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## Supplementary Information

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### 3      **Response of chlorinated hydrocarbons transformation and 4      microbial community structure in the aquifer to joint H<sub>2</sub> and O<sub>2</sub>**

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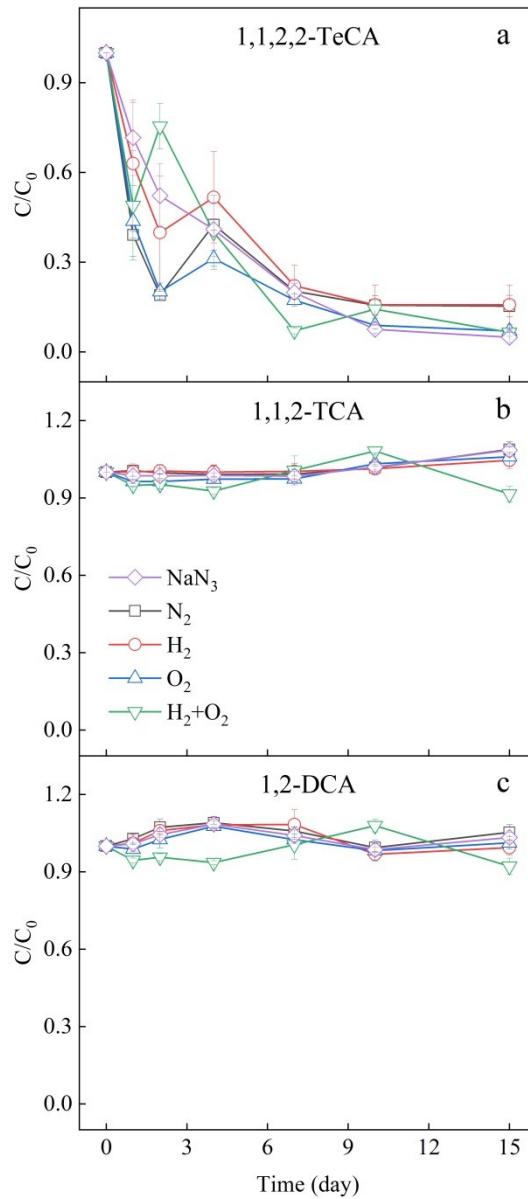
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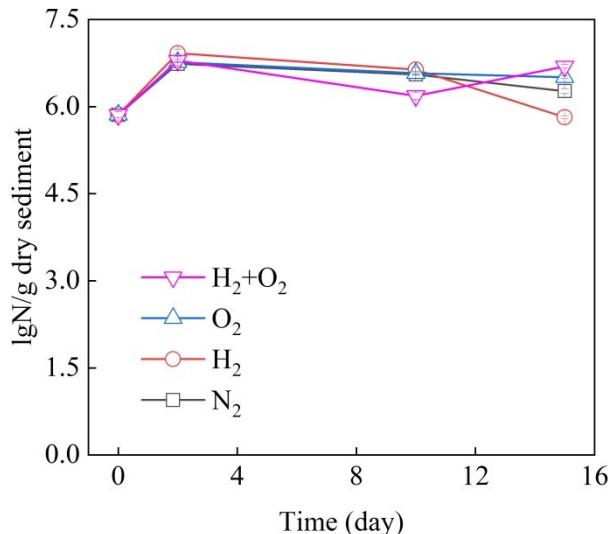


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14 **Fig. S1 Concentrations of chlorinated ethanes (1,1,2,2-TeCA (a), 1,1,2-TCA (b), 1,2-DCA**

15 **(c)) with time in different microcosms of  $\text{H}_2$  and  $\text{O}_2$  microcosms and with  $\text{NaN}_3$  as abiotic**

16 **control**

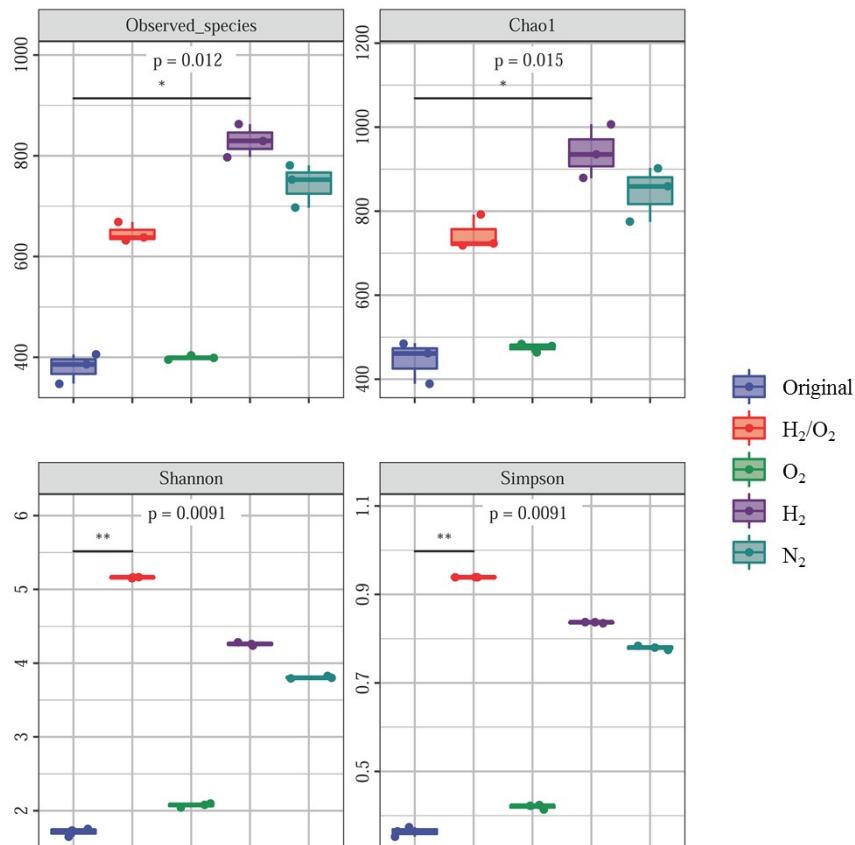


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**Fig. S2 Microbial numbers in different times of incubation in the four microcosms**

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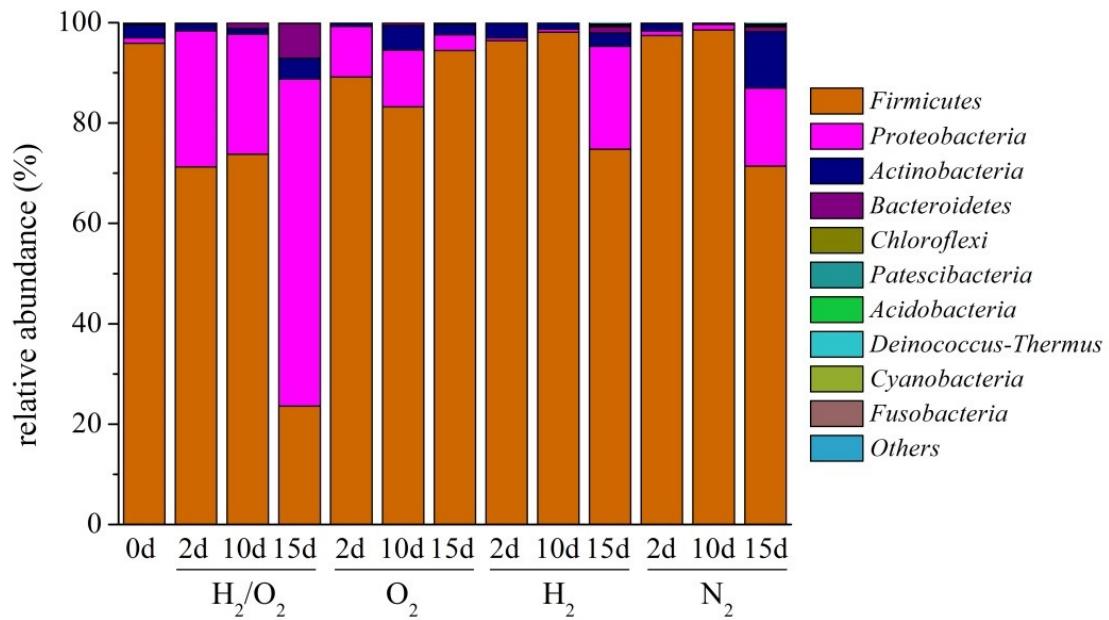
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**Fig. S3 Microbial diversity indices of the sediments before and after 15-days of incubation in**

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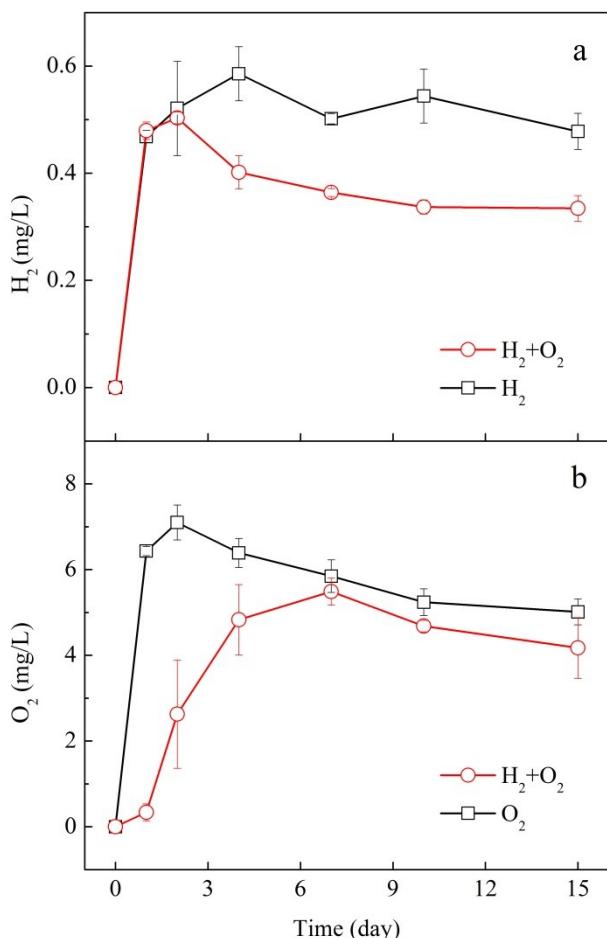
**the four microcosms**



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24 **Fig. S4 Microbial composition at phylum level in different experimental microcosms**

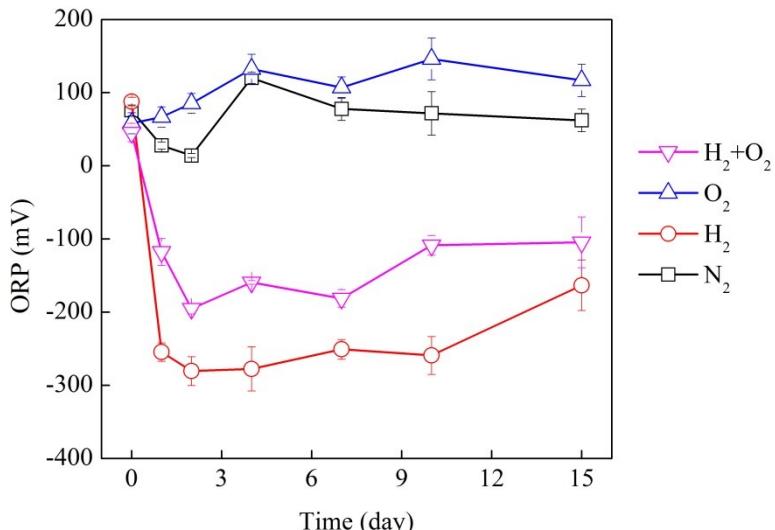
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27 **Fig. S5 Variations of  $\text{H}_2$  (a) and  $\text{O}_2$  (b) concentrations during chlorinated hydrocarbons  
28 degradation**

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31 **Fig. S6 Variations of oxidation-reduction potential (ORP) during chlorinated hydrocarbons  
32 degradation**

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**Table S1 Sediments properties used in this study**

<b>Contaminant</b>	<b>Concentration</b>	<b>Concentration</b>		<b>Concentration</b>	
	(mg/kg)	Cation	(mg/g)	Anion	(mg/kg)
Perchloroethylene	2.78±0.22	Fe	13.14	Cl <sup>-</sup>	189.44
Trichloroethylene	171.41±0.84	K	2.37	SO <sub>4</sub> <sup>2-</sup>	66.05
<i>cis</i> -Dichloroethene	88.51±14.42	Na	0.62	NO <sub>3</sub> <sup>-</sup>	34.64
<i>trans</i> -Dichloroethene	0.78±0.07	Ca	20.79	PO <sub>4</sub> <sup>3-</sup>	20.12
Vinyl chloride	12.1±1.75	Mg	7.39	F <sup>-</sup>	24.31
1,1,2-Trichloroethane	2.75±0.32	P	0.32		
1,2-Dichloroethane	4.46±0.73	Al	9.81		
1,1-Dichloroethane	1.38±0.19	Mn	0.32		
Chloroform	0.29±0.04	Si	33.57		
Chlorobromomethane	0.28±0.03	Li	2.37		
		Ti	0.38		

35 **Note:** TOC content of sediments was 1360 mg/kg, pH value was 7.4.

**Table S2 Material balance and variance analysis of CHCs in microcosms**

Pollutants	Conditions	Concentration ( $\mu\text{M}$ )			Transformation rate (%)	P-value	
		0 day	15 day	variation			
Perchloroethylene	$\text{N}_2+\text{NaN}_3$	47.36 $\pm$ 1.43	30.62 $\pm$ 0.14	-16.74	-35.35	3.53E-05	**
	$\text{N}_2$	43.86 $\pm$ 4.56	29.44 $\pm$ 0.68	-14.42	-32.88	5.63E-03	**
	$\text{H}_2$	52.38 $\pm$ 3.64	29.89 $\pm$ 0.89	-22.49	-42.94	4.82E-04	**
	$\text{O}_2$	41.10 $\pm$ 2.92	21.84 $\pm$ 0.72	-19.26	-46.86	3.76E-04	**
	$\text{H}_2+\text{O}_2$	30.15 $\pm$ 0.41	20.95 $\pm$ 1.33	-9.2	-30.51	3.35E-04	**
Trichloroethylene	$\text{N}_2+\text{NaN}_3$	27.14 $\pm$ 0.65	48.46 $\pm$ 0.48	21.32	78.56	7.26E-04	**
	$\text{N}_2$	40.56 $\pm$ 3.15	35.99 $\pm$ 4.2	-4.57	-11.27	0.21	
	$\text{H}_2$	43.31 $\pm$ 2.48	36.22 $\pm$ 3.32	-7.09	-16.37	4.14E-02	*
	$\text{O}_2$	40.10 $\pm$ 3.42	40.54 $\pm$ 4.56	0.44	1.10	0.90	
	$\text{H}_2+\text{O}_2$	40.56 $\pm$ 3.15	23.49 $\pm$ 2.34	-17.07	-42.09	1.66E-03	**
<i>trans</i> -Dichloroethene	$\text{N}_2+\text{NaN}_3$	31.45 $\pm$ 0.81	31.23 $\pm$ 0.36	-0.22	-0.70	0.70	
	$\text{N}_2$	29.67 $\pm$ 1.75	30.71 $\pm$ 0.56	1.04	3.51	0.38	
	$\text{H}_2$	32.80 $\pm$ 1.56	30.60 $\pm$ 0.93	-2.2	-6.71	0.10	
	$\text{O}_2$	30.21 $\pm$ 1.45	26.50 $\pm$ 0.16	-3.71	-12.28	1.14E-02	*
	$\text{H}_2+\text{O}_2$	26.79 $\pm$ 0.47	12.20 $\pm$ 0.55	-14.59	-54.46	4.01E-06	**
Carbon tetrachloride	$\text{N}_2+\text{NaN}_3$	31.92 $\pm$ 1.09	24.22 $\pm$ 2.11	-7.7	-24.12	4.94E-03	**
	$\text{N}_2$	28.56 $\pm$ 2.18	15.46 $\pm$ 0.65	-13.1	-45.87	5.68E-04	**
	$\text{H}_2$	33.55 $\pm$ 2.63	9.64 $\pm$ 0.48	-23.91	-71.27	1.02E-04	**
	$\text{O}_2$	27.39 $\pm$ 1.52	21.97 $\pm$ 0.73	-5.42	-19.79	5.12E-03	**
	$\text{H}_2+\text{O}_2$	34.15 $\pm$ 0.84	25.87 $\pm$ 1.11	-8.28	-24.25	4.91E-04	**
Chloroform	$\text{N}_2+\text{NaN}_3$	33.80 $\pm$ 0.82	34.68 $\pm$ 0.53	0.88	2.60	0.20	
	$\text{N}_2$	33.28 $\pm$ 1.10	37.70 $\pm$ 0.42	4.42	13.28	2.90E-03	**
	$\text{H}_2$	32.55 $\pm$ 0.70	40.82 $\pm$ 1.25	8.27	25.41	5.57E-04	**
	$\text{O}_2$	31.68 $\pm$ 1.16	31.86 $\pm$ 0.29	0.18	0.57	0.81	
	$\text{H}_2+\text{O}_2$	35.77 $\pm$ 0.94	28.49 $\pm$ 0.97	-7.28	-20.35	7.39E-04	**
Dichloromethane	$\text{N}_2+\text{NaN}_3$	28.33 $\pm$ 1.72	29.81 $\pm$ 0.69	1.48	5.22	0.24	
	$\text{N}_2$	28.02 $\pm$ 0.95	31.71 $\pm$ 0.74	3.69	13.17	6.06E-03	**
	$\text{H}_2$	29.18 $\pm$ 0.90	30.47 $\pm$ 1.14	1.29	4.42	0.20	
	$\text{O}_2$	29.86 $\pm$ 1.16	30.19 $\pm$ 0.61	0.33	1.11	0.69	
	$\text{H}_2+\text{O}_2$	24.80 $\pm$ 0.40	22.27 $\pm$ 0.92	-2.53	-10.20	1.21E-02	*
1,1,2,2-Tetrachloroethane	$\text{N}_2+\text{NaN}_3$	28.81 $\pm$ 3.90	1.38 $\pm$ 0.35	-27.43	-95.21	2.66E-04	**
	$\text{N}_2$	30.67 $\pm$ 1.14	4.70 $\pm$ 1.14	-25.97	-84.68	9.77E-06	**
	$\text{H}_2$	26.86 $\pm$ 2.89	4.28 $\pm$ 2.08	-22.58	-84.07	3.90E-04	**
	$\text{O}_2$	26.39 $\pm$ 2.72	1.83 $\pm$ 0.16	-24.56	-93.07	9.85E-05	**
	$\text{H}_2+\text{O}_2$	24.56 $\pm$ 4.23	1.61 $\pm$ 0.36	-22.95	-93.44	7.25E-04	**
1,1,2-Trichloroethane	$\text{N}_2+\text{NaN}_3$	29.10 $\pm$ 0.44	31.58 $\pm$ 0.08	2.48	8.52	6.40E-04	**
	$\text{N}_2$	28.98 $\pm$ 0.60	31.52 $\pm$ 0.27	2.54	8.76	2.64E-03	**
	$\text{H}_2$	28.90 $\pm$ 1.04	30.22 $\pm$ 1.27	1.32	4.57	0.24	
	$\text{O}_2$	28.88 $\pm$ 1.17	30.57 $\pm$ 0.30	1.69	5.85	0.07	
	$\text{H}_2+\text{O}_2$	24.69 $\pm$ 0.63	22.57 $\pm$ 0.35	-2.12	-8.59	7.18E-03	**
1,2-Dichloroethane	$\text{N}_2+\text{NaN}_3$	28.99 $\pm$ 0.54	29.96 $\pm$ 0.10	0.97	3.35	3.83E-02	*
	$\text{N}_2$	29.23 $\pm$ 0.47	30.77 $\pm$ 0.50	1.54	5.27	1.78E-02	*
	$\text{H}_2$	30.03 $\pm$ 0.94	29.84 $\pm$ 1.28	-0.19	-0.63	0.85	
	$\text{O}_2$	29.88 $\pm$ 0.93	30.23 $\pm$ 0.21	0.35	1.17	0.56	
	$\text{H}_2+\text{O}_2$	25.52 $\pm$ 0.36	23.51 $\pm$ 0.66	-2.01	-7.88	9.69E-03	**

37 \* Significance at  $P < 0.05$ , \*\* Significance at  $P < 0.01$