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Particular Photodegradation Processes of Organic Pollutants in Surface Waters

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Abstract

This article describes the processes that affect the chemical composition of surface waters, including annual, seasonal, and long-term variations. It provides an overview of the effect of sunlight penetration into the water column of natural water bodies. Photochemical processes are strongly affected by sunlight irradiation, the penetration of which into the water column depends on several factors, such as the magnitude and color of suspended solids and colloids, the presence of phytoplankton, particulate and dissolved organic matter, among others. Different photochemical processes are shown and discussed throughout this work. The photo transformation of pollutants in waters from natural environments, such as pesticides, detergents, and pharmaceuticals (drugs as emerging pollutants), is summarized. These techniques can be used in laboratory experiments to simulate the abiotic transformation of these pollutants in the euphotic zone (the zone with the greatest light reception), which sometimes leads to potentially harmful transformation intermediates. Similarly, the role played by reactive oxygen species (superoxide anion, singlet oxygen, hydroxyl radicals) in surface waters is described. The latter, produced in aqueous solution by photo-excited organic matter, has a micro-heterogeneous distribution in the solid phase and in the hydrophobic nuclei of humic substances, which can have important implications for the degradation of hydrophilic and hydrophobic pollutants.

Keywords

Organic Pollutants, Surface Waters, Photosensitizer, Photodynamic, Photo-Oxidation

1. Introduction

Photochemistry plays a fundamental role during the transformation of organic

pollutants in natural systems using solar energy, therefore, its application frontiers in this area are limited by those spaces to which sunlight penetrates.

For its part, environmental geochemistry is the discipline of Earth sciences dedicated to the study of the source, transformation, transport and destination of chemical pollutants on the planet, specifically in surface systems such as terrestrial, aquatic, biotic and atmospheric, in most of which sunlight is present and has a fundamental role in the degradation of some pollutants. This is how sunlight links photochemistry with environmental geochemistry, on the one hand, photochemistry studies the photodegradative reactions of organic pollutants, while on the other, environmental geochemistry provides information of interest on the natural factors that may be involved (Andrady, 2011; Auta et al., 2017; Bergmann et al., 2019). Affecting, either promoting or limiting, the photochemical reactions that transform organic pollutants as pesticide and drugs. In this sense, some authors have used the term photo-geochemistry, referring to the study of chemical reactions that occur in the surface terrestrial system due to the action of sunlight on minerals, plant residues, dissolved organic matter, organic and inorganic aerosols and gases, associated with terrestrial, aquatic or tropospheric systems (Schrauzer et al., 1983; Canfield et al., 2010; Propopapadaki et al., 2017; Walkinshaw et al., 2020).

2. Synergy between Photochemistry and Geochemistry

A relationship considered as synergistic can be observed between the climate change of our tropical region, the incidence of solar radiation, and the photochemical processes occurring in the emerging contaminants of our surface waters.

A brief review of the synergy between both disciplines and its impact on the understanding of the transformation phenomena of organic pollutants in the natural surface environment is described here. Information on the link between surface water photochemistry and climate is currently scarce, as only a few studies have been devoted to the subject (Ceccarini et al., 2018; Cox et al., 2019; GESAMP, 2015). Based on the limited knowledge currently available, the present inferences can be made as follows:

1) Global warming can cause greater leaching (or solid-liquid extraction) of ionic solutes from soils and sediments to surface waters, including, in addition to cations, major anions such as bicarbonate, product of the chemical weathering of siliciclastic minerals by means of hydrolysis reaction with carbonic acid, as well as biologically unstable species such as sulfate. Although the main source of bicarbonate supply is chemical weathering, the preferential biodegradation of sulfate followed by its microbiological elimination can also increase alkalinity, favoring the generation of the carbonate radical, CO_3^-. However, these phenomena would be easily compensated by fluctuations in dissolved organic carbon (DOC), which is inversely correlated with CO_3^-. Therefore, gaining insight into the evolution of dissolved organic carbon (DOC) is a key topic in understanding the link between photochemistry and climate (Holmes et al., 2012; Kane et al., 2020).

2) Climate change could exacerbate water scarcity in the dry season for some regions. Fluctuations in the water column could profoundly alter the photochemistry, which is generally favored in shallow waters. However, this negative water imbalance would strongly affect the predominant photoinduced processes. The decrease in the volume of runoff water without important changes in the concentration of solutes would mainly favor the reactions induced by hydroxyl and carbonate radicals ($\cdot OH$ and CO_3^-). On the contrary, concentration by evaporation would promote reactions mediated by singlet oxygen ($^1O_2$) and by the triple states of chromophoric dissolved organic matter (^{3}CDOM*).

3) In a warmer climate, the stratification period of the lakes would last longer, increasing the photochemical reactions in the epilimnion but at the same time keeping the water of the hypolimnion in the dark for longer periods.

Photochemical reactions are highly dependent on solar radiation, water chemistry, and depth. The chemistry of the water and the irradiation on it produced by incident sunlight affect the concentration values of photo-reactants (DOM, nitrate and nitrite) and of transient reactive species scavengers (mainly $\cdot OH$ and $CO_3^{\cdot-}$). Finally, depth is important because deep areas of a body of water are poorly lit by sunlight. Therefore, photochemical reactions are favored in shallow water bodies. Climate change can affect the chemistry and depth of water in several ways (Delpla et al., 2009; Leavitt et al., 2003). For example, a change in runoff and the rate of weathering can change the concentration of cationic and anionic species in receiving waters. The chemical weathering of minerals, as well as the biological lability of anions such as sulfate, would cause an increase in alkalinity, which would alter the concentration values of bicarbonate and carbonate and modify the rate of formation of CO_3 after carbonate oxidation and bicarbonate by OH (Nesbitt, 1980; Schindler, 2009).

As far as we know, the effects of climate on surface water photochemistry are largely unknown at this time, so it can be argued that the topic does not even exist as a research field in Venezuela. Therefore, this contribution is intended to present the results of some pioneering work and our personal opinion on the potential impact that climate change in tropical areas can have on photochemical processes in freshwater.

3. Photochemical Processes in Surface Waters

Sensitized photolysis is triggered by the absorption of radiation by photoactive compounds called photosensitizers, the main ones in surface waters are nitrate, nitrite and most notably. Dissolved organic matter (DOM) consists of organic compounds dissolved in water that can be derived from the microbiological transformation of animal and plant remains. DOM is composed of small primary molecular fractions (100 - 200 Da) that are organized into supramolecular structures. These aggregates are composed of aromatic and aliphatic hydrocarbon structures with various functional groups (e.g., $CONH_2$, -COOH, -OH, -CO) (Leavitt et al., 2003; Vione & Scozzaro, 2019). The aromatic/quinone fractions play a very important

role as they can absorb sunlight. They are contained in humic and fulvic acids, product of the biodegradation of lignin or of the oligomerization/supramolecular association of smaller compounds, caused by photo-oxidation (Leenher & Croue, 2003). The fraction of DOM that can absorb sunlight is called MODC (Boule et al., 2005). The absorption of radiation by DOMC causes the formation of excited singlet states (^{1}DOMC*) that are transformed by inter-system crossover (ISC) into excited triplet states (DOMC*). The reactivity of dissolved compounds with ^{3}DOM* is greater than with ^{1}MODC* due to the longer useful life of the former (Canonica, 2007; Halladja et al., 2007). The $\cdot OH$ radical is produced by irradiation of nitrate, nitrite and DOMC. In the latter case, the details of the process are not yet clear, although the significant but not exclusive role of H_2O_2 is known which could, for example, be involved in Fenton reactions (Page et al., 2011; Vermilyea & Voelker, 2009). On the other hand, there is ample evidence of an independent H_2O_2 pathway from OH generated by irradiated DOMC (Page et al., 2011; Dong & Rosario-Ortiz, 2012), which could possibly be counted among the processes by triplet-sensitized oxidation of OH- and/or H_2O (Sur et al., 2011). The radical CO_3^- is produced after the oxidation of CO_3^{2-} and HCO_3^- by OH, as well as by oxidation of carbonate by ^{3}DOMC*. The ^{3}DOMC* triplet state can degrade pollutants on its own, but can also react with O_2 to form the reactive transient $^1O_2$, which is also involved in pollutant breakdown. The reactions that can take place in natural water systems (apart from the not yet clear formation of OH from irradiated MODC) are illustrated below (Canonica, 2007; Singh & Sharma, 2008):

$$NO_3 + h\nu + H^+ \longrightarrow \cdot OH + NO_2$$

$$NO_2^- + h\nu + H^+ \longrightarrow \cdot OH + NO$$

$$OH + HCO_3^- \longrightarrow H_2O + CO_3^- \cdot$$

$$\cdot OH + CO_3^{2-} \longrightarrow OH^- + CO_3^-$$

$$MODC + h\nu \longrightarrow {}^1DOMC^* \xrightarrow{ISC} {}^3MODC^*$$

$${}^3MODC^* + CO_3^{2-} \longrightarrow DOMC^- + CO_3^-$$

$${}^3MODC^* + O_2 \longrightarrow DOMC + {}^1O_2$$

The transient species $\cdot OH$, CO_3^-, $^1O_2$ and ^{3}DOMC* can induce the degradation of xenobiotics, but with important differences. In particular, the $\cdot OH$ radical is primarily involved in decontamination with a generally limited formation of harmful intermediates. Compared with $\cdot OH$, the probability of forming harmful intermediates is considerably higher in the case of $^1O_2$, CO_3^- and ^{3}DOMC*.

Photochemical reactions involving $\cdot OH$, CO_3^-, $^1O_2$ and ^{3}DOMC* are highly dependent on the chemistry and depth of the water. The depth effect is mainly affected by absorbent substances dissolved in water, and most notably by DOMC. The latter is the main absorber of radiation in surface waters between 300 and 500 nm, which is the most significant wavelength range for photoinduced processes.

The absorption of sunlight by DOMC plays an important role in reducing the intensity of solar radiation in the water column. Therefore, the lower region of a body of water will be less illuminated than the surface, which also promotes faster photochemical processes in shallow water bodies compared to deep ones. In the latter, the high photo-activity in the superficial zone is compensated by the lack of photoactivity in depth. Such an effect reduces the importance of photochemical reactions as the depth of the water column increases, being that it also protects aquatic life from exposure to harmful UV radiation (Dattilo et al., 2005).

The absorption of DOMC shows an exponential decrease with the increase of the wavelength, and the absorption in the different spectral ranges decreases as UVB > UVA > visible. Consequently, penetration of the column by radiation is in the order UVB < UVA < visible. Nitrate primarily absorbs UVB radiation, and its photochemistry is highly inhibited with depth. A lower degree of inhibition is observed with nitrite that absorbs in UVA rays, while MODC, which also absorbs in visible rays, is less affected by depth. Nitrite and nitrate are direct sources of $\cdot OH$ and indirect sources of CO_3^-, while DOC produces only $^1O_2$ and $^3DOMC^*$. Therefore, the relative importance of $^1O_2$ and $^3DOMC^*$ versus $\cdot OH$ and CO_3^- increases as depth increases (Minella et al., 2011; De Laurentiis et al., 2013).

The photochemical reactivity of surface waters has been the subject of specific studies in some environments, but so far, no long-term campaigns have been carried out. Consequently, there is no information on photochemical behavior in defined environments in the last 20 - 30 years, and data for the next few decades is clearly not yet available (Gewert et al., 2015). For this reason, the relationship between climate change and surface water photochemistry is largely unknown and almost non-existent as a research topic.

Some photodegradation processes of drugs such as aceclofenac, clozapine and benzodiazepines are referred to as emerging contaminants in water (Vargas, Tropper, & Gavidia, 2001; Vargas et al., 2007).

The limited information currently available on the link between surface water photochemistry and climate change is explained by the relatively few studies that have been carried out so far on this topic. In the current state of knowledge, the following conclusions can be drawn:

1) Increased chemical weathering of siliciclastic and minerals with biologically labile sulfate could lead to higher alkalinity of the water and higher $[CO_3^{2-}]/[HCO_3^-]$ ratios. This could favor the formation of CO_3 because carbonate is considerably more reactive towards $\cdot OH$ than bicarbonate. However, this effect could be largely offset by the presence of COD because the MOD is inversely correlated with $[CO_3^-]$. Therefore, a key issue in the link between photochemistry and climate is represented by the understanding of the system, especially considering the variations of DOM by increasing temperature. An important confounding factor in this context is the fact that DOM variations are also connected to anthropogenic changes.

2) A decrease in the depth of the column of water bodies during the dry season

would generally favor photochemical reactions. In particular, the decrease in the volume of water without significant changes in the concentration of solutes would favor the reactions induced by OH and CO_3^-. In contrast, the phenomena of concentration by evaporation would favor the processes that involve ^{3}DOMC* and $^1O_2$ (Delpla et al., 2009).

3) A longer duration of the hot season would prolong the stratification period in the lakes, which would increase the importance of photochemical reactions in the epilimnion but, at the same time, would keep the deep waters of the hypolimnion in the dark for longer.

Relationship between environmental phenomena and photochemical processes.

Phenomenon Coloration and turbidity	Increased steps	Inhibited steps
Decrease in DOC	·OH , CO_3^-	^{3}MODC*, $^1O_2$
Photobleaching	direct photolysis	
Increases in DOC	direct photolysis	^{3}MODC*, $^1O_2$
Photobleaching		·OH , CO_3^-
Increased alkalinity	CO_3^- (rarely)	·OH
Stratification in lakes for long summers	All (in *epilimnion*)	All (in *hypolimnion*)
Shorter ice sheet period	All processes	
Evaporation concentration	^{3}MODC*, $^1O_2$	
Water outlet without compensation	·OH , CO^- direct photolysis	
Lower flow velocity in rivers	All processes	
Higher flow velocity in rivers		All processes

4. Conclusion

In conclusion, the main foreseeable effects of climate change for photochemical processes in surface waters and some types of pollutants that are preferentially degraded by the different photogenerated transient species (·OH , CO_3^- , ^{3}DOMC*, $^1O_2$) can be summarized. Climatic effects can also induce considerable fluctuations in the availability of water, with important changes in the water column of the lakes, as well as in the depth and speed of river flow. In both cases, conditions of water scarcity can improve the kinetics of photoinduced reactions. The importance of photochemistry in the degradation of pesticides under environmental conditions has been widely demonstrated, emerging as a possible technological solution for the complete mineralization of pesticides, with higher efficiencies and lower costs than conventional methodologies. However, it would be convenient to mention that the experimental studies to date have been directed towards the conditions related to the chemical characteristics of water. A greater understanding of the natural system could provide parameters that optimize the current methods of degradation

of pesticides, drugs, and other chromophore pollutants. This article aims to draw scientists' attention to this line of research in order to improve the treatment of contaminated surface waters and their significant relationship with climate change in tropical regions like Venezuela, which has high solar radiation. Photochemical reactions in surface waters have significant environmental consequences.

The connection between the global environment and the biosphere is striking, exemplified by the recent identification of ozone layer depletion and the ensuing rise in high-energy ultraviolet radiation. The harmful impacts of this radiation on living systems are now widely recognized. The revelation of these consequences brought the field of protosciences into the spotlight, driving an unprecedented collaboration between scientists and policymakers to establish international agreements aimed at curbing air pollution.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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