DOCTORAL THESIS

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

Sang, Ziye

Date of Award:
2016

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/TiO_2 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/TiO_2 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/TiO_2 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