



Influence of inorganic and organic compounds on the decay of peracetic acid in wastewater disinfection

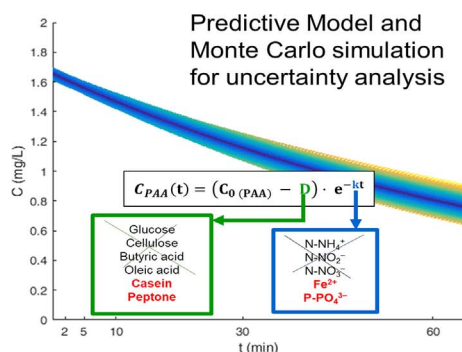
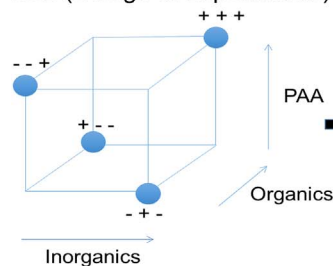


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GRAPHICAL ABSTRACT

Experiments based on DoE (Design of Experiments)



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ABSTRACT

The aim of this study was to evaluate the influence of the physical–chemical characteristics of wastewater on PAA decay, in multi-component solutions of inorganic and organic compounds (11 compounds in total) representative of secondary effluents of wastewater treatment plants, disinfected at various PAA concentrations (2–5 mg/L). Batch experiments were defined using the statistical method of the Design of Experiments (DoE) in order to evaluate the effect of each compound and their interaction on PAA decay. Results showed that the organics consumed immediately a considerable amount of PAA, independently from the initial PAA concentration, and consumption dropped rapidly to almost nil after 5 min, whereas PAA consumption due to the inorganics was slow, dependent on the initial PAA concentration and persisted until the end of the experiments (60 min). In detail, inorganics (such as reduced iron and orthophosphate) have shown to be the main drivers of the exponential decay: iron, particularly, has proved to directly consume PAA due to its catalysing capacity, whereas orthophosphate has shown to mainly interact with iron, acting as a chelating compound towards iron and consequently reducing the iron effect in consuming PAA. As for organics, proteins such as, casein and peptone, have been highlighted as the main cause of the initial PAA demand, probably due to the homolytic fission of PAA to generate peroxy and hydroxyl radicals, which are known to have a high reactivity towards proteins. Finally, a model for predicting the residual PAA concentration was obtained and validated; uncertainty analysis was also performed by a series of Monte Carlo simulations to propagate input uncertainties to the model output.

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1. Introduction

The ongoing population growth has often challenged the water supply systems in many areas of the world [1], leading to severe shortage of water. Consequently, the reuse of treated wastewater to reduce the gap between water demand and available water resources is drawing increasing attention in recent years [2]. In particular, reclaimed water from treated municipal effluents is increasingly used as an alternative source for a wide range of applications, such as irrigation of public areas and crops, industrial use and groundwater recharge [3]. Regardless of the type of wastewater reuse, a disinfection stage is required in wastewater treatment plants (WWTPs) to ensure a good microbiological quality of the discharged effluent [4]. Chlorine-based compounds are still the most widespread disinfectants even though many studies [5–7] reported that reactions between chlorine and organic matter lead to the formation of carcinogenic disinfection by-products in the treated effluents. For this reason, alternative disinfecting agents, such as peracetic acid (PAA), were investigated for wastewater treatment over the past years [8]. PAA is a strong oxidizing agent with an oxidation potential higher than chlorine, which determines its high antimicrobial activity [9]. PAA has proved to be effective against many microorganisms including bacteria, viruses, bacterial spores and protozoan cysts [10–13], guaranteeing no appreciable re-growth after disinfection [14]. In addition, no harmful disinfection by-products (DBPs) are formed during PAA treatment [15–18]. Concerning the application of PAA disinfection in WWTPs, initial PAA concentration and contact time were widely studied as the most relevant operating parameters for determining PAA disinfection efficacy against microorganisms. In particular, several works [12,19–21] showed that initial PAA concentrations from 1 to 15 mg_{PAA} L⁻¹ and contact times from 15 to 60 min can result in proper disinfection of primary, secondary and tertiary effluents. However, PAA decomposes rapidly in water solution and the rate of this process is highly affected by the water matrix composition [8,9,12,13,22,23]. A key step for guarantying a sufficient disinfectant residual concentration to reach bacterial inactivation targets, avoiding excessive residuals consists in the assessment of the influence of wastewater characteristics on PAA decay, in terms of easy-to-measure parameters, thus on the actual PAA dose. Currently, experimental works on this topic are either missing or were carried out as preliminary assessments. In more detail, past studies [24–26] reported that the presence of some inorganic compounds induces the catalytic decomposition of PAA, especially transition metals, such as Fe²⁺, Mn²⁺, Cu²⁺ and Co²⁺. Regarding salinity, the study of Liu et al. [22] revealed that it has a significant effect on PAA decay. In contrast, the same authors found that water hardness had only a slight impact on PAA degradation. On the other hand, to the best author's knowledge, no research work investigated the potential effect on PAA decay of many common inorganic ions, as inorganic nitrogen compounds or phosphorus compounds. The effect of pH on PAA disinfection and stability has been reported by various authors [8,9,13,27], who have observed that pH values above 9 lead to higher PAA decomposition rates. Indeed, given that the pK_a of PAA is 8.2, it is expected that at pH higher than this value the equilibrium will move toward the dissociated form (CH₃CO₃⁻). According to Yuan et al. [27], the hydrolysis of PAA to acetic acid is only slightly affected by pH ranging from 5.5 to 9.0. This was confirmed by Pedersen et al. [23] who found that PAA decay was comparable at pH adjusted between values of 6.4 and 8.6, which are typical values for wastewaters. As a consequence, pH exhibits a moderate effect on PAA decay in the usual range that this parameter assumes in conventional treatment conditions in WWTPs. As for organic compounds, the majority of previous works [12,22,28–30] focused on the effect of the organic content in the effluent through macro-indicators, as COD or DOC, highlighting a general trend according to which the higher is the organic content higher is the PAA degradation. However, the extent of PAA degradation is different among the studies available at similar COD/DOC concentrations. This observation can be

explained considering that organic compounds act differently on PAA decay depending on molecular characteristics, a major drawback of these macro-indicators is that they do not discriminate the individual effect of specific compounds, such as carbohydrates, lipids and proteins, in turn contained at different extent in WWTP effluents [31]. As for total suspended solids, although scarcely studied up to now, a significant PAA consumption has been outlined [12,19,20,30,32], but it has to be pointed out that most of the organic matter (around 86%) present in secondary effluents is expected to be in the soluble fraction [31,33]. Moreover, most of the past research works focused on investigating the effect of one factor at a time, without considering the occurrence of interaction effects between two or more factors.

The purpose of this study is firstly to identify which inorganic and organic compounds commonly present in the soluble fraction of wastewater can individually and/or jointly affect PAA decay, and secondly the development of a reliable mathematical model to predict residual PAA concentration over time as a function of the constituents of the WWTP effluent. In detail, eleven compounds were evidenced as potentially relevant for PAA decay based on a preliminary literature review as well as on the expertise of experimenters. Inorganic compounds were ammonia nitrogen (NH⁴⁺), nitrate (NO³⁻), nitrite (NO²⁻), reduced iron (Fe²⁺) and orthophosphate (PO₄³⁻), whereas organic compounds were surrogates of carbohydrates (glucose and cellulose), lipids (butyric and oleic acids) and proteins (peptone and casein). Experiments were divided in two experimental plans, one for inorganics and one for organics, which were defined using the statistical method of the Design of Experiments (DoE). This methodology allowed to evaluate the effect exerted by each compound and to evidence the occurrence of interaction effects. After the development of the model and its validation by a set of experiments in which inorganics and organics displaying a significant effect were simultaneously present, an uncertainty analysis was performed by Monte Carlo simulations to propagate the input uncertainties to model output.

2. Materials and methods

2.1. Reagents and experimental setup

PAA technical grade solution (VigorOx® WWTII) was supplied by PeroxyChem (USA), whose composition as weight percentage is 15% of peracetic acid, 23% of hydrogen peroxide and 16% of acetic acid. The PAA concentration in the commercial solution was checked monthly by iodometric titration [34]. All chemicals were reagent grade purchased from Sigma Aldrich (USA), except for DPD (N,N-diethyl-p-phenylenediaminesulphate) salt that was purchased from Hach Lange (USA). All the stock solutions for the inorganic compounds were prepared according to Standard Methods [35]. The stock solutions containing the organic compounds or their surrogates, were prepared to have a concentration 1000 mg L⁻¹ in terms of COD for each compound. The calculations required to prepare the stock solutions were based on the Theoretical Oxygen Demand (ThOD) values of each organic compound [36].

1-h PAA decay tests were performed in completely mixed batch reactors (1 L glass beakers) mixed by magnetic stirrer in dark conditions at room temperature (20 ± 1 °C). In each beaker, a given aliquot of the stock solutions, according to the experimental plan described in Sections 2.3 and 2.4, was added to deionized water and the pH of the solutions was adjusted to 7.5 at the beginning of the tests with sodium hydroxide (NaOH) or sulphuric acid (H₂SO₄) and monitored during the tests. After pH adjustment, a selected aliquot of PAA stock solution was added to obtain the given PAA initial concentration. Samples were collected at five contact times (2, 5, 10, 30 and 60 min). Preliminary tests were performed in deionized water to estimate PAA decay in absence of inorganic and organic compounds (blank tests). Experiments were performed in triplicate for three initial PAA concentrations (2, 5 and 10 mg_{PAA} L⁻¹).

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