9)