

DOCTORAL THESIS

Probing the environmental fate of emerging contaminants and their ecological impacts on aquatic environment

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Abstract

In recent years, new classes of aquatic pollutants have received growing concerns from the environmentalists due to their unclear effects and increasing release into the water environment. Generally, before entering the aquatic ecosystem, many of these emerging contaminants are deemed safe active ingredients in the commercial products. This may lead to the underestimation of their potential impacts to the environment, once these substances were drained into the natural waters. So far, a wide range of emerging contaminants are still not well studied. In this thesis work, the environmental behaviors for two typical groups of emerging contaminants, which are frequently used in personal care products, are systematically probed in the aquatic ecosystem. One is the hydrophilic compounds – artificial sweeteners and the other one is the moderate hydrophobic compounds – ultraviolet (UV) filters.

Acesulfame and sucralose are two of the most commonly used artificial sweeteners, which are a newly recognized class of emerging contaminants due to their widespread occurrence and extreme persistence in water environment. To date, however, their environmental behaviors, fates as well as long term ecotoxicological contributions in our water resources still remain largely unknown. In this work, a photodegradation investigation with UV/TiO₂ was performed on these two artificial sweeteners, which projected their potential impacts under prolonged exposure to intensive solar irradiation. Real-time observation of the degradation profiles in both sweeteners illustrated that formation of new photo by-products under prolonged UV irradiation is highly viable. For the first time, acute toxicity for the degradates of these two sweeteners were measured and the enhancement in their ecotoxicity has been observed during the irradiation. In an attempt to neutralize this prolonged environmental threat, the feasibility of UV/TiO₂ as an effective mineralization process in wastewater treatment was evaluated for both sweeteners. As a result, relatively higher removal efficiencies for acesulfame and sucralose were achieved in this study.

For UV filters, a comprehensive investigation on their environmental behaviors and impacts was described in this thesis, covering the occurrence study in surface water and biota, environmental fate and their ecological risks. In the first step, an analytical method based on ultra performance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS) was developed for the simultaneous determination of twelve UV filters in the environmental waters, which achieved good sensitivity to sub-ng/L levels. To monitoring their occurrence in local marine environment, the regional distribution of these sunscreen compounds was monitored along the coastline of Hong Kong. At the same time, considering the intensive consumption of sunscreen products in hot weathers, seasonal variation of UV filters between summer and winter was taken into consideration in our occurrence study. Additionally, we also monitored their occurrence in the drinking water supply system of Hong Kong, including the source water in Dongjiang, raw water in local reservoirs and tap water collected from urban communities, to support a good management on the reliable water supply. Furthermore, the distribution results indicated that the incomplete removal of UV filters in the effluents could be a major source for their release into the environmental waters. Aiming to achieve the complete removal of such

compounds, a powerful technique of ozonation was employed in this study and the preliminary operational conditions were further provided. Secondly, in order to gain a better understanding of their environmental fate in surface water, a simulative catalytic photodegradation study with UV/TiO₂ were performed to investigate the photostability, phototoxicity and the transformation pathways of seven selected UV filters. During the photodegradation, several transformation products were primarily identified. Thirdly, since lipophilic sunscreens were easily accumulated in the biota, we also made efforts to monitor their occurrence in marine organisms of different species, including farmed fish, wild mussels, prawns and sea urchin, which were collected from local offshore areas. Last but not the least, according to the measured concentrations of UV filters in Hong Kong marine environment, we further performed the ecological risk assessment with the approach specific to the marine aquatic compartment, so as to provide the overall understanding on the environmental impacts of these UV filters.

Table of Contents

Declaration	i
Abstract	ii
Acknowledgement	iv
Table of Contents	v
List of Tables	ix
List of Figures	x
List of Abbreviations and Symbols	xi
Chapter 1 Introduction	1
1.1 Emerging contaminants	1
1.2 Current status of studies in aquatic environment	3
1.3 Target compounds in this thesis work	7
1.3.1 Artificial sweeteners	7
1.3.2 UV filters	8
1.4 Objectives of this thesis work.....	9
1.5 References	10
Chapter 2 Photodegradation and Ecotoxicity Study of Artificial Sweeteners	17
2.1 Introduction	17
2.2 Experimental.....	19
2.2.1 Chemicals and reagents	19
2.2.2 Photodegradation profiling	19
2.2.3 Instrumental analysis	19
2.2.4 Bioluminescent Microtox test.....	20
2.2.5 Removal treatment.....	21
2.3 Results and discussions	22
2.3.1 Degradation toxicity	22
2.3.1.1 Sucralose.....	24
2.3.1.2 Acesulfame	26

2.3.2 Removal treatment.....	29
2.4 Chapter summary.....	34
2.5 References	35
Chapter 3 A Comprehensive Occurrence Investigation of UV Filters in Local Aquatic Environment and Their Removal Treatment Study	40
3.1 Introduction	40
3.2 Experimental.....	44
3.2.1 Chemicals and reagents	44
3.2.2 Sample locations and collection	44
3.2.2.1 Seasonal collection of seawater.....	44
3.2.2.2 Drinking water supply system of Hong Kong.....	46
3.2.3 Analytical method.....	48
3.2.4 Bench-scale ozonation experiments	49
3.2.5 Instrumental analysis	50
3.3 Results and discussions	52
3.3.1 Method validation.....	52
3.3.2 Environmental occurrence of UV filters in surface seawater.....	53
3.3.2.1 Occurrence in Hong Kong seawater.....	53
3.3.2.2 Seasonal pattern.....	57
3.3.2.3 Spatial distribution in Hong Kong.....	59
3.3.3 Occurrence of UV filters in drinking water supply system.....	63
3.3.4 Removal treatment of ozonation	65
3.4 Chapter summary.....	69
3.5 References	71
Chapter 4 Photodegradation and Ecotoxicity Study of UV Filters.....	77
4.1 Introduction	77
4.2 Experimental.....	80
4.2.1 Chemicals and reagents	80
4.2.2 Photodegradation	81
4.2.3 Instrumental analysis	82
4.2.4 Bioluminescent Microtox test.....	83
4.3 Results and discussions	85

4.3.1 Ethyl <i>p</i> -amino benzoic acid (Et-PABA)	85
4.3.1.1 Catalytic photodegradation profiles	85
4.3.1.2 Identification of photodegradation products and their transformation profiles	88
4.3.1.3 Phototoxicity evaluation	92
4.3.2 3-Benzylidene camphor (3-BC)	94
4.3.2.1 Catalytic photodegradation profile	94
4.3.2.2 Identification of photodegradation products and their transformation profiles	96
4.3.3 Benzophenone group	101
4.3.3.1 Catalytic photodegradation profiles	101
4.3.3.2 Phototoxicity evaluation	105
4.3.3.3 Degradation products of BP-type UV filters	106
4.4 Chapter summary.....	108
4.5 References	110

Chapter 5 Occurrence Study of UV filters in Marine Organisms and Their Ecological Risk Assessment in Hong Kong Marine Environment 115

5.1 Introduction	115
5.2 Experimental.....	117
5.2.1 Chemicals and reagents	117
5.2.2 Sample collection and preparation	117
5.2.3 Analytical procedures	119
5.2.4 Instrumental analysis	120
5.2.5 Analytical method validation.....	121
5.3 Results and discussions	122
5.3.1 Method validation results	122
5.3.1.1 Validation results	122
5.3.1.2 Matrix effect study	124
5.3.2 Environmental occurrence of UV filters in marine organisms....	125
5.3.2.1 Concentrations in marine organisms	125
5.3.2.2 Spatial distribution in Hong Kong.....	130
5.3.2.3 Global comparison.....	133
5.3.3 Ecological risk assessment	135
5.4 Chapter summary.....	140

5.5 References	141
Chapter 6 Conclusions and Outlook of Environmental Research	146
6.1 Thesis summary	146
6.2 Future needs.....	149
Outcome of This Thesis Work	150
Curriculum Vitae.....	151