



# Contribution of filamentous fungi to the musty odorant 2,4,6-trichloroanisole in water supply reservoirs and associated drinking water treatment plants



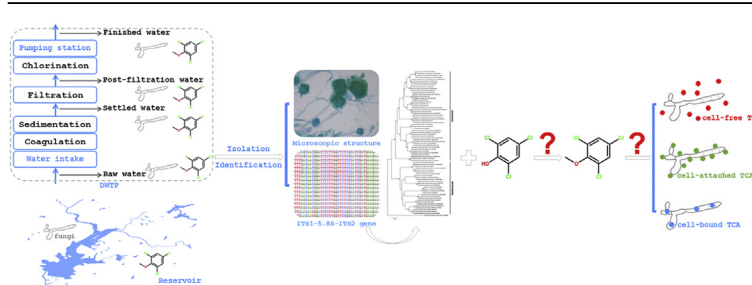
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## HIGHLIGHTS

- Filamentous fungi were the major contributors to 2,4,6-TCA formation in drinking water.
- Eleven fungal species isolated have the capability to convert 2,4,6-TCP to 2,4,6-TCA.
- 2,4,6-TCA was mainly distributed in extracellular environment of TCA-forming fungi.
- A possible metabolic pathway of 2,4,6-TCP in TCA-forming fungi was proposed.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this study, the distribution of 2,4,6-trichloroanisole (2,4,6-TCA) in two water supply reservoirs and four associated drinking water treatment plants (DWTPs) were investigated. The 2,4,6-TCA concentrations were in the range of 1.53–2.36 ng L<sup>-1</sup> in water supply reservoirs and 0.76–6.58 ng L<sup>-1</sup> at DWTPs. To determine the contribution of filamentous fungi to 2,4,6-TCA in a full-scale treatment process, the concentrations of 2,4,6-TCA in raw water, settled water, post-filtration water, and finished water were measured. The results showed that 2,4,6-TCA levels continuously increased until chlorination, suggesting that 2,4,6-TCA could form without a chlorination reaction and fungi might be the major contributor to the 2,4,6-TCA formation. Meanwhile, twenty-nine fungal strains were isolated and identified by morphological and molecular biological methods. Of the seventeen isolated fungal species, eleven showed the capability to convert 2,4,6-trichlorophenol (2,4,6-TCP) to 2,4,6-TCA. The highest level of 2,4,6-TCA formation was carried out by *Aspergillus versicolor* voucher BJ1-3: 40.5% of the original 2,4,6-TCP was converted to 2,4,6-TCA. There was a significant variation in the capability of different species to generate 2,4,6-TCA. The results from the proportions of cell-free, cell-attached, and cell-bound 2,4,6-TCA suggested that 2,4,6-TCA generated by fungi was mainly distributed in their extracellular environment. In addition to 2,4,6-TCA, five putative volatile by-products were also identified by gas chromatography and mass spectrometry. These findings increase our understanding on the mechanisms involved in the formation of 2,4,6-TCA and provide insights into managing and controlling 2,4,6-TCA-related problems in drinking water.

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## 1. Introduction

The increasing occurrence of taste and odor compounds in drinking water is considered an important water quality problem

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throughout the world (Antonopoulou et al., 2014). The earthy and musty odor is the most troublesome subgroup, which has been reported by many researchers (Ridal et al., 2001; Zamyadi et al., 2015; Liu et al., 2016). Generally, geosmin, 2-methylisoborneol (2-MIB), 2-isopropyl-3-methoxy pyrazine, 2-isobutyl-3-methoxy pyrazine, and 2,4,6-trichloroanisole (2,4,6-TCA) are responsible for the earthy and musty odor in drinking water. All five odorous compounds have microbial sources (cyanobacteria, actinomycetes, myxobacteria, and fungi) (Zhang et al., 2010; Callejón et al., 2016). Significant advances have been made in the understanding of the fundamental mechanisms responsible for the biosynthesis of geosmin and 2-MIB (Su et al., 2015). Unfortunately, the formation of 2,4,6-TCA in drinking water has not been systematically investigated.

The earthy and musty odor in water supply reservoirs and drinking water treatment plants (DWTPs) has recently received increasing attention. Both 2-MIB and geosmin had high detection rates in source water (Bai et al., 2017; Olsen et al., 2016). However, 2,4,6-TCA exhibited a low detection rate in source water and little attention was paid to the 2,4,6-TCA problem at DWTPs. Zhang et al. (2016a) first reported the occurrence of haloanisoles in tap water sampled from 22 locations across China. The concentrations of 2,4,6-TCA were greater than zero in all tap water samples. The absence in source water and wide occurrence in tap water indicated that 2,4,6-TCA was mainly generated in drinking water treatment plants and/or distribution systems; however, its formation mechanisms have not yet been clarified. Simultaneously, the elimination efficiency of 2,4,6-TCA in a conventional water treatment process (coagulation, sedimentation, filtration, and chlorination) has also remained unclear. These two issues are key to effective management of 2,4,6-TCA problems at DWTPs.

2,4,6-Trichloroanisole is known for causing cork taint in wines (Buser et al., 1982). Subsequently, a number of studies suggested that 2,4,6-TCA was formed *via* the methylation of 2,4,6-trichlorophenol (2,4,6-TCP) with some fungi in wines, food-packaging materials, rice-koji, and coffee (Whitfield et al., 1991; Álvarez-Rodríguez et al., 2002; Miki et al., 2005; Iamanaka et al., 2014). Several isolates such as *Paecilomyces variotii*, *Trichoderma longibrachiatum*, and *Aspergillus oryzae* were shown to have the TCA-forming potential. In drinking water, chlorine is widely used for its chemical disinfection properties. Previous studies showed that 2,4,6-TCA was identified in drinking water after chlorination (Corbi et al., 2007; Peter and von Gunten, 2009). Recently, Zhang et al. (2016b) reported that 2,4,6-TCA could be produced by the chlorination of anisole with chlorine under acidic conditions (pH = 4). In fact, the pH of water is adjusted to 7.5–8.0 throughout the water treatment process (sodium hypochlorite as a disinfectant). Since anisole is not ionizable at a pH greater than 6.4 (Sivey et al., 2015), it is surmised that the chlorination of anisole with chlorine is hard to achieve at DWTPs. On the other hand, 2,4,6-TCP is an environmental pollutant found in surface water, sediment, soil, and atmosphere (Campoy et al., 2009). At DWTPs, 2,4,6-TCP can be formed from dissolved organic matters present in chlorinated water (Michalowicz et al., 2011). 2,4,6-TCP is also an aromatic halogenated disinfection byproduct that has been identified in chlorinated drinking water (Pan and Zhang, 2013) and chlorinated wastewater effluents (Yang and Zhang, 2013). Consequently, the methylation of 2,4,6-TCP with fungi may be a major biosynthetic route in drinking water treatment. Fungi as the provider of methyltransferase are heterotrophs. They acquire food by absorbing dissolved molecules, typically by secreting digestive enzymes into their environment. However, there is a general lack of knowledge about how the fungi are involved in the methylation of 2,4,6-TCP. It is not known if the 2,4,6-TCP is absorbed into fungal cells to be methylated. The methylation capability of 2,4,6-TCP among and

within different taxa should be elucidated. Moreover, to minimize the occurrence of 2,4,6-TCA in drinking water, the formation mechanisms of 2,4,6-TCA should be further studied.

The purpose of this study was to investigate the distribution of 2,4,6-TCA in water supply reservoirs and associated DWTPs. For the *in-situ* investigation, the potentially TCA-forming fungi were isolated from the conventional treatment processes (coagulation, sedimentation, filtration, and chlorination) and identified through microscopic observations combined with the 16S rDNA analysis using a fungi-specific primer (White et al., 1990). A comprehensive analysis of the capability of the isolated fungi for the methylation of 2,4,6-TCP was performed in laboratory experiments. Putative volatile by-products were also identified by gas chromatography and mass spectrometry (GC-MS). This study provides a better understanding of the formation mechanisms of 2,4,6-TCA in drinking water and useful information for a more efficient management of DWTPs experiencing 2,4,6-TCA-related problems.

## 2. Materials and methods

### 2.1. Study sites and sampling

The investigation was performed in Shenzhen Reservoir (R1), Xikeng Reservoir (R2), and four associated DWTPs (DWTP1, DWTP2, DWTP3, and DWTP4) in Guangdong Province, China. The two water supply reservoirs have total storage capacities of 44.96 and 19.82 million cubic meters, respectively. Both of them are the main water sources and drinking water storage reservoirs. R1 provides source water for DWTP1, DWTP2, and DWTP3, while R2 provides source water for DWTP4. DWTP4 is a large-scale drinking water treatment plant with a production rate of 195,600 m<sup>3</sup> d<sup>-1</sup>. There are two independent projects (Project I and Project II) at DWTP4, started in September 2006 and February 2014, respectively. The characteristics of the treatment processes used at DWTP4 are shown in Table S1 (Supplementary Information). Water samples from the two water supply reservoirs were taken from the water intake(s) and the center area in October and December 2015. Surface water samples for the 2,4,6-TCA analysis were collected in brown glass bottles without headspace from a depth of 0.5 m, then stored at 4 °C, and analyzed as soon as possible. Water samples from the DWTP4 were collected from the sampling points during different stages of the conventional treatment processes, namely: (1) raw water, (2) settled water, (3) post-filtration water, and (4) finished water. Sampling was performed four times a month. Water samples from the other three DWTPs were collected from finished water and sampling was performed once a month. These samples were collected from the taps of each sampling point and water was allowed to run for about 20 min before collection. Additional sampling procedures were applied to reservoir water samples.

### 2.2. Chemicals and reagents

The standards 2,4,6-TCA (1 mg mL<sup>-1</sup> in methanol) and acenaphthene-d10 (4 mg mL<sup>-1</sup> in methanol) were from AccuStandard (New Haven, USA). Acenaphthene-d10 was used as the internal standard. Methanol (HPLC grade) was from Sigma-Aldrich (Guangzhou, China). NaCl (analytical grade) provided by Sino-pharm Chemical Reagent Co., Ltd (Shanghai, China) was heated and dried at 450 °C for 4 h in a Muffle furnace (SX-5-12, Beijing, China). The fungal Czapek-Dox Agar and Martin Agar Medium, Modified were from Guangdong Huankai Microbial Sci. & Tech. Co., Ltd (Guangzhou, China). Pure water was generated by a Milli-Q laboratory water purification system (Direct-Q 3, Millipore SAS, Molsheim, France).

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