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Q13 **Intrinsic chemiluminescence production from the degradation**
 2 **of haloaromatic pollutants during environmentally-friendly**
 3 **advanced oxidation processes: Mechanism, structure–activity**
 4 **relationship and potential applications**

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ABSTRACT

The ubiquitous distribution of halogenated aromatic compounds (XAr) coupled with their 17
 carcinogenicity has raised public concerns on their potential risks to both human health and the 18
 ecosystem. Recently, advanced oxidation processes (AOPs) have been considered as an 19
 “environmentally-friendly” technology for the remediation and destruction of such recalcitrant 20
 and highly toxic XAr. During our study on the mechanism of metal-independent production of 21
 hydroxyl radicals ($\cdot\text{OH}$) by halogenated quinones and H_2O_2 , we found, unexpectedly, that an 22
 unprecedented $\cdot\text{OH}$ -dependent two-step intrinsic chemiluminescence (CL) can be produced by 23
 H_2O_2 and tetrachloro-*p*-benzoquinone, the major carcinogenic metabolite of the widely used 24
 wood preservative pentachlorophenol. Further investigations showed that, in all $\cdot\text{OH}$ -generating 25
 systems, CL can also be produced not only by pentachlorophenol and all other halogenated 26
 phenols, but also by all XAr tested. A systematic structure–activity relationship study for all 19 27
 chlorophenolic congeners showed that the CL increased with an increasing number of 28
 Cl-substitution in general. More importantly, a relatively good correlation was observed between 29
 the formation of quinoid/semiquinone radical intermediates and CL generation. Based on these 30
 results, we propose that $\cdot\text{OH}$ -dependent formation of quinoid intermediates and electronically 31
 excited carbonyl species is responsible for this unusual CL production; and a rapid, sensitive, 32
 simple, and effective CL method was developed not only to detect and quantify trace amount of 33
 XAr, but also to provide useful information for predicting the toxicity or monitoring real-time 34
 degradation kinetics of XAr. These findings may have broad chemical, environmental and 35
 biological implications for future studies on halogenated aromatic persistent organic pollutants. 36
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82 Introduction

83 Halogenated aromatic compounds (XAr) have been widely used
 84 as pesticides, herbicides, wood preservatives, personal care
 85 agents, pharmaceuticals, flame retardants, and many other
 86 industrial products (Zhu and Shan, 2009; Zhu et al., 2011a; Dann
 87 and Hontela, 2010; De Wit, 2002). Most of these compounds are
 88 poorly biodegradable both in water and soil, which makes them
 89 persistent and widely distributed in the environment, and have
 90 earned them the name persistent organic pollutants (POPs)
 91 from UNEP (United Nations Environment Program) (Zhu and
 92 Shan, 2009; Zhu et al., 2011a; Dann and Hontela, 2010; De Wit,
 93 2002). The ubiquitous distribution of these recalcitrant and
 94 highly toxic XAr coupled with their carcinogenicity has raised
 95 public concerns on their potential risks to both human health
 96 and the ecosystem (Dann and Hontela, 2010; De Wit, 2002;
 97 Ramamoorthy, 1997; Fang et al., 2000; Zimbron and Reardon,
 98 2009; Lan et al., 2008; Gupta et al., 2002; Sorokin et al., 1995;
 99 Zhang and Huang, 2003; Zhong et al., 2012; Peller et al., 2003).
 100 Polyhalogenated phenols are an important class of XAr, and
 101 some of them, such as the widely used wood preservative
 102 pentachlorophenol (PCP) and 2,4,6-trichlorophenol (TCP), have
 103 been listed by the U.S. Environmental Protection Agency (EPA)
 104 as priority pollutants. Recently, PCP has been classified as a
 105 group I human carcinogen by the International Agency for
 106 Research on Cancer (IARC Working Group, 2016).

107 Among all the techniques used or tested so far, advanced
 108 oxidation processes (AOPs) have been increasingly favored as
 109 an “environmentally-friendly” technology for treating recalci-
 110 trant chlorinated phenols (CPs) and other XAr in the remedi-
 111 ation of contaminated water or soil (Von Sonntag, 2008; Wang
 112 and Xu, 2012; Pera-Titus et al., 2004). Several alternative
 113 techniques have been well established for the oxidation and

114 degradation of CPs and XAr, such as Fenton and Fenton-like
 115 oxidation (Zimbron and Reardon, 2009; Liou et al., 2004; Liao
 116 et al., 2007), UV-photolysis (Lente and Espenson, 2003; Chu,
 117 1999), and ozonation (Hong and Zeng, 2002), during which the
 118 recalcitrant XAr were degraded or even mineralized. In those
 119 “environmentally-green” AOP systems, which are based on
 120 hydrogen peroxide (Fenton and Fenton-like reactions) and
 121 ozone (O₃, O₃/H₂O₂), the most reactive radical intermediate
 122 formed is the hydroxyl radical (Von Sonntag, 2008).

123 Chemiluminescence (CL) is a phenomenon in which mole-
 124 cules in a chemically generated excited state liberate energy with
 125 light emission. CL frequently accompanies organic peroxide
 126 decomposition and free radical formation (Schuster, 1979;
 127 Matsumoto, 2004; Almeida de Oliveira et al., 2012; Widder, 2010;
 128 Adam et al., 2005). Since the CL intensity is governed by the rate
 129 of the chemical reaction, it can be used to quantify any analyte
 130 whose concentration is rate-determining (Grayeski, 1987). CL
 131 intensity-based analytical assays are inherently highly sensitive,
 132 rapid, and simple to operate, without requiring pre-treatment of
 133 samples. Therefore, they are being increasingly used as a
 134 sensitive analytical method in various research fields (Grayeski,
 135 1987; McCapra, 2000). For example, the reactive oxygen species
 136 (ROS) generated during UV-irradiation of nano-TiO₂ were
 137 selectively and sensitively detected by CL method (Wang et al.,
 138 2014); trace amount of transition metal ions such as ferrous(II)
 139 and cobalt(II) in estuarine and coastal waters could be quantita-
 140 tively determined by a pyrogallol-H₂O₂ CL system (Cannizzaro
 141 et al., 2000); and a good molecular imprinting CL sensor could be
 142 designed to recognize and separate target molecules when
 143 combining the molecular imprinting with high-sensitivity CL
 144 method (Lin and Yamada, 2000).

145 The hydroxyl radical ([•]OH) is an extremely reactive ROS
 146 which is important in chemistry, biology, toxicology, medicine,

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