Supplementary Information (SI) for Environmental Science: Processes & Impacts.

## Supplementary data

**Text S1.** High-throughput sequencing primer sequences.

The V4 region of the bacterial 16S rRNA gene was subsequently amplified with sequencing primers 515F (5’-GTGCCAGCMGCCGCGGTAA-3’) and 806R (5’-GGACTACHVGGGTWTCTAAT-3’).

**Table S1.** Relationship between alpha diversity of microbial communities in three groups of reactors at different time period.

| **Alpha diversity** | | **Shannon** | **Inverse Simpson** | **Richness** | **Chao** |
|---|---|---|---|---|---|
| Original | GC-0 | 3.99±0.046 | 0.96±0.0026 | 905 | 1214.38 |
| Day 30 | GC-30 | 4.11±0.027 | 0.96±0.0011 | 879 | 1189.18 |
| | GL-30 | 4.08±0.042 | 0.96±0.00091 | 903 | 1251.01 |
| | GH-30 | 4.07±0.072 | 0.96±0.0017 | 907 | 1272.88 |
| Day 60 | GC-60 | 4.19±0.033 | 0.96±0.0006 | 891 | 1203.91 |
| | GL-60 | 4.17±0.039 | 0.97±0.0016 | 892 | 1208.79 |
| | GH-60 | 4.11±0.022 | 0.96±0.00111 | 841 | 1169.13 |
| Day 90 | GC-90 | 4.07±0.020 | 0.94±0.0057 | 944 | 1267.40 |
| | GL-90 | 3.79±0.15 | 0.93±0.016 | 821 | 1052.46 |
| | GH-90 | 4.19±0.022 | 0.96±0.0014 | 898 | 1128.66 |
| Day 120 | GC-120 | 3.38±0.083 | 0.84±0.016 | 873 | 1088.91 |
| | GL-120 | 3.29±0.079 | 0.82±0.017 | 876 | 1114.04 |
| | GH-120 | 4.14±0.085 | 0.96±0.0048 | 914 | 1167.73 |

**Table S2.** Dissimilarity results of microbial communities between different groups based on the Bray-Curtis matrix

| | **Groups** | **Day0** | **GC30-60** | **GL30-60** | **GH30-60** | **GC90-120** | **GL90-120** | **GH90-120** |
|---|---|---|---|---|---|---|---|---|
| MRPP | **Day0** | 0 | 0.016 | 0.012 | 0.016 | 0.012 | 0.011 | 0.012 |
| | **GC30-60** | 0.2147 | 0 | 0.002 | 0.012 | 0.002 | 0.003 | 0.002 |
| | **GL30-60** | 0.2591 | 0.2228 | 0 | 0.001 | 0.002 | 0.003 | 0.004 |
| | **GH30-60** | 0.2511 | 0.2168 | 0.2501 | 0 | 0.005 | 0.004 | 0.004 |
| | **GC90-120** | 0.2760 | 0.2355 | 0.2688 | 0.2627 | 0 | 0.043 | 0.004 |
| | **GL90-120** | 0.2829 | 0.2407 | 0.2740 | 0.2679 | 0.2866 | 0 | 0.004 |
| | **GH90-120** | 0.2684 | 0.2298 | 0.2631 | 0.2570 | 0.2757 | 0.2809 | 0 |
| ANOSIM | **Day0** | 0 | 0.009 | 0.017 | 0.014 | 0.011 | 0.009 | 0.01 |
| | **GC30-60** | 1 | 0 | 0.001 | 0.009 | 0.004 | 0.005 | 0.003 |
| | **GL30-60** | 0.9938 | 0.7370 | 0 | 0.004 | 0.002 | 0.004 | 0.003 |
| | **GH30-60** | 1 | 0.3888 | 0.6592 | 0 | 0.007 | 0.002 | 0.003 |
| | **GC90-120** | 1 | 0.9981 | 0.9870 | 1 | 0 | 0.065 | 0.002 |
| | **GL90-120** | 1 | 0.9925 | 0.8074 | 0.9944 | 0.2888 | 0 | 0.004 |
| | **GH90-120** | 1 | 1 | 1 | 0.9425 | 0.9944 | 0.9925 | 0 |
| PERMANOVA | **Day0** | 0 | 0.013 | 0.017 | 0.006 | 0.008 | 0.016 | 0.018 |
| | **GC30-60** | 11.7138 | 0 | 0.004 | 0.017 | 0.005 | 0.002 | 0.003 |
| | **GL30-60** | 10.4375 | 7.3806 | 0 | 0.003 | 0.006 | 0.004 | 0.002 |
| | **GH30-60** | 10.2239 | 3.6861 | 5.7288 | 0 | 0.004 | 0.001 | 0.003 |
| | **GC90-120** | 20.4698 | 27.5258 | 18.9836 | 23.7787 | 0 | 0.05 | 0.002 |
| | **GL90-120** | 19.2084 | 26.2625 | 15.2660 | 21.5032 | 3.4052 | 0 | 0.003 |
| | **GH90-120** | 23.8480 | 30.7686 | 22.7044 | 20.4259 | 14.6709 | 12.8921 | 0 |

Note: The values of the upper triangular matrix are P values. The values of the lower triangular matrix are delta, R, and f values for MRPP, ANOSIM, and PERMANOVA, respectively.

**Table S3.** Topological properties of the empirical functional molecular ecological networks (fMENs) in comparison to the random networks.

| **Empirical networks** | | | | | | | | | **Random networks**[a] | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Group | Total nodes | Edges | $R^2$ of power law | Similarity threshold ($s_t$) | Avg connectivity (avgK)[b] | Avg path length (GD)[c] | Avg clustering coefficient (avgCC) | Modularity (No. of modules)[d] | Avg path length (GD) | Avg clustering coefficient (avgCC) | Modularity (M) |
| GC | 203 | 922 | 0.96 | 0.94 | 9.08 | 4.21 | 0.35 | 0.39 | 2.76±0.04 | 0.09±0.012 | 0.24±0.006 |
| GL | 153 | 948 | 0.94 | 0.94 | 12.39 | 4.44 | 0.41 | 0.22 | 2.53±0.04 | 0.14±0.013 | 0.16±0.005 |
| GH | 187 | 1006 | 0.96 | 0.94 | 10.76 | 3.05 | 0.37 | 0.30 | 2.69±0.04 | 0.12±0.012 | 0.19±0.004 |

a. Random networks were generated by rewiring all nodes and links corresponding to empirical networks 100 times.

b. Significant difference ($P<0.001$) in average connectivity between any two groups based on Student's t test.

c. Significant difference ($P<0.001$) in average path for Group A with other two groups based on Student's t test.

d. Significant difference ($P<0.001$) in modularity between any two groups based on Student's t test.

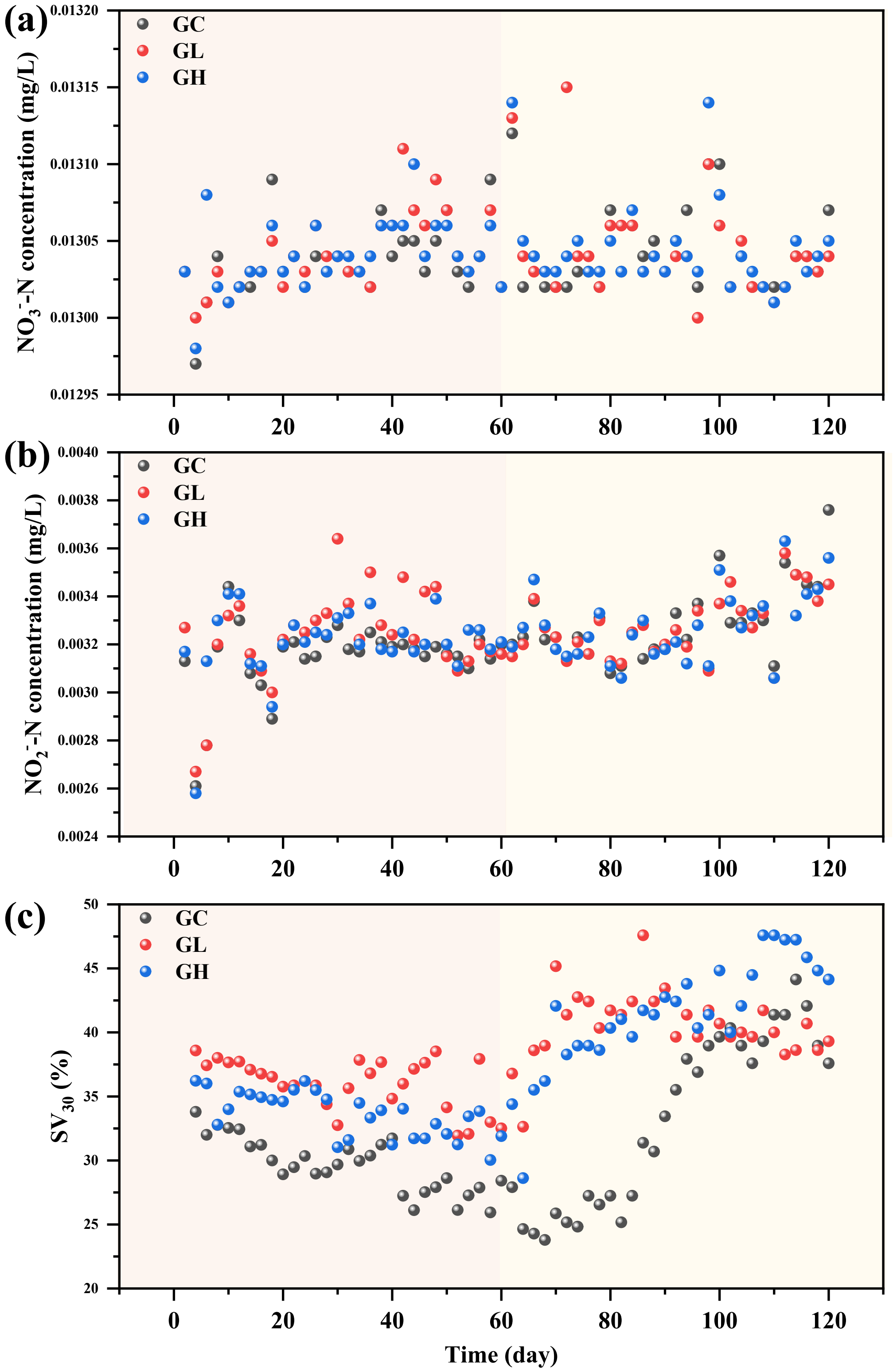

**Fig. S1** The concentrations of $NO_3$-N (a) and $NO_2$-N (b), and $SV_{30}$ values in the three groups

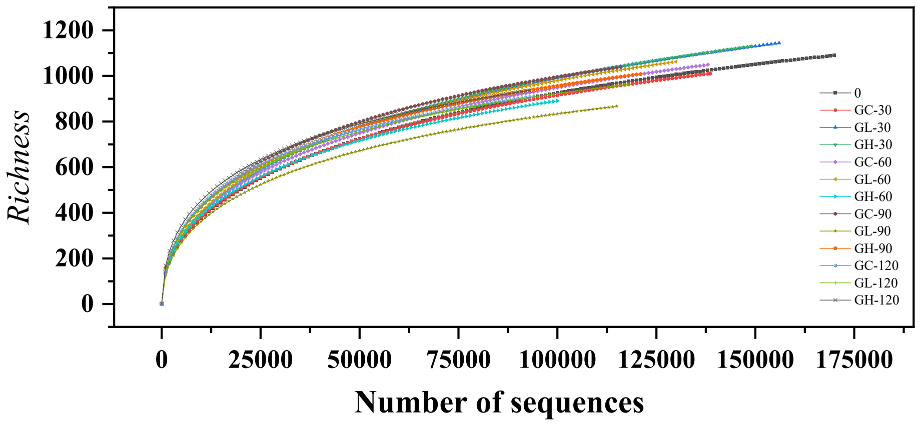

**Fig. S2** Rarefaction curves were obtained from all samples N in the three groups

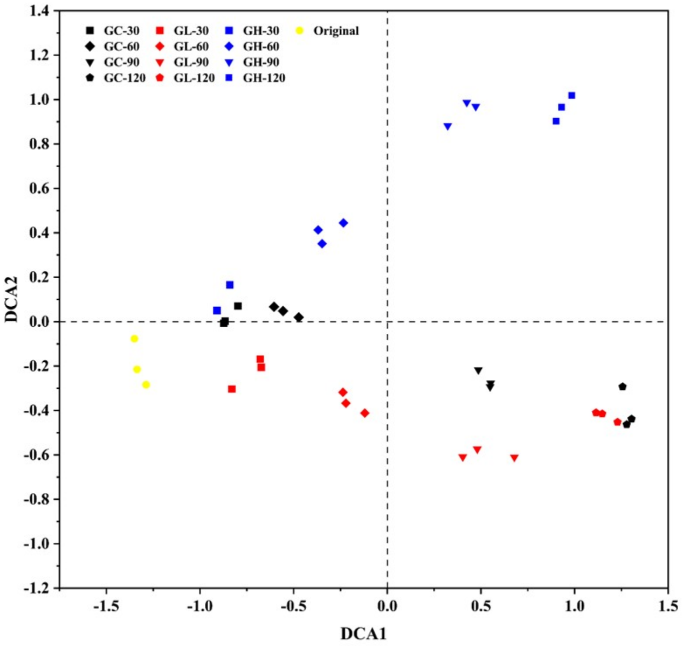

**Fig. S3** Detrended correspondence analysis of the samples at the phyla level

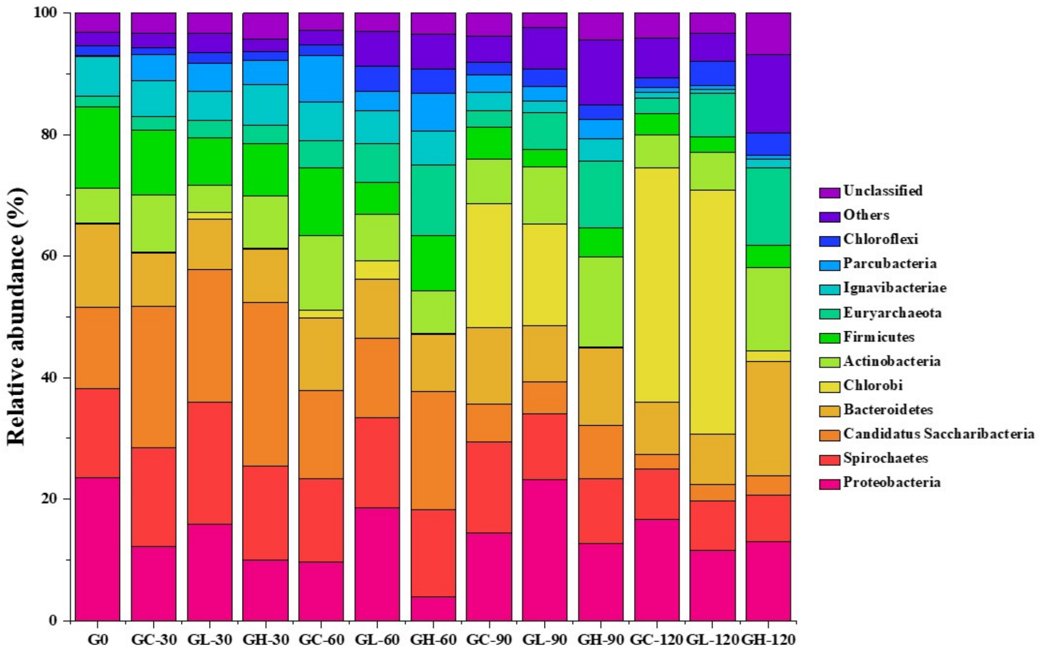

**Fig. S4** Dynamic changes of the bacterial communities at the phylum level during the whole operation process. Major means the relative abundance was >1 % in each sample

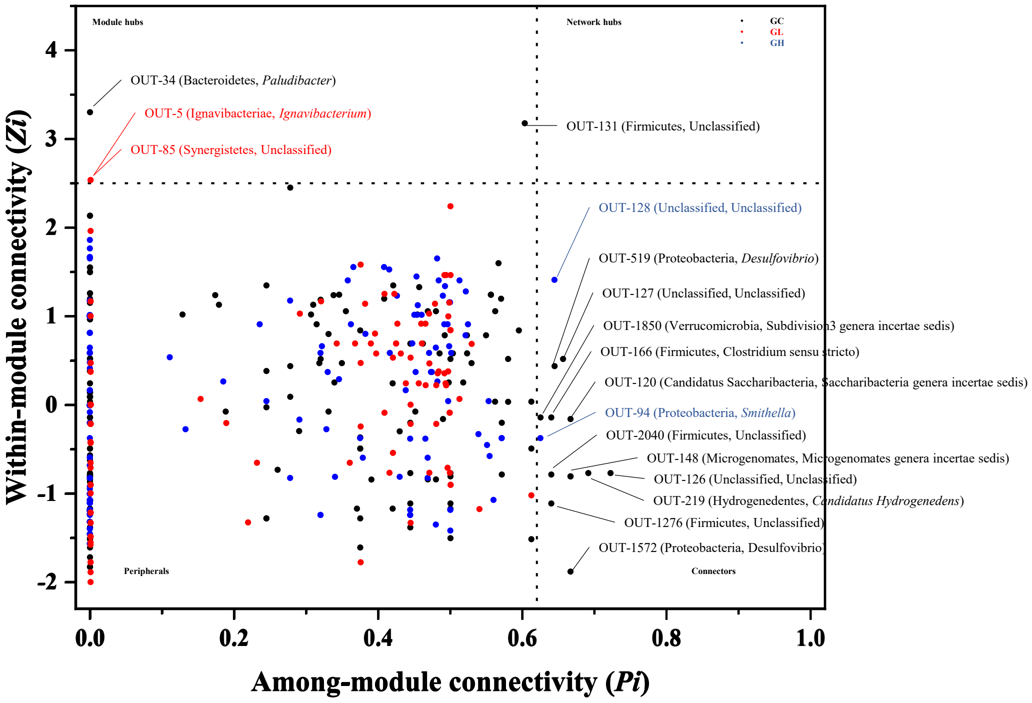

**Fig. S5** Representation of nodes of keystone species in the three networks. Each triangle represents an OTU for detailed module analysis

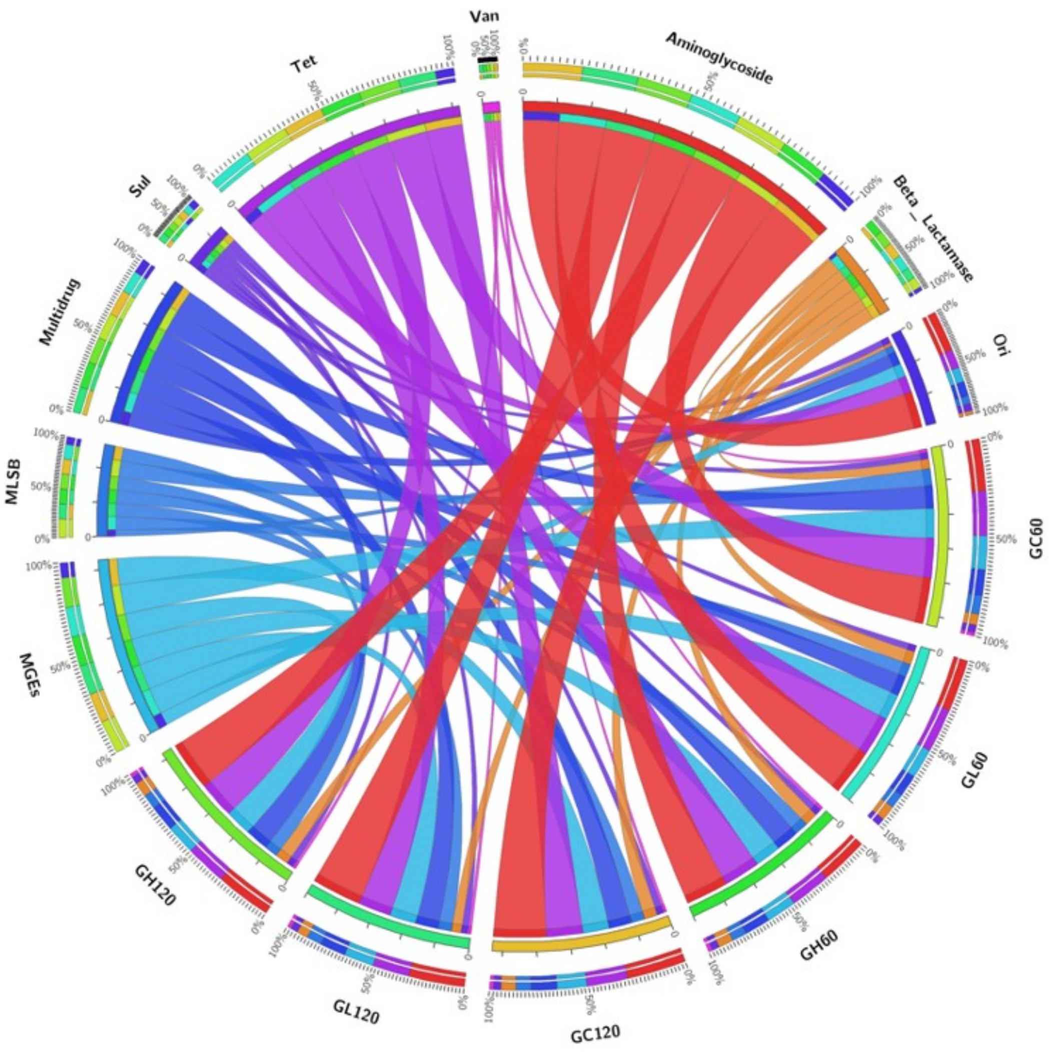

**Fig. S6** The number of typical ARGs on Day 60 and 120

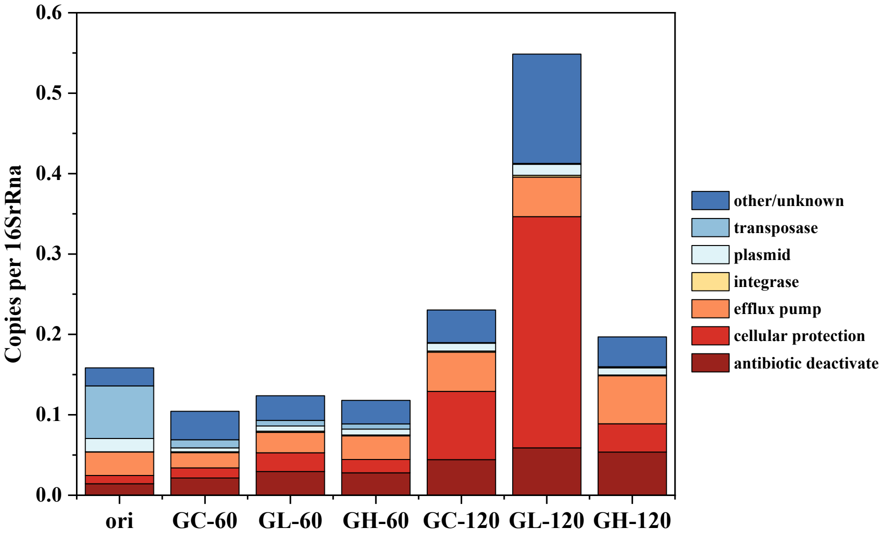

**Fig. S7** The effects of graphene on the distributions of ARGs belonging to different resistance mechanism.