

Methodology of Key Odorants Selection from Wastewater Treatment Plants Based on their Kinetics with Selected Atmospheric Oxidants

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Nowadays, wastewater treatment plants (WWTPs) are considered significant contributors to odour emissions, which can have a sensorineural impact on surrounding communities. In this study, calculations of the half-lives of selected odorants commonly found in wastewater treatment plants were carried out, considering varying concentrations of atmospheric oxidants that exhibit diurnal and geographical fluctuations. Odorants displaying quick reactivity and brief half-lives, which were deemed less significant contributors to odour nuisances, reacted with nitrate radicals (NO_3^*) during nighttime and chlorine atoms (Cl^*) with $T_{1/2}$ ranging from 1.23 to 2.77×10^5 minutes and from 6.57×10^{-4} to 3.14×10^3 minutes, respectively. On the other hand, extended half-lives were observed during reactions with ozone (O_3), ranging between 1.81×10^3 and 1.61×10^{11} minutes, suggesting a potentially smaller role in the degradability of odorants in the atmosphere. These insights into odorant-oxidant kinetics may aid in predicting atmospheric lifetimes and their contribution to secondary aerosol formation, thus informing regulatory and mitigation strategies to improve air quality.

1. Introduction

The nuisance caused by unpleasant odours is a significant concern from both a societal and industrial standpoint (Chaignaud et al., 2014). Nuisance due to odour generation by WWTPs, and animal production operations is one of the major sources of complaints of people living near these facilities. It also has triggered increased emphasis on controlling the impact of atmospheric pollutants on neighbouring areas (Rincón et al., 2019). Undesirable odours can elicit a range of emotional and adverse responses in individuals, spanning from irritation to documented health effects, ultimately resulting in a diminished quality of life (Blanes-Vidal, 2015). The odours from WWTPs emissions form an intricate mixture containing a diverse range of odorants derived from various chemical groups, including sulphur and nitrogen compounds, aldehydes, acids, alcohols, aromatics, ketones, terpenes, indole, and skatole (Barczak et al., 2021; Byliński et al., 2019). Only certain malodorous compounds are detected in significant concentrations.

The atmosphere possesses oxidizing capabilities, with key atmospheric daytime oxidant hydroxyl radical (OH). Additionally, oxidation processes can be initiated by NO_3 , O_3 , and Cl or Br atoms, and organic peroxy radicals (RO_2). These oxidants have the ability to transform initial air contaminants like carbon monoxide (CO), hydrocarbons (HCs), other organic compounds, nitrogen oxides (NO_x), and sulphur dioxide (SO_2) into carbon dioxide and some other products, rendering them more readily removable from the atmosphere (Chapleski et al., 2016). The understanding of odorant molecules kinetics in the presence of atmospheric oxidants is crucial for elucidating their fate and impact on air quality.

In the lower atmosphere known as the troposphere, hydroxyl radicals play a crucial role as the main oxidizer for both naturally occurring and man-made compounds, which contributes to the formation of ozone. Within the troposphere, hydroxyl radicals are generated when oxygen atoms resulting from the breakdown of ozone photolysis ($< 282 \text{ nm}$), interact with water vapor (Avallone, 2003). Therefore, OH radicals are formed, and active as an oxidant only during the daytime.

The aim of this paper is to assess the role of different oxidants active mainly in the time when OH concentration is very low. Ozone undergoes gas-phase reactions with alkenes, or it reacts heterogeneously with building materials or surface-sorbed alkenes like squalene or monoterpenes. Ozone is active as an oxidant during the day as well as night. Ozone also serves as the initiator and primary driver of indoor chemistry, predominantly entering indoor environments through outdoor-to-indoor transport. Indoor ozone concentrations commonly range from 20% to 70% of ambient values.

Nitrate radicals are generated through O_3 reacting with nitrogen dioxide (NO_2) to produce NO_3 and O_2 (Nazaroff & Cass, 1986). Following its formation, the oxidation facilitated by NO_3 is primarily notable during nighttime and plays a role in the initiation of the oxidation of many trace gases at night, especially in the boundary layer. Also, NO_3 has a rapid photolysis, characterized by a brief lifetime of approximately 5 seconds during the day. Due to the instability of NO_3 in daylight and the rapid reaction between NO_3 and NO , both N_2O_5 and NO_3 are commonly depleted during the day and increase during the night (Wang & Ruiz, 2017). In contrast, this lifetime is notably curtailed in the presence of NO , especially in a photochemical steady state with NO_2 , during daytime conditions. Nevertheless, studies have demonstrated that NO_3 can assume a pivotal role as a daytime oxidant within the canopy of forested environments.

The chlorine atoms are the exceptionally reactive radicals that could have a notable impact on the oxidative chemistry of the troposphere, especially over oceanic and coastal regions abundant in available chloride. The photochemical cycling of reactive chlorine species is a process through which their interactions and transformations occur. The chlorine atoms can be formed in marine air through multiphase photochemical mechanisms. The oxidation of chloride in marine aerosols by various species leads to the production of gas-phase species such as Cl_2 and $ClNO_2$. These compounds undergo photolysis during the daytime, releasing Cl atoms. In heavily polluted air, the generation of reactive chlorine is initiated by the reaction of N_2O_5 with aerosol chloride, as demonstrated in both laboratory and field observations. When gaseous N_2O_5 reacts heterogeneously with particulate chloride, it produces nitrile chloride ($ClNO_2$). In urban sites, Cl atoms are derived from particle-phase chloride in the presence of both NO_2 and ozone (O_3). This trend encourages the accumulation of $ClNO_2$ during nighttime. Following sunrise, $ClNO_2$ undergoes photolysis, leading to the liberation of chlorine.

Globally, oxidant concentrations vary depending on the surrounding environment, whether it be marine, urban, rural, and/or mountainous areas. Usually, ozone concentration is less in marine environment, medium in urban areas, and high in mountains, whereas chlorine is completely opposite. While for nitrate radicals are high in marine and mountains and less in urban areas as reported for oxidants in Table 2.

In the present era, there is a gap in knowledge about the kinetics of some odorants with atmospheric oxidants. The study focuses on calculating the half-lives of odorants commonly found in WWTPs, considering varying atmospheric oxidants. The understanding of odorant molecules' kinetic in the presence of atmospheric oxidants is important because odorants with extended half-lives suggest a potentially heightened role in causing odour nuisances, while those with quick reactivity and brief half-lives in the presence of air oxidants are deemed less significant contributors to odour nuisances.

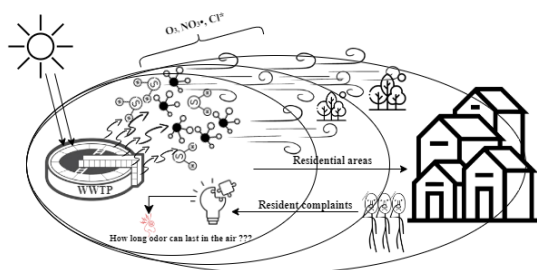


Figure 1: The schematic illustrates how the odour can be emitted from WWTPs and its effect on surrounding residential communities.

2. Methodology

2.1 Selection of Odorants

Targeted odorants listed in Table 1 have been identified in a previous work done by Barczak et al. (2022). Thus, based on the NIST Chemical Kinetics Database, these odorants have been assessed reaction rates and mechanisms of, spanning volatile organic compounds to sulfur-containing compounds, with atmospheric oxidants like ozone, nitrate radicals, and chloride atoms. In addition, depending on the reaction rate coefficients, (k), which have been extracted from the NIST Chemical Kinetics Database, and the oxidants concentration as

well as the odorants concentrations taken from the literature, it will allow to choose potentially important odorants by calculating their half-lives.

2.2 Sites description

In this study, we employed ongoing measurements of oxidants, expanding upon insights gleaned from prior research endeavors. To comprehensively assess oxidant concentrations, our data primarily originated from monitoring activities conducted at three specific locations: marine, urban, and areas characterized by mountainous or rural landscapes. This approach aimed to capture a diverse range of environmental conditions and contribute to a more nuanced understanding of oxidant dynamics for odorants across varied settings. Table 2 describes the different concentrations of the mentioned oxidants.

2.3 Half-Lives Calculations

This section utilizes the reaction information presented in Table 1 to construct models that explore the kinetics and reactions of the selected oxidants O₃, NO₃^{*}, and Cl^{*} atoms with the odorants. Based on the reactions of these odorants, which are Ammonia (NH₃), Trimethylamine (TMA), Carbonyl sulfide (COS), Methanethiol (MT), Carbonyl disulfide (CS₂), Ethanethiol (ET), Dimethyl sulfide (DMS), Dimethyl disulfide (DMDS), α-pinene (C₁₀H₁₆), Hydrogen sulfide (H₂S), p-Xylene (C₈H₁₀), Toluene (C₆H₅CH₃), 1,3,5-Trimethylbenzene (C₉H₁₂), Butanol (C₄H₉OH), with each oxidant, and using the previously determined reaction rate coefficients, at 298K. Consequently, the half-lives of the odorants computed by considering the concentrations of the oxidants [A] are constant and vary depending on diurnal and geographical fluctuations as shown in Table 2. The observed equation as reported by Atkins and De Paula (2006) is used for the targeted calculations (3). We obtain the equation for the half-life of a second-order reaction from the equation (1) as follows:

$$\left(\frac{[B]_0[A]_t}{[B]_t[A]_0}\right) = kt([A]_0 - [B]_0) \quad (1)$$

When the concentration of the odorant [B] is half of its initial:

$$[B]_t = \frac{[B]_0}{2} \quad (2)$$

Substituting this into the equation, we get:

$$T \frac{1}{2} = \frac{1}{k([A]_0 - [B]_0)} \ln \left(\frac{[A]_0[B]_0}{[A]_t \frac{[B]_0}{2}} \right) \quad (3)$$

Table 1 Selected odorants to be applied for kinetics determination.

Reactant	Reaction	k ₂₉₈ /cm ³ molecule ⁻¹ s ⁻¹
Ammonia	F [*]	F [*]
	NH ₃ + NO ₃ → HNO ₃ + ·NH ₂	5.99 X10 ⁻¹⁶
	NH ₃ + ·Cl → HCl + ·NH ₂	1.05 X10 ⁻¹³
Trimethylamine	(CH ₃) ₃ N + O ₃ → Products	7.84 X10 ⁻¹⁸
	F [*]	F [*]
	(CH ₃) ₃ N + ·Cl → HCl + CH ₂ N(CH ₃) ₂	4.83 X10 ⁻¹⁰
Carbonyl sulfide	F [*]	F [*]
	COS + NO ₃ → Products	1.00 X10 ⁻¹⁶
	COS + ·Cl → CO + SCl	1.00 X10 ⁻¹⁶
Methanethiol	F [*]	F [*]
	CH ₃ SH + NO ₃ → Products	9.20 X10 ⁻¹³
	CH ₃ SH + ·Cl → HCl + ·CH ₂ SH	4.30 X10 ⁻¹²
Carbonyl disulfide	F [*]	F [*]
	CS ₂ + NO ₃ → Products	4.00 X10 ⁻¹⁶
	CS ₂ + ·Cl → Products	4.00 X10 ⁻¹⁵
Ethanethiol	(CH ₃) ₂ S + O ₃ → Products	1.00 X10 ⁻¹⁸
	(CH ₃) ₂ S + NO ₃ → HNO ₃ + CH ₃ SCH ₂	1.10 X10 ⁻¹²
	(CH ₃) ₂ S + ·Cl → Products	3.30 X10 ⁻¹⁰
Dimethyl sulfide	(CH ₃) ₂ S + O ₃ → Products	1.00 X10 ⁻¹⁸
	(CH ₃) ₂ S + NO ₃ → HNO ₃ + CH ₃ SCH ₂	1.09 X10 ⁻¹²
	(CH ₃) ₂ S + ·Cl → Products	4.30 X10 ⁻¹⁰

Dimethyl disulfide	$((\text{CH}_3)_2\text{S} + \text{O}_3 \rightarrow \text{Products})$	F*
	$(\text{CH}_3)_2\text{S} + \text{NO}_3 \rightarrow \text{Products}$	7.00×10^{-13}
	$(\text{CH}_3)_2\text{S} + \cdot\text{Cl} \rightarrow \text{CH}_3\text{S}\cdot + \text{CH}_3\text{S}\cdot$	2.02×10^{-10}
Alpha pinene	$\text{alpha-pinene} + \text{O}_3 \rightarrow \text{Products}$	9.60×10^{-17}
	$\text{alpha-pinene} + \text{NO}_3 \rightarrow \text{Products}$	6.16×10^{-12}
	$\text{alpha-pinene} + \cdot\text{Cl} \rightarrow \text{Products}$	4.70×10^{-10}
Hydrogen sulfide	$\text{H}_2\text{S} + \text{O}_3 \rightarrow \text{SO}_2 + \text{H}_2\text{O}$	3.99×10^{-16}
	$\text{H}_2\text{S} + \text{NO}_3 \rightarrow \text{Products}$	1.00×10^{-15}
	$\text{H}_2\text{S} + \cdot\text{Cl} \rightarrow \text{HCl} + \text{SH}$	7.44×10^{-11}
p-xylene	$1,4\text{-Dimethylbenzene} + \text{O}_3 \rightarrow \text{Products}$	5.00×10^{-21}
	$1,4\text{-Dimethylbenzene} + \text{NO}_3 \rightarrow \text{Products}$	4.53×10^{-16}
	$1,4\text{-Dimethylbenzene} + \cdot\text{Cl} \rightarrow \text{Products}$	1.40×10^{-10}
Toluene	$\text{Toluene} + \text{O}_3 \rightarrow \text{Products}$	3.90×10^{-22}
	$\text{Toluene} + \text{NO}_3 \rightarrow \text{Products}$	6.79×10^{-17}
	$\text{Toluene} + \cdot\text{Cl} \rightarrow \text{Other Products} + \text{HCl}$	5.81×10^{-11}
1,3,5-trimethylbenzene	$1,3,5\text{-Trimethylbenzene} + \text{O}_3 \rightarrow \text{Products}$	F*
	$1,3,5\text{-Trimethylbenzene} + \text{NO}_3 \rightarrow \text{Products}$	8.00×10^{-16}
	$1,3,5\text{-Trimethylbenzene} + \cdot\text{Cl} \rightarrow \text{Products}$	Not- found
Butanol	F*	F*
	$\text{sec-C}_4\text{H}_9\text{OH} + \text{NO}_3 \rightarrow \text{Products}$	2.34×10^{-15}
	$\text{sec-C}_4\text{H}_9\text{OH} + \cdot\text{Cl} \rightarrow \text{CH}_3\text{CH}_2\text{C}\cdot(\text{OH})\text{CH}_3 + \text{HCl}$	2.57×10^{-9}

F* refers to that no data has been published in the NIST database.

Table 2 Observed oxidant concentrations in different tropospheric environments.

Oxidant	Marine environment (molecules/cm ³)	Urban environment (molecules/cm ³)	Mountain environment (molecules/cm ³)
Ozone	5.93×10^{11} (Ding et al., 2020)	10.6×10^{11} (Haslett et al., 2023)	1.31×10^{12} (Chevalier et al., 2007)
	24.3×10^{12} (Li et al., 2022)	2.16×10^{12} (Haslett et al., 2023)	0.00044×10^{12} (Yun et al., 2024)
Nitrate Radicals	31.7×10^3 (Lawler et al., 2011)	14.5×10^3 (Priestley et al., 2018)	-

3. Results and discussion

In this section, we present the findings from assessing how various odorants emitted from wastewater treatment plants interact with air oxidants (such as ozone, nitrate radicals, and chlorine atoms). The assessment considers differences in oxidant concentrations across the areas where measurements were taken. However, certain odorants were excluded from analysis due to the lack of their reaction rate coefficients (k_{298}) in the NIST database. As a result, it was not feasible to calculate their half-lives or estimate their persistence in the air as confirmed by (Ng et al., 2017). The excluded odorants due to lack of data published on the NIST database, which showed no reaction with O_3 , include ammonia, carbonyl sulfide, methanethiol, carbonyl disulfide, 1,3,5-trimethylbenzene, and butanol. Additionally, two other odorants were not shown any reaction with nitrate radicals and chlorine atoms, which are trimethylamine and 1,3,5-Trimethylbenzene, respectively. Furthermore, to align with the primary objective of our study, we focused on identifying odorants that potentially play a significant role in causing odor nuisances with a high half-life duration. The nitrate radicals during the night showed a fast decrease in odorants concentration (brief half-lives) compared to those reacted with ozone, while chlorine atoms were fluctuating depending on the geographical environment. Table 3 shows all odorants achieved the maximum range of half-lives calculations.

Table 3 List of odorants that achieved the maximum range of half-lives values.

Reactant	Environment	T $\frac{1}{2}$ O ₃ (min)	T $\frac{1}{2}$ NO ₃ [*] (min)	T $\frac{1}{2}$ Cl [*] (min)
Methanethiol	Marine site	N [*]	1.51	1.63
	Urban site	N [*]	5.43×10 ¹	1.52
	Mountain site	N [*]	1.42×10 ¹	N [*]
Ethanethiol	Marine site	6.13×10 ⁷	1.23	3.20×10 ⁻²
	Urban site	2.14×10 ⁹	2.38×10 ¹	2.97×10 ⁻²
	Mountain site	1.09×10 ⁸	1.79×10 ¹	N [*]
Trimethylamine	Marine site	1.04×10 ⁵	N [*]	6.57×10 ⁻⁴
	Urban site	1.07×10 ⁵	N [*]	6.11×10 ⁻⁴
	Mountain site	1.09×10 ⁵	N [*]	N [*]
Ammonia	Marine site	N [*]	1.86×10 ¹	3.70×10 ⁻²
	Urban site	N [*]	1.71×10 ¹	3.44×10 ⁻²
	Mountain site	N [*]	1.21×10 ¹	N [*]
Carbonyl sulfide	Marine site	N [*]	2.89×10 ⁴	3.14×10 ³
	Urban site	N [*]	8.81×10 ³	2.92×10 ³
	Mountain site	N [*]	5.85×10 ³	N [*]
Carbonyl disulfide	Marine site	N [*]	3.86×10 ³	7.13×10 ²
	Urban site	N [*]	4.24×10 ⁴	6.73×10 ²
	Mountain site	N [*]	1.33×10 ⁴	N [*]
Dimethyl sulfide	Marine site	1.77×10 ⁷	1.31	1.19×10 ⁻²
	Urban site	2.58×10 ⁷	1.85×10 ⁴	1.11×10 ⁻²
	Mountain site	2.34×10 ⁷	8.74	N [*]
Dimethyl disulfide	Marine site	N [*]	1.93	5.39×10 ⁻²
	Urban site	N [*]	3.63×10 ¹	5.00×10 ⁻²
	Mountain site	N [*]	2.90×10 ¹	N [*]
Alpha pinene	Marine site	1.01×10 ⁶	2.18×10 ⁻¹	2.67×10 ⁻²
	Urban site	1.64×10 ⁶	3.69	2.49×10 ⁻²
	Mountain site	6.92×10 ⁵	3.80	N [*]
Hydrogen sulfide	Marine site	1.81×10 ³	2.13×10 ³	3.80×10 ⁻³
	Urban site	1.87×10 ³	7.87×10 ²	3.53×10 ⁻³
	Mountain site	1.90×10 ³	5.26×10 ²	N [*]
p-xylene	Marine site	1.30×10 ¹⁰	2.88×10 ³	4.80×10 ⁻¹
	Urban site	6.34×10 ⁹	3.22×10 ⁴	4.46×10 ⁻¹
	Mountain site	4.99×10 ⁹	2.77×10 ⁵	N [*]
Toluene	Marine site	6.08×10 ¹⁰	2.06×10 ⁴	1.08×10 ⁻¹
	Urban site	1.03×10 ¹¹	1.08×10 ⁶	1.00×10 ⁻¹
	Mountain site	1.61×10 ¹¹	1.72×10 ⁵	N [*]
1,3,5-trimethylbenzene	Marine site	N [*]	1.66×10 ³	N [*]
	Urban site	N [*]	2.19×10 ⁴	N [*]
	Mountain site	N [*]	5.14×10 ⁴	N [*]
Butanol	Marine site	N [*]	5.57×10 ²	3.12×10 ⁻²
	Urban site	N [*]	6.15×10 ³	2.89×10 ⁻²
	Mountain site	N [*]	6.39×10 ⁴	N [*]

N^{*} refers to that no available data published for certain oxidants concentrations.

4. Conclusion

The evaluation indicates that the chosen odorants and VOCs emitted from the WWTPs exhibit prolonged persistence, lasting billions of seconds when exposed to air oxidants as reported in this study. Given the intricate nature of odor mixtures and perception, further in-depth studies are required to comprehensively understand the kinetics of these odorants. Understanding the kinetics of odorant-oxidant interactions is essential for predicting atmospheric lifetimes and their role in secondary aerosol formation, thereby guiding regulatory and mitigation efforts aimed at enhancing air quality.

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