

Supplementary Information

Aquatic photo-transformation and enhanced photoinduced toxicity of ionizable tetracycline antibiotics

Linke Ge^{a,b}, Jinshuai Zheng^a, Crispin Halsall^b, Chang-Er Chen^c, Xuanyan Li^a, Shengkai Cao^b, Peng Zhang^{a,*}

^a School of Environmental Science and Engineering, Shaanxi University of Science & Technology, Xi'an 710021, China

^b Lancaster Environment Centre, Lancaster University, Lancaster LA1 4YQ, UK

^c Environmental Research Institute/School of Environment, South China Normal University, Guangzhou 510006, China

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* Corresponding author.

E-mail addresses: zhangpeng4477@sust.edu.cn (P. Zhang)

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Methods for plotting the dissociation forms distribution

The dissociation-form distribution of model substances (S) was plotted based on the relationship between pH and distribution fraction (δ). δ was calculated from $[H^+]$ or pH, and the dissociation constants (k_{a1} , k_{a2}). S are regarded as dibasic acids and exist in three forms (*i*): H_2S^+ , HS^0 , and S^- . The distribution fractions (δ_i) of the three dissociated forms at different pHs can be calculated by the following equations for $pH \sim \delta_i$,

$$\delta_{H_2S^+} = \frac{[H_2S^+]}{[H_2S^+] + [HS^0] + [S^-]} = \frac{[H^+]^2}{[H^+]^2 + [H^+]k_{a1} + k_{a1}k_{a2}} \quad (1)$$

$$\delta_{HS^0} = \frac{[HS^0]}{[H_2S^+] + [HS^0] + [S^-]} = \frac{[H^+]k_{a1}}{[H^+]^2 + [H^+]k_{a1} + k_{a1}k_{a2}} \quad (2)$$

$$\delta_{S^-} = \frac{[S^-]}{[H_2S^+] + [HS^0] + [S^-]} = \frac{k_{a1}k_{a2}}{[H^+]^2 + [H^+]k_{a1} + k_{a1}k_{a2}} \quad (3)$$

Thus, the dissociation-form distribution of S can be plotted.

Detailed analytical procedures

To investigate photo-transformation intermediates of tetracycline (TC), oxytetracycline (OTC), and chlortetracycline (CTC), their irradiated samples were analyzed by an Agilent 6470B HPLC-MS/MS. The mobile phases were pure water (containing 0.1% formic acid) (A) and acetonitrile (B). The flow rate was 0.3 mL/min, with injection volume 5 μ L. The mass spectrometry conditions were as follows. Electrospray ionization (ESI): positive mode; drying gas flow: 5 L/min, drying gas temperature: 300 $^{\circ}$ C; nebulizer pressure: 35 psi; capillary voltage: 3.5 kV; Sheath temperature: 250 $^{\circ}$ C, flow rate: 11 L/min.

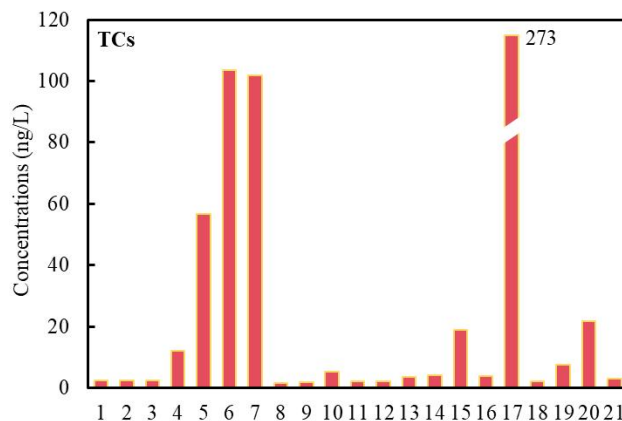


Fig. A1 The distribution of tetracyclines antibiotics (TCs) in surface waters of China (ng/L; 1. Daliao River, Liaoning (Yang et al., 2012); 2. Yellow Sea coast in Liaoning (Na et al., 2011); 3. coastal seawater of Dalian (Na et al., 2013); 4. Haihe River, Tianjin (Luo et al., 2011); 5. Haihe River estuary (Zou et al., 2011); 6. Wei River, Xi'an (Wang et al., 2019); 7. Ba River, Xi'an (Jia et al., 2018); 8. Nansi Lake, Shandong (Zhang et al., 2020a); 9. Surface waters, Jiangsu (Wei et al., 2011); 10. Tai Lake, Jiangsu (Zhou et al., 2016); 11. Huangpu River, Shanghai (Jiang et al., 2011; Chen and Zhou, 2014); 12. Lower reaches of the Yangtze River (Zhang et al., 2020b); 13. Han River (Hu et al., 2018); 14. Chongqing section of the Yangtze River (Wang et al., 2020); 15. Three Gorges Reservoir (Yan et al., 2018); 16. East China Sea (Li et al., 2020); 17. Honghu Lake (Wang et al., 2017); 18. Fuxian Lake (Zhao et al., 2021); 19. Pearl River estuary (Liang et al., 2013); 20. Coastal waters, Hong Kong (Gulkowska et al., 2007); 21. Bosten Lake, Xinjiang (Lei et al., 2015).

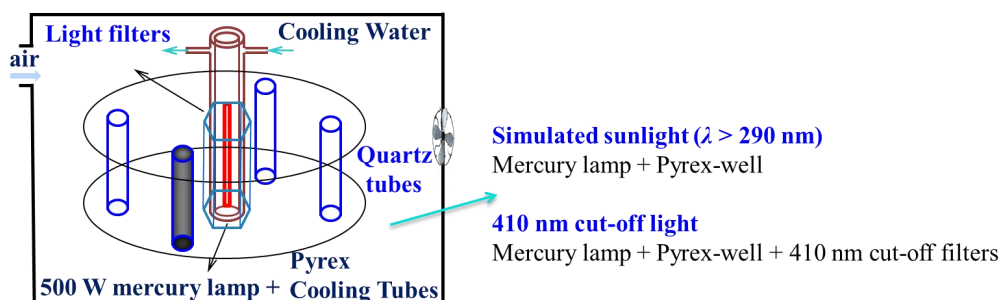


Fig. A2 Schematic diagram of the photochemical experimental apparatus.

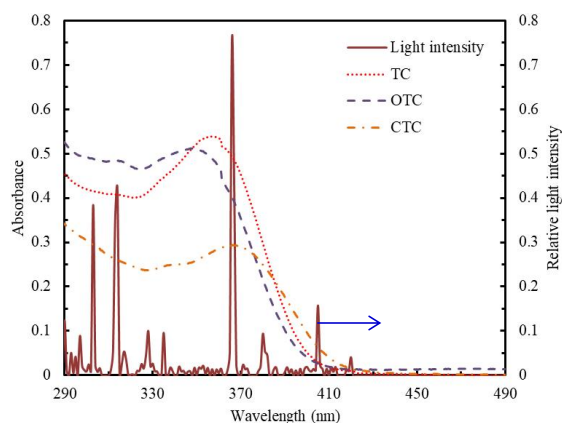


Fig. A3 UV absorption spectra of tetracycline (TC), oxytetracycline (OTC), and chlortetracycline (CTC) and the relative light intensity of a 500 W high-pressure mercury lamp with a water-cooled Pyrex well ($\lambda > 290$ nm).

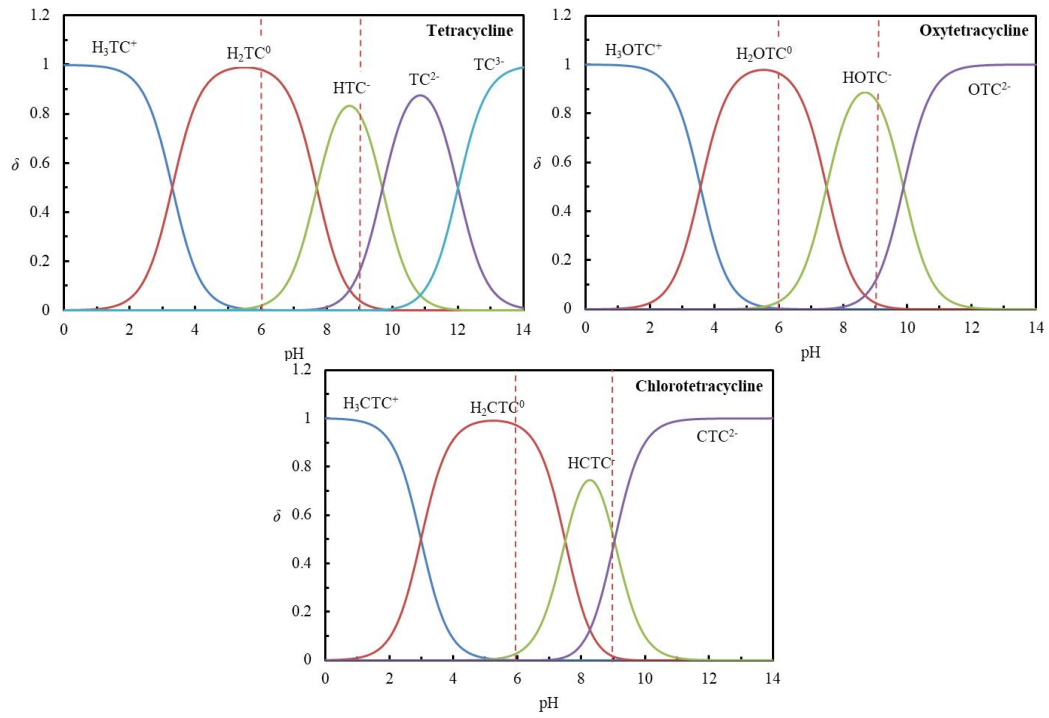


Fig. A4 The distribution of dissociated species of tetracycline antibiotics under different pH conditions.

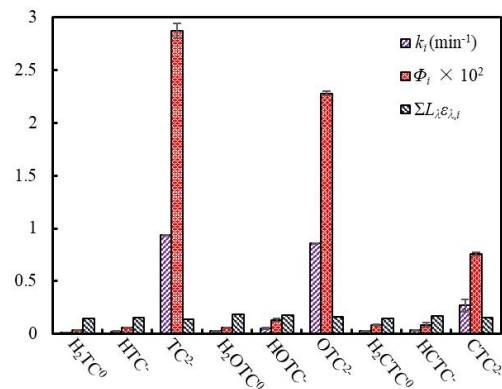


Fig. A5 The photolytic rate constants (k_i), quantum yields (Φ_i) and cumulative light absorption ($\Sigma L_j \epsilon_{\lambda,i}$) for each dissociated species (i) of tetracycline antibiotics (TCs).

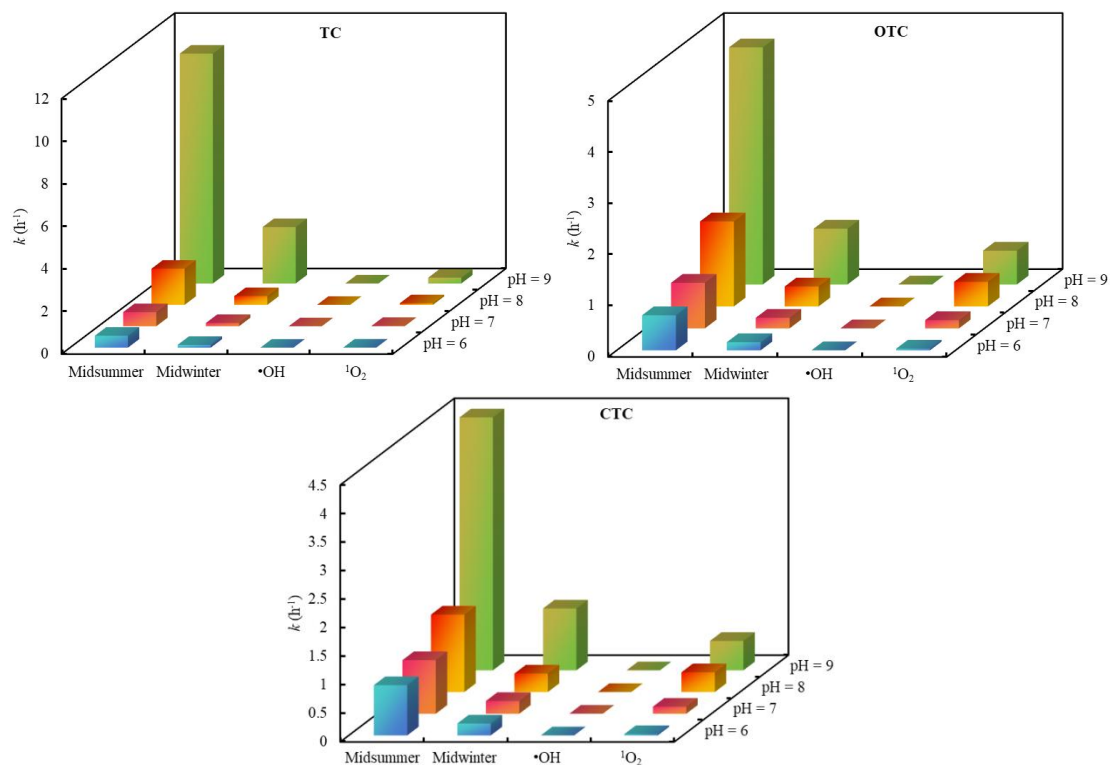


Fig. A6 Rate constants (k) of apparent photolysis (in midsummer and midwinter) and photooxidation degradation ($\bullet\text{OH}$ and $^1\text{O}_2$) of tetracycline antibiotics (TCs) in surface waters with different pH at 45°N latitude.

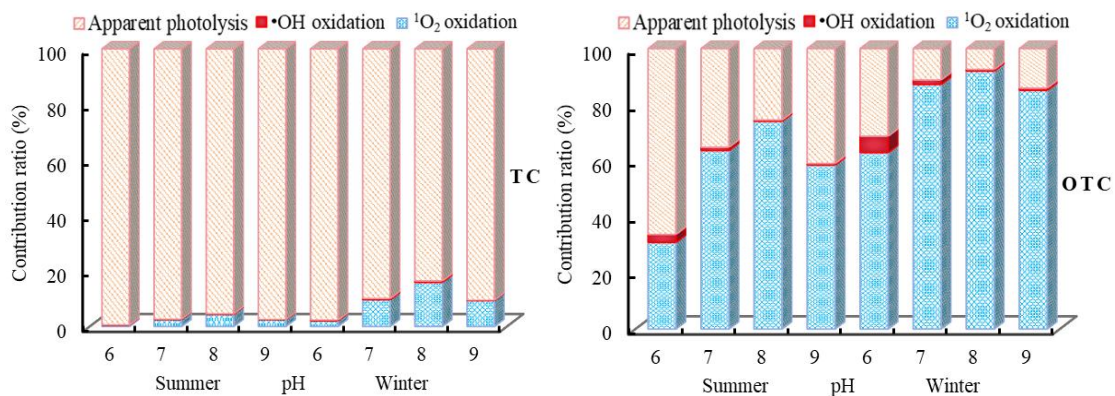


Fig. A7 Relative contributions of apparent photolysis and reactions with $\bullet\text{OH}$ and $^1\text{O}_2$ to the photo-transformation of tetracycline antibiotics (TCs) in surface waters at 45°N latitude.

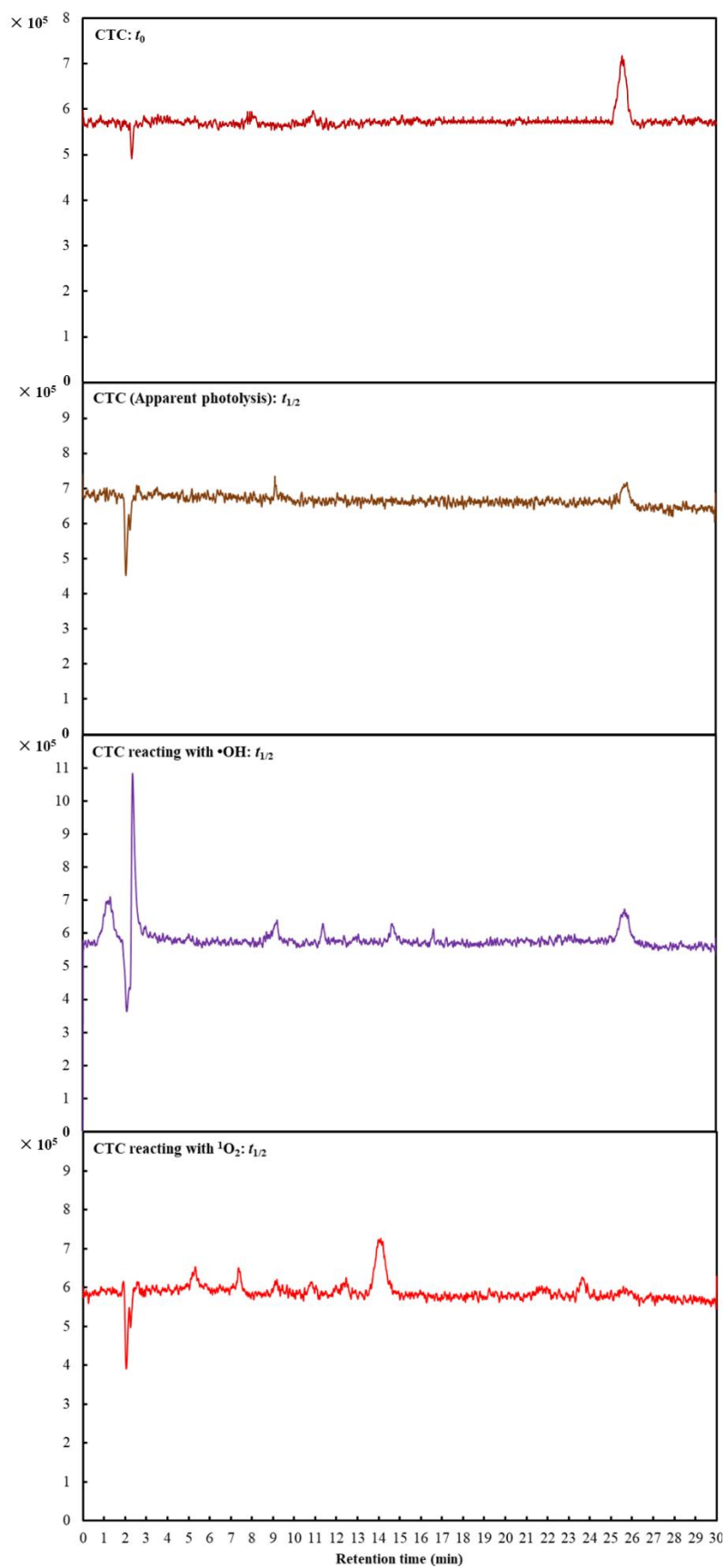
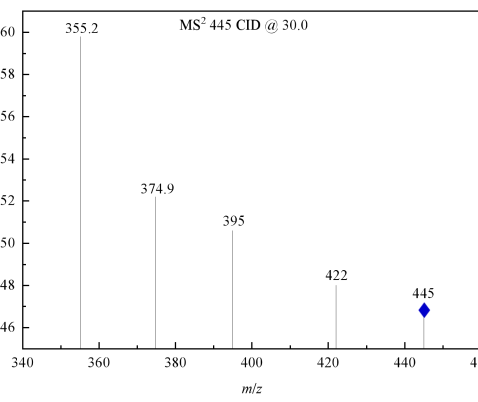
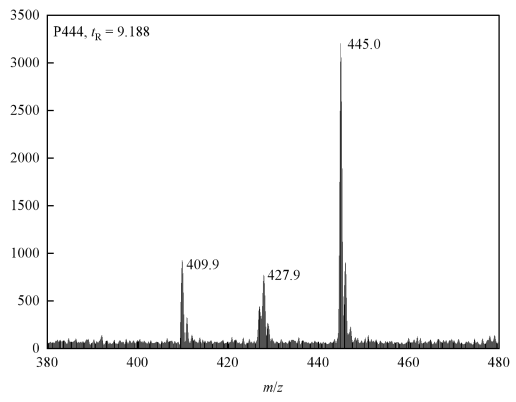
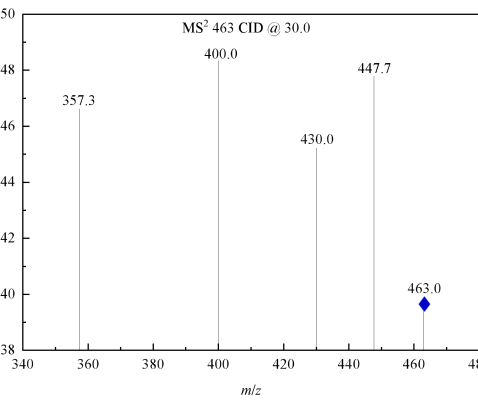
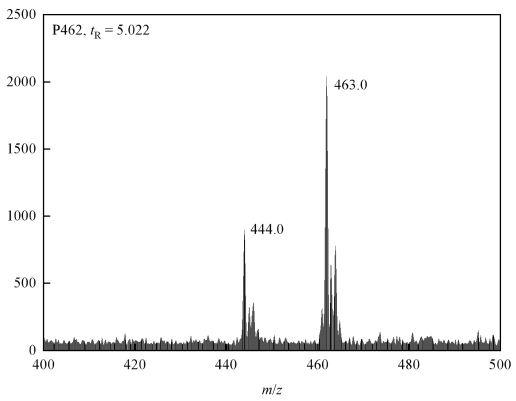
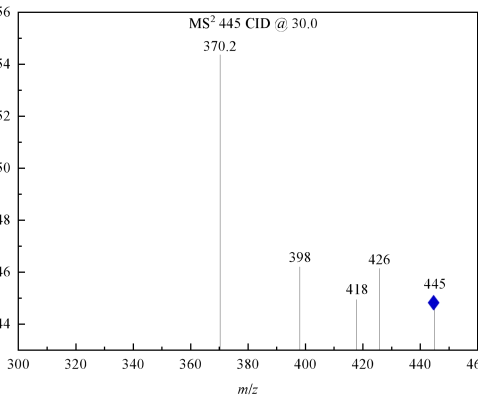
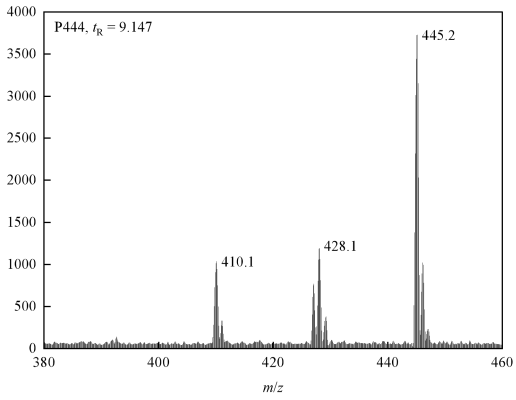
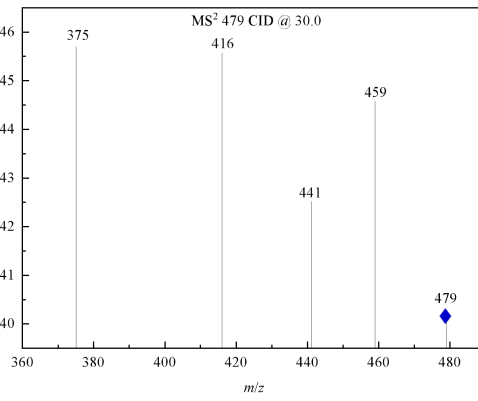
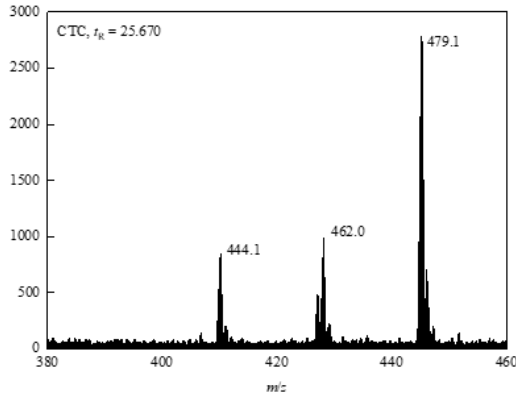
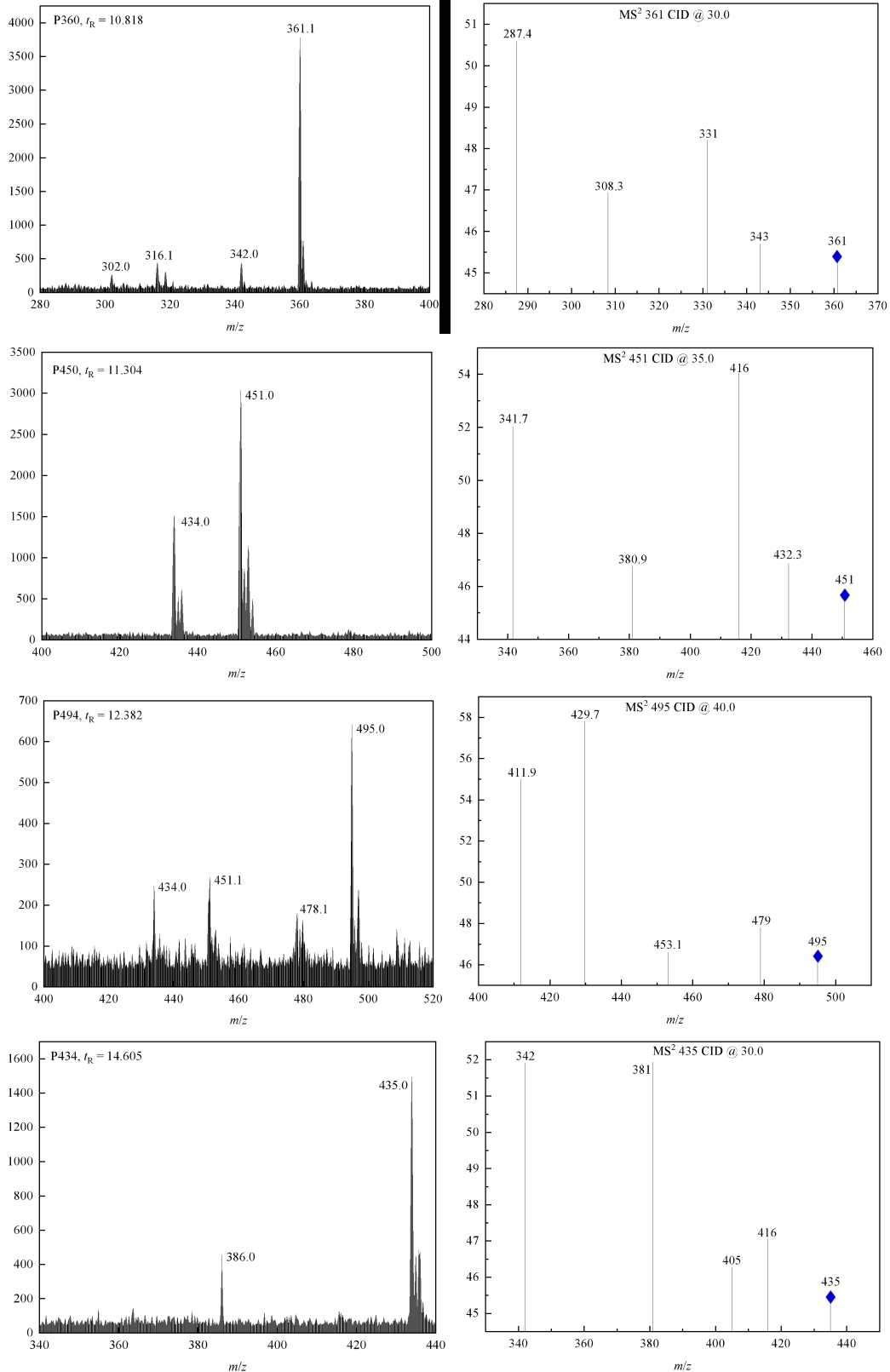
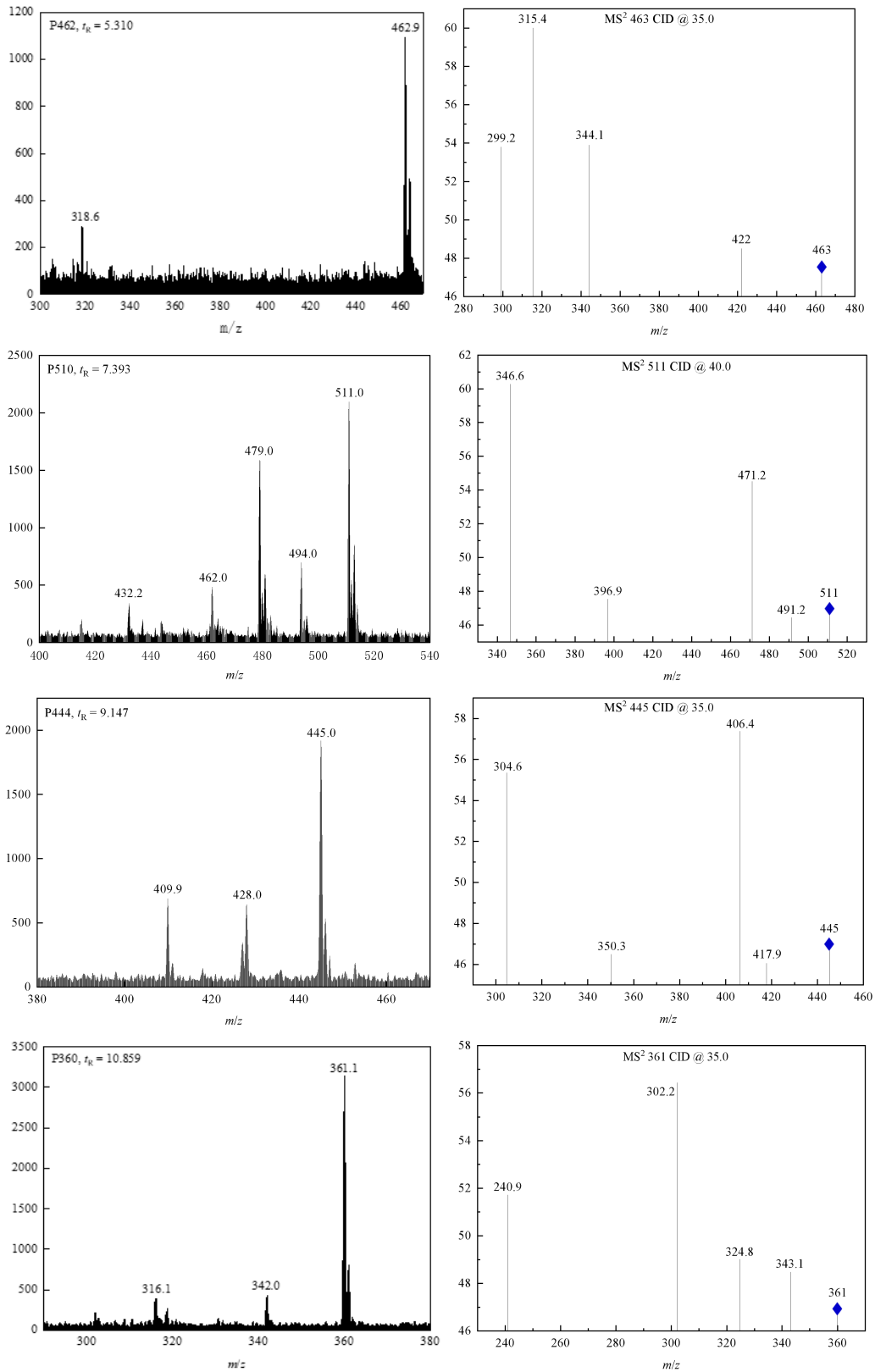


Fig. A8 Total ion chromatograms for irradiated samples of chlortetracycline (CTC) in different reaction systems at initial times (t_0) and half-lives ($t_{1/2}$).







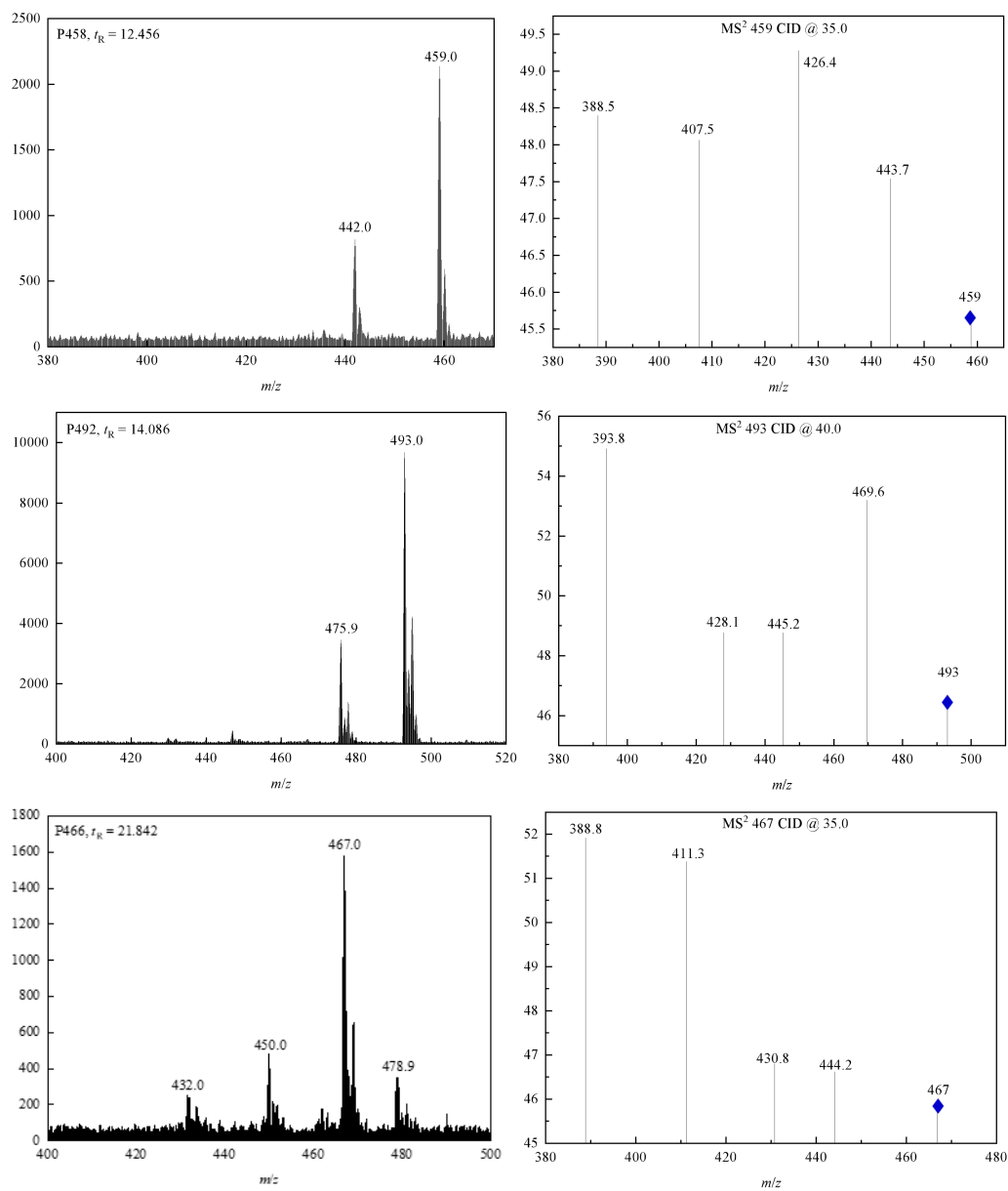


Fig. A9 ESI (+) MS and MS² spectra for chlortetracycline (CTC) and the photo-transformation products.

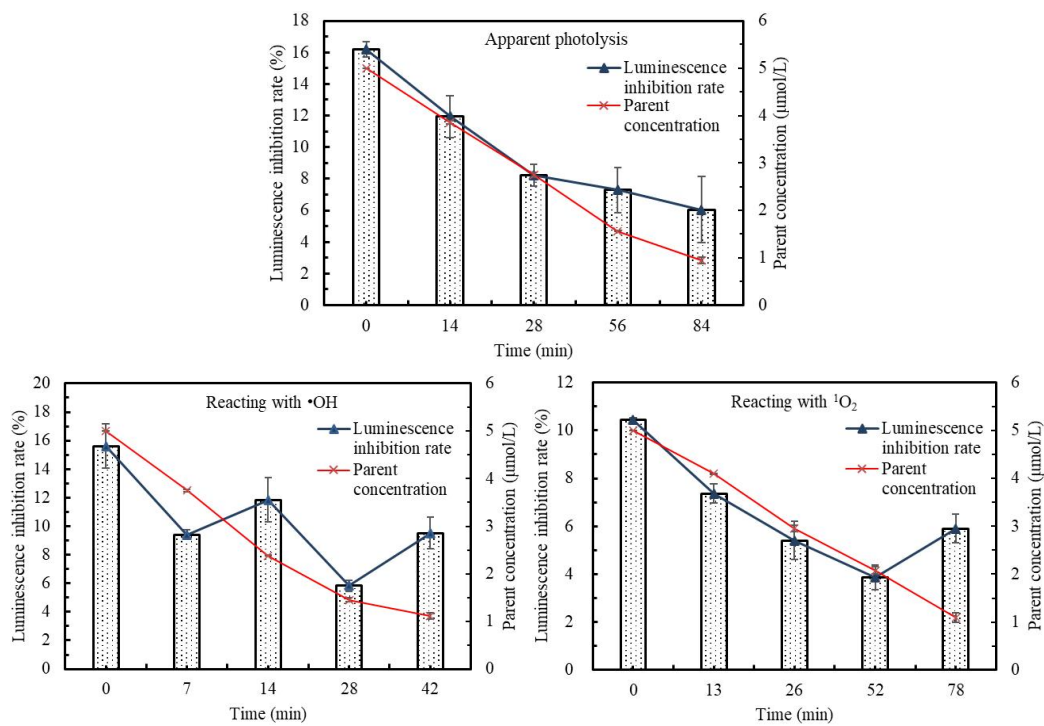


Fig. A10 Changes of parent concentrations and luminescence inhibition rates during chlortetracycline (CTC) degradation processes.

Table A1 Concentration levels of tetracyclines (TCs) in environmental waters in China and other countries

Waters, Regions	TCs	Minimum (ng/L)	Maximum (ng/L)	Median (ng/L)	Ref.
Huangpu River, Shanghai, China	TC	ND	54.3	4.2	(Chen and Zhou, 2014)
	OTC	ND	219.8	78.3	
	CTC	ND	46.7	3.6	
Dongting Lake, Hubei, China	TC	ND	21.5	–	(Liu et al., 2018)
	CTC	ND	6.5	–	
The coastal environment of Dalian, China	TC	ND	3.82	1.03	(Na et al., 2013)
	OTC	1.09	6.28	2.46	
	CTC	1.01	3	1.49	
The coastal areas of Guangdong, China	TC	0.26	3.78	–	(Xu et al., 2019)
	OTC	ND	81.54	–	
	CTC	0.43	22.53	–	
Wastewater and surface water, Jiangsu, China.	TC	ND	0.81	0.27	(Wei et al., 2011; Scheers et al., 2012)
	OTC	ND	2.2	0.22	
	CTC	ND	2.42	0.32	
Daliao River, Liaoning, China	TC	ND	10.8	1.05	(Yang et al., 2012)
	OTC	ND	13.6	2.89	
Haihe River basin, Shanxi, China	TC	ND	32	8.36	(Luo et al., 2011)
	OTC	ND	45	8.64	
Groundwater, Northern and Southwestern Regions of China	TC	ND	184	–	(Chen et al., 2018)
	OTC	ND	237	–	
	CTC	ND	8	–	
Changshou Lake, Chongqing, China	TC	10.59	16.81	12.81	(Lu et al., 2020)
	OTC	2.71	13.66	5.57	
	CTC	ND	5.60	2.46	
Municipal WWTPs, Gulf, Iran	TC	0.1	65.6	11.1	(Kafaei et al., 2018)
Surface and groundwater, Tehran, Iran	TC	280	540	–	(Javid et al., 2016)
The urban canals and lakes Hanoi, Vietnam	TC	ND	101	–	(Tran et al., 2019)
	OTC	ND	116	–	
	CTC	ND	ND	ND	
Waters, Regions	TCs	Minimum (ng/L)	Maximum (ng/L)	Median (ng/L)	Ref.
The swine treatment lagoons, United States	TC	570	1840	–	(Zhang et al., 2013)
	OTC	630	1470	–	
	CTC	300	2010	–	

Note: ND: not detected; –: not reported.

Table A2 Basic information on the tetracycline antibiotics (TCs) selected in the study

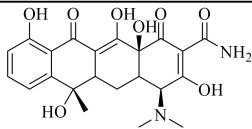
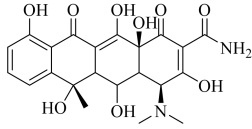
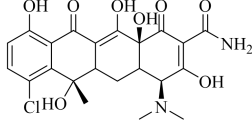
Compounds (Abbr.)	Molecular formula	CAS No.	Structures	M_w	Water solubility (g/L)
Tetracycline (TC)	$C_{22}H_{24}N_2O_8$	60-54-8		444.45	0.231
Oxytetracycline (OTC)	$C_{22}H_{24}N_2O_9$	79-57-2		460.43	0.200
Chlortetracycline (CTC)	$C_{22}H_{23}ClN_2O_8$	57-62-5		478.88	0.259

Table A3 The conditions for liquid chromatographic analysis of tetracycline antibiotics (TCs)

Compounds (Abbr.)	Injection volume (μ L)	Mobile phase v:v		Column temperature ($^{\circ}$ C)	Detection wavelength (nm)	Retention time (min)
		(Trifluoroacetic acid solution (pH = 2.4) and acetonitrile)	Flow rates (mL/min)			
Tetracycline (TC)	10	80:20	0.15	25	355	3
Oxytetracycline (OTC)	10	80:20	0.15	25	355	3
Chlortetracycline (CTC)	10	80:20	0.2	25	369	6.9

Table A4 Apparent photodegradation kinetic parameters and quantum yields (Φ_{TCs}) of tetracyclines (TCs) under different pH conditions in pure water

Compounds	pH values	k_{TCs} (min^{-1})	$t_{1/2,\text{TCs}}$ (min)	Φ_{TCs}
TC	6	$(1.21 \pm 0.09) \times 10^{-2}$	57.28 ± 2.99	$(3.53 \pm 0.25) \times 10^{-4}$
	8	$(2.96 \pm 0.30) \times 10^{-2}$	23.42 ± 1.55	$(8.57 \pm 0.86) \times 10^{-4}$
	10	$(6.26 \pm 0.16) \times 10^{-1}$	1.11 ± 0.02	$(1.89 \pm 0.05) \times 10^{-2}$
OTC	6	$(2.53 \pm 0.06) \times 10^{-2}$	27.40 ± 0.64	$(6.17 \pm 0.14) \times 10^{-4}$
	8	$(4.46 \pm 0.64) \times 10^{-2}$	15.54 ± 1.95	$(1.33 \pm 0.14) \times 10^{-3}$
	10	$(5.11 \pm 0.04) \times 10^{-1}$	1.36 ± 0.01	$(1.39 \pm 0.12) \times 10^{-2}$
CTC	6	$(2.87 \pm 0.13) \times 10^{-2}$	24.15 ± 1.15	$(7.94 \pm 0.59) \times 10^{-4}$
	8	$(4.17 \pm 0.37) \times 10^{-2}$	16.62 ± 1.35	$(1.09 \pm 0.14) \times 10^{-3}$
	10	$(2.29 \pm 0.35) \times 10^{-1}$	3.03 ± 0.39	$(6.37 \pm 1.36) \times 10^{-3}$

Table A5 The bimolecular rate constants ($k_{\text{OH,TCs}}$ and $k_{\text{1O2,TCs}}$) for the reaction between tetracycline antibiotics (TCs) and $\bullet\text{OH}/^1\text{O}_2$ under different pH conditions

Compounds	$k_{\text{ROS,TCs}}$	pH = 6	pH = 8	pH = 10
TC	$k_{\text{OH,TC}} \times 10^{-9}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	8.30 ± 0.52	72.10 ± 2.40	58.35 ± 1.91
	$k_{\text{1O2,TC}} \times 10^{-7}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	0.77 ± 0.15	21.70 ± 4.67	236.50 ± 31.82
OTC	$k_{\text{OH,OTC}} \times 10^{-9}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	8.30 ± 0.52	8.65 ± 0.28	58.39 ± 1.91
	$k_{\text{1O2,OTC}} \times 10^{-7}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	1.85 ± 0.12	131.96 ± 44.20	252.73 ± 18.00
CTC	$k_{\text{OH,CTC}} \times 10^{-9}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	7.93 ± 0.37	8.45 ± 0.20	58.35 ± 1.35
	$k_{\text{1O2,CTC}} \times 10^{-7}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	2.55 ± 0.40	95.48 ± 15.72	192.71 ± 10.01

Table A6 The bimolecular rate constants ($k_{\text{OH},i}$ and $k_{\text{1O2},i}$) for the reaction between the different dissociated species (i) of tetracycline antibiotics (TCs) and reactive oxygen species (ROS, $\bullet\text{OH}$ and $^1\text{O}_2$)

Compounds	$k_{\text{ROS},i}$	H_2TCs^0	HTCs^-	TCs^{2-}
TC	$k_{\text{OH},i} \times 10^{-9}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	6.37 ± 0.47	105.78 ± 3.40	35.29 ± 1.19
	$k_{\text{1O2},i} \times 10^{-7}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	0.26 ± 0.03	25.99 ± 6.18	344.96 ± 45.07
OTC	$k_{\text{OH},i} \times 10^{-9}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	8.35 ± 0.56	7.58 ± 0.60	97.05 ± 3.82
	$k_{\text{1O2},i} \times 10^{-7}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	3.63 ± 2.04	171.45 ± 59.25	314.99 ± 13.25
CTC	$k_{\text{OH},i} \times 10^{-9}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	8.42 ± 0.56	5.69 ± 0.63	68.93 ± 2.42
	$k_{\text{1O2},i} \times 10^{-7}$ ($\text{L}\cdot\text{mol}^{-1}\cdot\text{s}^{-1}$)	1.90 ± 0.03	114.27 ± 9.72	208.46 ± 12.08

Table A7 Rate constants of apparent photolysis and photooxidation degradation (k_{ap} and $k_{\text{oxidation}}$), and environmental multivariate photo-transformation half-lives ($t_{1/2,E}$) of tetracycline antibiotics (TCs) in surface waters with different pH at 45°N latitude

pH	k_{ap} (h ⁻¹) ^a		$k_{\text{oxidation}}$ (h ⁻¹) ^b		$t_{1/2,E}$ (h)	
	Midsummer	Midwinter	•OH	¹ O ₂	Midsummer	Midwinter
Tetracycline (TC)						
6	0.560	0.131	0.000299	0.00276	1.23	5.18
7	0.663	0.156	0.000825	0.0168	1.02	3.98
8	1.70	0.411	0.00260	0.0781	0.389	1.41
9	10.8	2.65	0.00326	0.273	0.0624	0.237
Oxytetracycline (OTC)						
6	0.685	0.159	0.000299	0.0319	0.966	3.63
7	0.890	0.207	0.000295	0.161	0.659	1.88
8	1.66	0.389	0.000311	0.481	0.324	0.796
9	4.63	1.09	0.000639	0.660	0.131	0.395
Chlortetracycline (CTC)						
6	0.880	0.209	0.000299	0.0223	0.768	3.00
7	0.938	0.223	0.000278	0.123	0.653	2.00
8	1.356	0.325	0.000311	0.346	0.407	1.03
9	4.430	1.083	0.000954	0.516	0.140	0.433

a: Estimation of the intensity of continuous illumination from the noon surface waters reported by Leifer (Leifer, 1988).

b: Estimated based on the average concentrations of •OH and ¹O₂ in natural waters (1×10^{-15} mol/L and 1×10^{-12} mol/L, respectively).

Table A8 Retention time (t_R), molecular weight (M_w , calculated based on the most abundant isotopes), and MS fragment m/z of chlortetracycline (CTC) and the photo-transformation products

reaction types	compounds	t_R (min)	M_w	ESI(+)MS m/z	ESI(+)MS ² m/z
Apparent photolysis	CTC	25.827	478	479 [M+H] ⁺	479 > 459, 441, 416, 375
	P444	9.147	444	445 [M+H] ⁺	445 > 426, 418, 398, 370
Reacting with •OH	CTC	25.670	478	479 [M+H] ⁺	479 > 454, 431, 415, 371
	P462	5.022	462	463 [M+H] ⁺	463 > 448, 430, 400, 357
	P444	9.188	444	445 [M+H] ⁺	445 > 422, 395, 375, 355
	P360	10.818	360	361 [M+H] ⁺	361 > 343, 331, 308, 287
	P450	11.304	450	451 [M+H] ⁺	451 > 432, 416, 381, 342
	P494	12.382	494	495 [M+H] ⁺	495 > 479, 453, 430, 412
	P434	14.605	434	435 [M+H] ⁺	435 > 416, 405, 381, 342
	CTC	25.678	478	479 [M+H] ⁺	479 > 460, 445, 434, 388
Reacting with ¹ O ₂	P462	5.310	462	463 [M+H] ⁺	463 > 422, 344, 315, 299
	P510	7.393	510	511 [M+H] ⁺	511 > 491, 471, 397, 347
	P444	9.147	444	445 [M+H] ⁺	445 > 418, 406, 350, 305
	P360	10.851	360	361 [M+H] ⁺	361 > 343, 325, 302, 241
	P458	12.456	458	459 [M+H] ⁺	459 > 444, 426, 408, 389
	P492	14.086	492	493 [M+H] ⁺	493 > 470, 445, 428, 394
	P466	21.842	466	467 [M+H] ⁺	467 > 444, 431, 411, 389

Note: The molecular weights (M_w) calculated based on the most abundant isotopes.

Table A9 Acute toxicities of chlortetracycline (CTC) and the 10 intermediates to fish, daphnid, and green algae

Compounds	96 h LC_{50} (Fish)	48 h LC_{50} (Daphnid)	96 h EC_{50} (Green Algae)
CTC	5340.0	466.0	712.0
P444	13100.0	1060.0	1890.0
P462	777.0	78.4	89.9
P494	7800.0	663.0	1070.0
P450	8050.0	677.0	1110.0
P434	2030.0	190.0	254.0
P360	11.5	7.54	1.05
P510	2640.0	244.0	332.0
P458	2560.0	235.0	324.0
P492	1040.0	103.0	122.0
P466	85400.0	6010.0	14100.0

Note: LC_{50} and EC_{50} are the semi-lethal concentration and the semi-maximum effect concentration (mg/L), respectively.

Table A10 The significant difference analysis in toxicity of chlortetracycline (CTC) at different degradation times in diverse reaction processes

Time	0.5 $t_{1/2}$	1 $t_{1/2}$	2 $t_{1/2}$	3 $t_{1/2}$
Apparent photolysis				
0	0.073	0.420	0.066	0.040*
0.5 $t_{1/2}$		0.145	0.867	0.297
$t_{1/2}$			0.125	0.060
2 $t_{1/2}$				0.359
Reacting with •OH				
0	0.037*	0.984	0.042*	0.394
0.5 $t_{1/2}$		0.038*	0.770	0.056
$t_{1/2}$			0.042*	0.403
2 $t_{1/2}$				0.067
Reacting with ¹ O ₂				
0	0.046*	0.026*	0.036*	0.032*
0.5 $t_{1/2}$		0.151	0.539	0.346
$t_{1/2}$			0.292	0.455
2 $t_{1/2}$				0.040*

Note: * indicates significant difference ($p < 0.05$).

References

- Chen K, Zhou J (2014). Occurrence and behavior of antibiotics in water and sediments from the Huangpu River, Shanghai, China. *Chemosphere*, 95: 604-612
- Chen L, Lang H, Liu F, Jin S, Yan T (2018). Presence of antibiotics in shallow groundwater in the northern and southwestern regions of China. *Groundwater*, 56(3): 451-457
- Gulkowska A, He Y, So M K, Yeung L W, Leung H W, Giesy J P, Lam P K, Martin M, Richardson B J (2007). The occurrence of selected antibiotics in Hong Kong coastal waters. *Marine Pollution Bulletin*, 54(8): 1287-1293
- Hu Y, Yan X, Shen Y, Di M, Wang J (2018). Antibiotics in surface water and sediments from Hanjiang River, Central China: Occurrence, behavior and risk assessment. *Ecotoxicology and Environmental Safety*, 157: 150-158
- Javid A, Mesdaghinia A, Nasserli S, Mahvi A H, Alimohammadi M, Gharibi H (2016). Assessment of tetracycline contamination in surface and groundwater resources proximal to animal farming houses in Tehran, Iran. *Journal of Environmental Health Science and Engineering*, 14: 1-5
- Jia J, Guan Y, Cheng M, Chen H, He J, Wang S, Wang Z (2018). Occurrence and distribution of antibiotics and antibiotic resistance genes in Ba River, China. *Science of the Total Environment*, 642: 1136-1144
- Jiang L, Hu X, Yin D, Zhang H, Yu Z (2011). Occurrence, distribution and seasonal variation of antibiotics in the Huangpu River, Shanghai, China. *Chemosphere*, 82(6): 822-828
- Kafaei R, Papari F, Seyedabadi M, Sahebi S, Tahmasebi R, Ahmadi M, Sorial G A, Asgari G, Ramavandi B (2018). Occurrence, distribution, and potential sources of antibiotics pollution in the water-sediment of the northern coastline of the Persian Gulf, Iran. *Science of the Total Environment*, 627: 703-712
- Lei X, Lu J, Liu Z, Tong Y, Li S (2015). Concentration and distribution of antibiotics in water-sediment system of Bosten Lake, Xinjiang. *Environmental Science and Pollution Research*, 22: 1670-1678

- Leifer A (1988). The kinetics of environmental aquatic photochemistry: Theory and practice. American Chemical Society
- Li F, Chen L, Chen W, Bao Y, Zheng Y, Huang B, Mu Q, Wen D, Feng C (2020). Antibiotics in coastal water and sediments of the East China Sea: Distribution, ecological risk assessment and indicators screening. *Marine Pollution Bulletin*, 151: 110810
- Liang X, Chen B, Nie X, Shi Z, Huang X, Li X (2013). The distribution and partitioning of common antibiotics in water and sediment of the Pearl River Estuary, South China. *Chemosphere*, 92(11): 1410-1416
- Liu X, Lu S, Guo W, Xi B, Wang W (2018). Antibiotics in the aquatic environments: A review of lakes, China. *Science of the Total Environment*, 627: 1195-1208
- Lu L, Liu J, Li Z, Zou X, Guo J, Liu Z, Yang J, Zhou Y (2020). Antibiotic resistance gene abundances associated with heavy metals and antibiotics in the sediments of Changshou Lake in the three Gorges Reservoir area, China. *Ecological Indicators*, 113: 106275
- Luo Y, Xu L, Rysz M, Wang Y, Zhang H, Alvarez P J (2011). Occurrence and transport of tetracycline, sulfonamide, quinolone, and macrolide antibiotics in the Haihe River Basin, China. *Environmental Science & Technology*, 45(5): 1827-1833
- Na G, Fang X, Cai Y, Ge L, Zong H, Yuan X, Yao Z, Zhang Z (2013). Occurrence, distribution, and bioaccumulation of antibiotics in coastal environment of Dalian, China. *Marine Pollution Bulletin*, 69(1-2): 233-237
- Na G, Gu J, Ge L, Zhang P, Wang Z, Liu C, Zhang L (2011). Detection of 36 antibiotics in coastal waters using high performance liquid chromatography-tandem mass spectrometry. *Chinese Journal of Oceanology and Limnology*, 29: 1093-1102
- Scheers T, Appels L, Dirckx B, Jacoby L, Van Vaeck L, Van Der Bruggen B, Luyten J, Degève J, Van Impe J, Dewil R (2012). Evaluation of peroxide based advanced oxidation processes (AOPs) for the degradation of ibuprofen in water. *Desalination and Water Treatment*, 50(1-3): 189-197
- Tran N H, Hoang L, Nghiem L D, Nguyen N M H, Ngo H H, Guo W, Trinh Q T, Mai N H, Chen H, Nguyen D D (2019). Occurrence and risk assessment of multiple classes of antibiotics in urban canals and lakes in Hanoi, Vietnam. *Science of the Total Environment*, 692: 157-174
- Wang G, Zhou S, Han X, Zhang L, Ding S, Li Y, Zhang D, Zarin K (2020). Occurrence, distribution, and source track of antibiotics and antibiotic resistance genes in the main rivers of Chongqing city, southwest China. *Journal of Hazardous Materials*, 389: 122110
- Wang J, Wei H, Zhou X, Li K, Wu W, Guo M (2019). Occurrence and risk assessment of antibiotics in the Xi'an section of the Weihe River, northwestern China. *Marine Pollution Bulletin*, 146: 794-800
- Wang Z, Du Y, Yang C, Liu X, Zhang J, Li E, Zhang Q, Wang X (2017). Occurrence and ecological hazard assessment of selected antibiotics in the surface waters in and around Lake Honghu, China. *Science of the Total Environment*, 609: 1423-1432
- Wei R, Ge F, Huang S, Chen M, Wang R (2011). Occurrence of veterinary antibiotics in animal wastewater and surface water around farms in Jiangsu Province, China. *Chemosphere*, 82(10): 1408-1414
- Xu K, Wang J, Gong H, Li Y, Zhou L, Yan M (2019). Occurrence of antibiotics and their associations with antibiotic resistance genes and bacterial communities in Guangdong coastal areas. *Ecotoxicology and Environmental Safety*, 186: 109796
- Yan M, Xu C, Huang Y, Nie H, Wang J (2018). Tetracyclines, sulfonamides and quinolones and their corresponding resistance genes in the Three Gorges Reservoir, China. *Science of the Total Environment*, 631: 840-848
- Yang C, Wang L, Hou X, Chen J (2012). Analysis of pollution levels of 16 antibiotics in the river of the Daliao River System. *Chinese Journal of Chromatography*, 30(8): 756-762

- Zhang G, Liu X, Lu S, Zhang J, Wang W (2020a). Occurrence of typical antibiotics in Nansi Lake's inflowing rivers and antibiotic source contribution to Nansi Lake based on principal component analysis-multiple linear regression model. *Chemosphere*, 242: 125269
- Zhang G, Lu S, Wang Y, Liu X, Liu Y, Xu J, Zhang T, Wang Z, Yang Y (2020b). Occurrence of antibiotics and antibiotic resistance genes and their correlations in lower Yangtze River, China. *Environmental Pollution*, 257: 113365
- Zhang Y, Zhang C, Parker D B, Snow D D, Zhou Z, Li X (2013). Occurrence of antimicrobials and antimicrobial resistance genes in beef cattle storage ponds and swine treatment lagoons. *Science of the Total Environment*, 463: 631-638
- Zhao B, Xu J, Zhang G, Lu S, Liu X, Li L, Li M (2021). Occurrence of antibiotics and antibiotic resistance genes in the Fuxian Lake and antibiotic source analysis based on principal component analysis-multiple linear regression model. *Chemosphere*, 262: 127741
- Zhou L, Wu Q, Zhang B, Zhao Y, Zhao B (2016). Occurrence, spatiotemporal distribution, mass balance and ecological risks of antibiotics in subtropical shallow Lake Taihu, China. *Environmental Science: Processes & Impacts*, 18(4): 500-513
- Zou S, Xu W, Zhang R, Tang J, Chen Y, Zhang G (2011). Occurrence and distribution of antibiotics in coastal water of the Bohai Bay, China: Impacts of river discharge and aquaculture activities. *Environmental Pollution*, 159(10): 2913-2920