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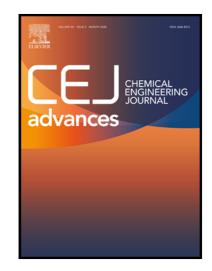
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Highlights

- Perspective from a wide interdisciplinary international network of experts in water T&O.
- Gaps and prospects in sensory and chemical analysis, risk assessment and management, water treatment.
- Improve capabilities of water supplies to solve water T&O incidents.

boundance

Perspective

Water taste and odor (T&O): challenges, gaps and solutions from a perspective of the WaterTOP network

Reyhan Akcaalan¹, Ricard Devesa-Garriga², Andrea Dietrich³, Martin Steinhaus⁴, Andreas Dunkel⁴, Veronika Mall⁴, Maura Manganelli⁵, Simona Scardala⁵, Emanuela Testai⁵, Geoffrey A. Codd⁶, Frantisek Kozisek⁷, Maria Antonopoulou⁸, Ana Rita Lado Ribeiro⁹, Maria José Sampaio⁹, Anastasia Hiskia¹⁰, Theodoros M. Triantis¹⁰, Dionysios D. Dionysiou¹¹, Gianluca Li Puma¹², Linda Lawton¹³, Christine Edwards¹³, Henrik Rasmus Andersen¹⁴, Despo Fatta-Kassinos¹⁵, Popi Karaolia¹⁵, Audrey Combès¹⁶, Kristel Panksep¹⁷, Sevasti-Kiriaki Zervou¹⁰, Meriç Albay¹, Latife Köker¹, Ekaterina Chernova¹⁸, Sofia Iliakopoulou^{8,10}, Elisabeth Varga¹⁹, Petra M. Visser²⁰, Angelika Ioanna Gialleli²¹, Zuhal Zengin¹, Nikos Deftereos²², Phani Miskaki²², Christophoros Christophoridis¹⁰, Aikaterina Paraskevopoulou¹⁰, Tsair-Fuh Lin²³, Arash Zamyadi^{24,25}, Galina Dimova-Boykinova²⁶, Triantafyllos Kaloudis^{10,21}*

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Abstract

Aesthetic aspects of drinking water, such as Taste and Odor (T&O), have significant effects on consumer perceptions and acceptability. Solving unpleasant water T&O episodes in water supplies is challenging, since it requires expertise and know-how in diagnosis, evaluation of impacts and implementation of control measures. We present gaps, challenges and perspectives to advance water T&O science and technology, by identifying key areas in sensory and chemical analysis, risk assessment and water treatment, as articulated by WaterTOP (COST Action CA18225), an interdisciplinary European and international network of researchers, experts, and stakeholders.

Keywords: water quality, water treatment, taste, odor

1. Introduction

Would you drink a glass of water that has an unusual smell? Probably not. But what about a slight chlorine smell? In many drinking water supplies a chlorinous odor can be accepted by consumers, because they are aware that water is chlorinated to protect public health from dangerous pathogens. There are other common and periodic tastes and odors (T&O), such as "musty" or "earthy" which are aesthetic but not health problems in water supplies. Earthy/musty odors result in varying levels of acceptance depending on prior exposure of the consumers or the level of proactive communications about the odor issue by the water supplier. Sometimes, massive consumer complaints about water T&O may arise from a change of the water source or an unusual contamination event, which alters the sensory character of water; consumers largely perceive such alterations as signs of degraded quality.

The answer to the "to drink or not to drink" question may not be obvious, since a "global" water quality assessment needs to encompass two seemingly "orthogonal" (independent) dimensions: a) evaluation of the water safety by microbiological and physicochemical laboratory testing, and b) sensory evaluation by untrained consumers or expert panels. Laboratory testing assesses the suitability of the water to be distributed, using a public health risk approach, while sensory perceptions largely determine the acceptability by consumers, a critical constraint that cannot be ignored by water suppliers. A simplified illustration of the combined safety and sensory assessment of water is presented in Figure 1. While decision making in cases (B) and (D) is straightforward, since laboratory testing is aligned to consumers' perceptions, cases (A) and (C) are challenging, as sensory evaluation may not be supportive of laboratory results. In particular, where consumers sense an unpleasant T&O, but the source cannot be unambiguously identified (case C), the tasks required in terms of laboratory analysis, risk evaluation and communication to consumers can overwhelm the capabilities and capacities of the water suppliers. However, such cases may also offer new opportunities to detect new or unknown T&O chemicals, identify their sources, evaluate and, if needed, mitigate the associated risks, aiming to better safeguard the quality of the distributed water.

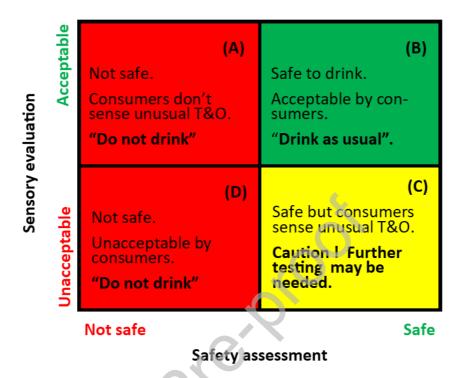


Figure 1: Simplified illustration of safety and sensory (T&O) assessment of drinking water

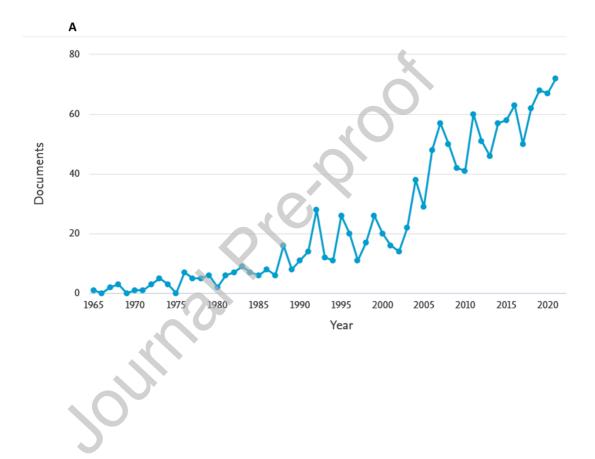
Unpleasant water T&O can be caused by a wide range of chemicals including organic compounds, minerals, and metals. Common sources of T&O are industrial pollution, aquatic microbial and plant metabolism, biotic and abiotic chemical transformations in natural waters or during water treatment processes, and migration from materials in contact with the water. A plethora of anthropogenic and natural organic compounds that present some volatility can bind to human olfactory receptors, triggering unpleasant odors. For example, the occurrence and growth of T&O producing cyanobacteria and algae, which are particularly relevant to surface water reservoirs, are increasing due to climate change and eutrophication. The response of the human olfactory system to organic compounds varies widely and this is reflected in the broad range of odor threshold concentrations that may differ by orders of magnitude among various compounds. Therefore, only a small number of organics found in water at relevant concentrations can be sensed by consumers, while the ability of humans to detect an odor in water varies among individuals and depends on the physical

condition and other parameters. However, some common T&O, such as the widely encountered earthy/musty geosmin and 2-methylisoborneol (MIB), have odor thresholds at the low-ng/L levels, which challenge the detection capabilities of chemical analysis. The need to remove traces of T&O that cause unpleasant odors is a common issue for water suppliers, since the treatment processes are generally non-selective and natural organic matter (NOM), usually present at 3 - 6 orders of magnitude higher concentrations (mg/L levels) competes with the T&O species. Integrating T&O into risk assessment and management in the framework of EU mandated Water Safety Plans (EU Drinking Water Directive 2020/2184) is complicated due to gaps of knowledge about T&O human bioactivity, possible public health and ecosystem impacts and prevention-mitigation strategies.

Drinking water treatment and supply has many challenges, starting with naturally varying source water qualities due to regional geology, hydrology, ecology, and seasonal factors such as temperature, drought, and flood. Engineered water treatment is designed to accommodate these varying factors with the aim of producing a consistent drinking water quality. Despite the increasing research on water T&O, knowledge and expertise regarding the management of these problems remain largely scattered and fragmented, especially within water supply organizations, where practices and know-how developed by several groups are not efficiently disseminated, they are discontinued, or they are regionally specific but not generally adaptable. Figure 2 illustrates the increase in the number of research publications and the global authorship networks for a common water T&O, geosmin, showing that the research field is generally dominated by a few countries and research groups.

Diagnostic methods of sensory testing by expert panels and non-targeted chemical analysis of T&O are currently not harmonized among water supplies and water quality laboratories. In addition, the water sector has not taken much advantage of the state-of-the art sensory and instrumental analysis techniques that are widely used in the food and cosmetics sectors. The development of guidelines for risk assessment and management of T&O is impeded by the need to involve expertise from a variety of disciplines, including environmental and analytical chemistry, sensory science, aquatic ecology, toxicology, and water engineering. A particularly important topic that needs to be further explored is the potential of using the detection of T&O as an early indicator of other associated, potential water quality problems.

To fill the above gaps, international cross-discipline and cross-sector networks are needed to share research results and expertise and to train the next generation of water quality researchers and practitioners. Here we present challenges and future perspectives for the advancement of water T&O science and technology, focusing on sensory and chemical analysis, risk assessment/management, and water treatment, as articulated by WaterTOP (COST Action CA18225), a current pan-European and international network of researchers, experts, and stakeholders (WaterTOP, 2019).



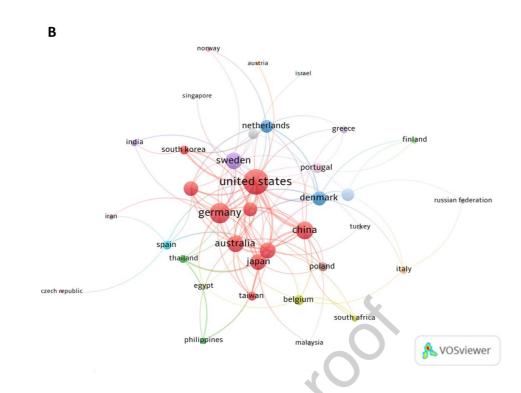


Figure 2: (A) Number of publications and (B) authorship networks for publications on geosmin (title, abstract, keywords). Data source: Scopus. Network Visualization: VOSviewer.

2.Taste and odor (T&O), and sensory analysis in waters

2.1 Taste and odor

Water companies are continuously addressing the need to improve water quality, safety, and treatment processes for their consumers. For the past few decades, this has been most notably accomplished by implementing membrane technologies (Devesa & Dietrich, 2018; Garcia et al., 2015; Vingerhoeds et al., 2016). However, consumers are not usually aware of the complexity of the drinking potabilization process and they primarily value tap water according to its organoleptic properties. Consumer expectations are that the water must be colorless and clear, and the perception of its quality depends fundamentally on its flavor, where flavor is a combination of T&O.

It is well known that water taste depends fundamentally on its mineral composition, i.e., the total dissolved solids (TDS) and their relative distribution as cation and anion species and their concentrations (Devesa and Dietrich, 2018). Disinfectants and other

chemicals added during treatment, water-aging, and changes in the distribution system also influence taste, odor, and flavor. In addition, water may contain numerous other organic and inorganic compounds, both of natural or anthropogenic origin, which, although at low concentrations, can provide the water with characteristic (usually undesirable) odors, tastes, flavors, or tactile sensations such as astringency or dryness.

2.1.1 Influence of TDS on the taste of water

Total dissolved solids (TDS) constitute the most common aggregate parameter of the mineralization level and taste-quality of water. International regulations and recommendations establish significantly different maximum levels for TDS: 500 mg/L in USA and Canada; 1000 mg/L according to World Health Organization (WHO) Guidelines, and 1600 mg/L in the European Community (corresponding to the established 2500 μ S/cm conductivity at 25 °C). Several studies show that consumers do not like high levels of minerals in their tap water (Platikanov et al., 2013; Vingerhoeds et al. 2016) and may have a negative perception of water quality in the range of 500-1600 mg/L TDS. A recent article provides guidance about how increases and decreases of TDS affect consumers' perception and human sensitivity to detect these changes (Devesa and Dietrich 2018).

2.1.2 Role of specific species

Waters with a very low mineral content are perceived as sweet, bitter, or rough depending on personal perception. The effect of the dissolved species changes depending on concentration, and the interaction among species introduces a great complexity to the subject. In general, calcium, magnesium and bicarbonate are considered positive for taste; whereas sodium, potassium and chloride have a negative impact (Platikanov et al., 2013; Vingerhoeds et al. 2016). Sulfate is regulated from aesthetic and health perspective because of its salty or gypsum-like taste at high concentrations or laxative effects in combination with magnesium, but a positive effect has also been suggested at low to medium concentrations (López et al. 2017).

2.1.3 Improvement in the taste of tap water due to membrane use

Membrane techniques are characterized by greatly reducing TDS and altering the proportion of the anions and cations. The resulting water requires remineralization to mitigate its aggressiveness to metallic distribution materials. This is usually

accomplished by adding calcium and magnesium salts, and sometimes carbon dioxide. The remineralization process further provides two advantages: the water is safer for human consumption (alkaline-earth salts are considered beneficial for many physiological processes) and the taste is improved. Several works have shown the improvement of the taste of water on the network by using membranes and a proper blending of resources (García et al. 2015).

2.1.4 Taste-and-odor events

Malodorous water and abnormal changes on T&O are frequently associated with an unsafe product by consumers. Geosmin and MIB are by far the most common compounds causing T&O episodes in source and drinking waters across the globe (Devi et al. 2020). It should be noted that these two metabolites are produced by cyanobacteria, algae, and actinomycetes (Watson and Jüttner 2019) naturally present in the environment and have a tremendous negative economic impact on the drinking water industry, and on aquaculture and recreational waters.

Other reported episodes are totally anthropogenic. A severe crisis in the Elk River (West Virginia, USA) occurred due to a spill of crude 4-methylcyclohexane-methanol (4-MCHM), a product of the coal mining industry, which affected thousands of citizens and was declared as a national emergency. The *trans* isomer of this compound presents a sweet-licorice odor and represents an excellent example of isomers with a huge difference of odor intensity; indeed, the cis-4-MCHM has approximately a 2000-fold higher odor threshold concentration with different odor descriptors (fermented fruit, mint-like) (Gallagher et al. 2015). Chemical contamination events caused by dioxanes, a family of compounds with a long history of odor episodes due to intensive industrial use, have also been recently reported (Carrera et al. 2019). Besides industrial chemicals, anthropogenic compounds from municipal wastewater, such as indole, also can cause odor issues in tap water (Wang et al. 2019).

2.2 Sensory Test Methods

Good aesthetic water quality and consumer satisfaction are critical to the success and value of the drinking water industry. Sensory analysis is essential for good water quality, either by helping to identify issues and determine treatments for source water

or to assure consumer confidence and satisfaction of the drinking water. Sensory methods, mostly adapted from the food science industry, are widely available to monitor water quality. These methods have been described in detail (Dietrich and Ömür-Özbek 2019) and are briefly summarized below. Except for the checklist methods, the others have been used to varying degrees in the water industry for decades.

Discrimination Methods: These compare two or more samples to determine which sample has more (or less) of a specific attribute, e.g., chlorinous or earthy/musty/moldy odors, or salty taste. Common methods are a paired comparison, triangle test, 1-out-of-five test, and ranking test.

Threshold Methods: Taste threshold concentrations (TTC) and odor threshold concentrations (OTC) represent the concentrations at which 50% of the population can detect a taste or an odor. The approach provides a single value although it is important to recognize that thresholds are better represented by a range of values, as many consumers detect tastes or odors above and below the TTC or OTC, respectively. A typical threshold method uses triangle tests and a series of 8 concentrations of the pure chemical. Another method is the Threshold Odor Number (TON) (APHA 2017; Standard Method 2150 B), which is based on the serial dilution of a water sample with odor-free water until any odor is no longer detected.

Descriptive Methods: Flavor Profile Analysis (FPA) (APHA 2017; Standard Method 2170) is the gold standard for describing all possible tastes and odors in a product. FPA requires extensive training and practice. A simpler and more direct approach is the Quantitative Descriptive Analysis (QDA), where a limited number of tastes and odors associated with specific products are the focus of the training and rating. The food and beverage industries are moving toward QDA, and away from FPA, as QDA requires less training. For consumer-based sensory studies, the Check-All-That-Apply or Check-If-Apply approaches involve providing consumers with a checklist of 10-40 attributes based on those best describing the product, as well as an "other" category (Carneiro et al. 2021). By providing descriptors, the checklist method is faster than FPA/QDA and allows consumers to provide relevant information to industry.

Intensity Methods: These methods assess the strength, or intensity of the overall taste or odor of water. FPA does this for individual tastants and/or odorants

using a 0-12 scale: 0 (no taste or odor); 4 (weak); 8 (moderate); 12 (strong). The Total Intensity of Odor (APHA 2017; Standard Method 2150 C) assesses the overall odor of a water sample without identifying descriptors. Smelling a series of hexanal standards is used to define the scale applied to determine the odor intensity of the water sample. The Threshold Odor Number (TON) (European Standard EN 1622; APHA 2017; Standard Method 2150) measures the amount of dilution with odor free water required to produce a drinking water sample with no perceivable odor. The endpoint is no perceivable odor and descriptors are not used.

Hedonic Methods: Hedonic methods assess consumer satisfaction. The methods use a nine-, seven-, or five-point scale with extreme dislike and extreme like at either ends of the scale and neither like, nor dislike, in the middle to rate the overall liking of the water.

Assessing Consumer Feedback: Consumers provide feedback to their water suppliers in the form of complaints and compliments. Data analytics are applied to assess feedback and identify trends and issues that consumers identify (Gallagher and Dietrich, 2014; Dietrich et al. 2014).

2.3 Gaps and Recommendations

The global drinking water industry requires more focus on aesthetic water quality, routine implementation of sensory methods, and routine data analytics of consumer feedback. Increased demand for potable water for a growing global population is occurring at a time of increased challenges to source water quality due to climate change, salinization of freshwaters, algal and cyanobacterial blooms, floods, droughts, and other factors. Sensory monitoring of source and finished water quality can assist in the early identification of problems, thus implementing appropriate and timely treatment, and maintaining consumer satisfaction.

Many gaps exist in the knowledge of which chemicals cause tastes and odors. Performing research to connect specific chemicals with TTC or OTC and their associated sensory descriptors is required (Burlingame and Doty, 2018). Substantial differences occur in the literature for threshold values because of different test conditions: sensory method and statistical approach, type of tasters (trained or untrained), temperature in case of odor evaluation (in general, room temperature, 25 or 45°C), physicochemical properties of the water where the stimulus is dissolved, and

other experimental considerations. Some degree of coordination and standardization would benefit the water industry. Also, the occurrence of geometric isomers has to be taken into account, because studies have shown that stereoisomers and enantiomers can present hugely different organoleptic properties (Bentley 2006; Piriou et al. 2009; Gallagher et al. 2015).

The situation in real drinking water samples is extremely complex due to interactions (additive, synergistic or antagonistic) between mineral species, odorants, but also considering the disinfectant agents and the organic compounds (natural or anthropogenic) at trace levels. More research is needed on this subject.

The FPA method is continuously enhanced thanks to the T&O Wheel (TOW) that is periodically revised with new compounds and descriptors (Suffet et al. 2019). From the quantitative point of view, the scale of intensities is poorly defined and only a few calibration standards are described. Therefore, results from different countries, even regions, cannot be readily compared. The implementation of international intercomparison exercises would be a useful tool to improve this issue.

Consumers are always present throughout the drinking water distribution network to monitor water quality. Establishing a dialogue with consumers about water quality and harnessing positive and negative consumer feedback through social media and on-line tools, such as Check-If-Apply lists, can thus provide the water industry with valuable data. Consumers can be part of a monitoring network, with a high degree of granularity. However, at present, consumer feedback data are not usually tracked or analyzed with the same rigor as regulatory data or waterflow/main break data, even though consumers judge the aesthetic quality of their drinking water daily and may only assess the regulatory reports occasionally. The water industry to effectively track consumer data, should broaden their understanding of how consumers describe tap water tastes and odors (Dietrich and Burlingame 2020) and how consumers respond to water quality changes (Devesa and Dietrich 2018).

An easily accessible and searchable global repository of sensory properties of known water-related T&O compounds and acknowledged treatment strategies, will aid the water industry in providing safe and palatable water to consumers across the globe.

3. Chemical analysis of water T&O

Diagnosis of the causes of water T&O incidents is an essential requirement to initiate timely and effective control and management strategies. Expert sensory panels can confirm the presence of undesirable T&O, describing their characteristics, providing clues about possible sources and narrowing the focus of investigation on specific sectors of the TOW. However, the sensory description of water T&O generally corresponds to the occurrence of several different compounds, possibly of various origins, their analysis being further complicated when multiple T&O are present. Consequently, unambiguous detection and identification by chemical analysis is fundamental in solving T&O incidents.

Gas chromatography – mass spectrometry (GC-MS) is the standard fit-for-purpose technique to detect, identify and quantify odorous organic compounds, that constitute the largest part of the chemical contributors to water T&O. Various GC-MS technologies and methods can be applied as diagnostic tools, to detect and confirm the presence of compounds causing undesirable T&O. In addition, GC-MS can be applied in the spatio-temporal monitoring of T&O in waters and has the ability of accurate quantification of known, commercially available compounds. However, GC-MS may not be successful in all cases, as it is also susceptible to some drawbacks and limitations.

A great challenge in the analysis of water T&O by GC-MS is the need for low limits of detection, as several T&O have extremely low odor threshold concentrations, sometimes at sub-ng/L levels. As a consequence, efficient methods of extraction and pre-concentration have to be applied at the sample preparation stage. For this purpose and to increase sensitivity, conventional laborious extraction techniques such as liquid-liquid extraction and solid-phase extraction are being increasingly replaced by advanced methods such as Head-Space Solid Phase Microextraction (HS-SPME), Stir-Bar Sorptive Extraction (SBSE) or Closed-Loop Stripping Analysis (CLSA) (Bruchet, 2006). Applications of fully automated SPME samplers are increasing as they offer improved sensitivity, reproducibility and high throughput and these have been standardized for the analysis of a range of volatile organics (VOC) in water (ISO 17943:2016). However, to capture a wide range of T&O, HS-SPME methods require

further in-house development and optimization. SBSE is simple and fast and facilitates the transfer of samples from distant locations to the laboratory, however, SBSE requires an upgrade of the GC system. CLSA combined with large volume GC-MS injection has been successfully applied for the diagnosis of water T&O episodes and a CLSA device has been commercialized, but this technique may not be efficient for routine monitoring or for the analysis of a large numbers of samples because it is more time-consuming and laborious, since samples are processed one-by-one.

Identification of compounds causing T&O by GC-MS normally involves non-targeted analytical approaches (NTA) aiming to cover as wide range of T&O as possible. This is a challenge, as known T&O in addition to known-unknowns (i.e., known molecules not previously reported as water T&O) and unknown-unknowns (i.e., new molecules not included in chemical registries and repositories) must be included in the scope of analysis. NTA approaches are largely based on matching the obtained mass spectra to those of open-source, commercialized or in-house spectral libraries. This means that good quality mass spectra must be produced, which may be a demanding task, especially when concentrations of T&O are close to detection limits, or when overlapping spectra occur. Mass spectra need to be processed for deconvolution and elimination of background signals with open-source or commercialized cheminformatics software packages before they can be matched to standardized library spectra. The level of confidence in identifying T&O is an issue that should not be neglected, especially for unknowns or when commercial standards of the suspected compounds are not available. Use of complementary existing data including retention indices or fragmentation spectra may improve confidence levels; however, identification may remain inconclusive.

The widely used NIST/EPA/NIH MS library includes over 350000 Electron Ionization (EI) spectra covering more than 300000 compounds with over 130000 retention indices. However, such broad databases do not specifically indicate compounds that may be relevant to water T&O, to assist laboratories investigating T&O episodes. Specialized databases exist in other related areas, such as in food flavor research, The Odorant Database of the Leibniz Institute for Food Systems Biology at the Technical University of Munich (Leibniz-

LSB@TUM) includes sensory and chromatographic data of more than 1700 food odorants extracted from more than 700 publications (Kreissl et al., 2022). By developing an open-source database coupled with an expert system dedicated to water T&O, that would include all compounds that have been reported in T&O incidents, with relevant information, water utilities and research laboratories would be able to considerably improve their diagnostic capabilities. To support the integration of a future WaterTOP database into already existing data analysis pipelines and the connection to supplementary databases, the FAIR principles Findable, Accessible, Interoperable and Reusable will be followed (Wilkinson et al., 2016).

Identification of the T&O compounds among a plethora of chromatographic peaks characterizing a water sample is not immediately possible, since intensities or other features in the chromatogram are not related to their sensory properties. Gas chromatography – olfactometry (GC-O) and GC-MS-O can assist in solving these problems, by simultaneously providing sensory, chromatographic and mass spectrometric data. Although these techniques have been applied in the elucidation of water T&O incidents (Hocherau & Bruchet, 2004) and identification of odorants that migrate to water from cross-linked polyethylene water pipe (Kalweit et al. 2019), their use is not widespread in the water sector, as they require specialized know-how and training. This is another area where water utilities could benefit from transfer of expertise from the food or cosmetics sectors, where GC-O and GC-MS-O are more commonly applied.

Many water T&O are products of microbial metabolism: more than 200 volatile and odorous compounds have been reported as metabolites of cyanobacteria and algae which can thrive in surface water reservoirs (Watson, 2004). Analytical and cheminformatics techniques applied in the context of metabolomics can be used to study this complex microbial "volatilome" (Meredith & Tfaily, 2022). Gas chromatography coupled to high-resolution mass spectrometry (GC-HRMS) is expected to enable the discovery of new microbial metabolites that could be relevant to water T&O. Emerging technologies, such as the comprehensive two-dimensional gas chromatography (GC×GC), that provides enhanced separation power, can further enable identification of unknown compounds in complex samples, especially when

coupled to HRMS. The potential of these technologies in water T&O studies is largely unexplored, offering a new promising area in water quality research.

Quantitative monitoring of known water odor compounds can be carried out by GC-MS using targeted analytical approaches, if standard compounds are available for calibration. If needed, sensitivity can be further increased with GC-MS/MS (e.g. triple quadrupole instruments), to reach very low detection limits. The gold standard in odorant quantitation is application of a Stable Isotope Dilution Assay (SIDA), especially when it comes to ultra-trace amounts and complex matrices. SIDA uses isotopically substituted analogues of the target compounds as internal standards, to compensate for any losses during sample preparation and measurement procedures. However, SIDA applications are rare in water T&O testing as T&O isotopologues are scarcely available, emphasizing the future need for chemical synthesis and commercialization (Porcelli et al., 2020).

Targeted identification and quantitation of taste-active organic compounds in food primarily uses triple quadrupole mass spectrometers hyphenated to liquid chromatography (LC-MS/MS), while inorganic anions and cations are monitored by means spectrometric approaches such as inductively coupled plasma mass spectrometry (ICP-MS) or ion-exchange chromatography coupled with suppressed conductivity detection. As analytical methods based on liquid chromatography often permit the direct injection of beverage samples with minimal sample preparation, recent developments enabling the simultaneous quantitation of odorants and tastants by UHPLC-MS/MS open up new paths for the detection of water T&O issues (Hofstetter et al., 2019).

4. Hazards of T&O compounds and risk assessment

T&O compounds could in principle represent a health risk, but being perceived by nose or taste, exposure is generally prevented by the fact that consumers refuse to consume water with unacceptable organoleptic characteristics. Therefore, to understand possible risks to human health of T&O compounds, it is important to know their odor or taste perception threshold concentrations (OTC or TTC) when

dissolved in water. These can be compared with health-based guidance values (GV), when available: when the GV is much higher (e.g., two or more orders of magnitude) than the OTC/TTC, the T&O compounds should not present a health risk, since exposure is prevented. The recommended drinking water GVs are derived to protect humans from long-term to lifetime consumption and may be considerably lower than the levels considered 'safe' for short-term exposure, as it occurs for many T&O compounds. This represents a conservative factor to estimate possible risk associated with the presence of T&O. Nevertheless, the margin between the OTC and the GV should be sufficiently high (> by a factor of 100), as sensory detection of these compounds (i.e., the relationship between the perceived odor intensity and the concentration of the compounds) can vary between people and even within one individual over time. It is therefore very difficult to establish a single threshold concentration (TC) that can be applied to the whole population, thus TCs would be better described by a range of values (Dietrich 2015).

Although the above consideration can be valid for single substances, T&O perception is not always a reliable alert, because the likely concurrent presence in water of different T&O producing substances can generate an altered perception. Since having mixtures of compounds in water is the rule rather than the exception, understanding the interactions among T&O species represents a significant challenge.

The WHO drinking water guidelines (WHO, 2022) report a list of compounds (of both natural and anthropogenic origin) that can change the aesthetic parameters of drinking water at concentrations well below those which can cause known adverse health effects, therefore no GVs have been derived for most T&O. The WHO list is not exhaustive and there are many additional T&O compounds, which have been detected in waters. Some of these are also used as food or feed additives, and the evaluation of oral exposure by international authorities (e.g., EFSA, EPA, WHO, ECHA) can be directly used to assess GVs for drinking water. For some flavoring agents, the evaluation for inhalation exposure is sometimes available: it is important to stress that a route-to-route extrapolation can be considered only if the potential kinetic differences associated with the different exposure routes are known.

A more critical situation is for those T&O substances produced by microorganisms, for which data are generally scant. The production of T&O compounds by microorganisms in terrestrial, natural and controlled aquatic environments is welldocumented and includes actinobacteria (actinomycetes), fungi, micro-algae and cyanobacteria (e.g., Lanciotti et al., 2003). Among many identified compounds, WHO considers only two, geosmin and MIB, which occur widely and have been mostly studied. Their toxicological thresholds are well above their OTC and no GV have been defined although recent studies suggest possible adverse effects in a test organism (zebrafish) (Zhou et al., 2020). Moreover, the presence of many still uncharacterized T&O compounds of biological origin creates problems in the assessment of the risk. Few field and lab studies show the co-occurrence of multiple T&O compounds and of T&O compounds plus other natural contaminants, including cyanotoxins. This could give rise to combined exposures and unknown effects, but more mechanistic studies are still needed to understand the relationships between the different compounds and to assess if and how T&O could be used as early warning for more complex water quality problems. Other data gaps include the environmental fates of T&O compounds, potential exposure via multiple routes, and bioaccumulation - biomagnification - depuration of T&O compounds and possible transfer along food chains.

Due to the revised EU Drinking Water Directive (2020/2184) water safety planning (WSP), a proactive approach based on risk assessment and management, will become mandatory for all EU drinking water-producers and -suppliers after January 2023. The T&O of water should always be taken as a generic hazard, considering the difficulties described in previous paragraphs in identifying the causative compounds in T&O incidents. An optimum tool for identifying this type of hazard is a reliable system for complaint tracking and handling. Even if there are currently no T&O complaints, water suppliers should develop their investigation and management plans to anticipate possible future T&O episodes. Dealing with such cases, one must keep in mind that the cause of the problem may originate anywhere from source to tap, including the raw water, the treatment plant, the distribution network and the domestic system. To maximize the effective implementation of WSPs, the water-supplier may establish and train a consumer T&O panel to provide early warning of water quality changes.

Such a panel is also a suitable medium to strengthen communication with consumers and increase utility confidence and proficiency.

5. Water treatment for removal of T&O

Despite the current growing concern about the improvement of the sensory quality of drinking water, the removal of T&O compounds from the water is highly challenging, since the T&O thresholds are extremely low, e.g., in the range of ng/L. Effective water purification requires highly efficient, cost-effective, and sustainable methods. A few conventional water treatment methods exhibit efficiency to remove T&O compounds from water, but they present significant limitations. Common disinfectants and oxidants such as chlorine, chlorine dioxide, or potassium permanganate alone are unable to control most T&O in drinking water, while ozone and a combination of ozone/hydrogen peroxide has led to superior performance (Antonopoulou et al. 2014; Glaze et al., 1990). As demonstrated in a study of 95 odorants in raw and finished water at full scale conventional or O₃/Biological Activated Carbon (BAC) water treatment plants, overall greater removal of odorants was observed for O₃/BAC plants, although the removal of indoles, phenols and sulfides were similar for both treatment processes (Wang et al. 2021). Adsorption by powdered/granular activated carbon (PAC/GAC) has been effectively employed in large-scale applications, however reduced adsorption capacity and efficiency are often exhibited due to the presence of NOM (Zamyadi et al., 2015; Chestnutt et al., 2007). Activated carbon is also used in point-of-use filters to improve water quality. In this field, alternative carbon materials with fine-tuned properties such as high surface area, high affinity, and adsorption performance, could be also adopted in the fabrication of filters for T&O removal.

The limited efficiency and drawbacks presented by conventional treatment technologies have led to the increasing interest of the scientific community for the development of novel treatment options, known as Advanced Oxidation Processes (AOPs). AOPs are based on the production of various reactive oxygen species with low selectivity, including hydroxyl radicals (HO[•]), able to degrade a wide range of chemically stable organic pollutants. Established and emerging AOPs, such as

 UV/H_2O_2 , UV/O_3 , $UV/O_3/H_2O_2$, UV/Cl_2 , photo-Fenton and heterogeneous photocatalysis, have been recently studied for the treatment of T&O compounds in various aqueous matrices (Antonopoulou et al., 2020; Antonopoulou et al. 2014; Kim et al., 2016).

UV-based AOPs have been extensively studied to improve the efficiency and effectiveness of T&O removal (Antonopoulou et al. 2014). These processes offer fast kinetics, high degrees of mineralization, and can simultaneously be used to remove odorants and other micropollutants as well as for disinfection. Among the UV-based AOPs, homogeneous processes seem to be more promising as they can combine high efficiency and cost-effectiveness. The homogeneous processes are commonly more attractive compared to the heterogeneous systems, due to their capacity of generating oxidative species in the absence of a solid catalyst, which poses high costs due the catalyst separation requirement after treatment. A representative and promising AOP with potential applicability for T&O control is UV/Cl₂. Currently, UV and chlorination are widely used processes in Drinking Water Treatment Plants (DWTPs) and their integration (UV/Cl₂) is easily applicable, with residual chlorine potentially acting as disinfectant. This can reduce the complexity and the total cost of the process (Remucal and Manley, 2016).

Heterogeneous photocatalysis could also be characterized as a suitable AOP for T&O control, and its nature brings more capabilities/opportunities for providing drinking water in locations where centralized water treatment is not available (e.g., small-scale, or point-of-use applications). Other prospective methods include catalytic ozonation, sonolysis, and electro-AOPs. Since AOPs are based on various reactive species, different reaction pathways can occur. The reactivity of each species is significantly related to the chemical structure of the target molecule and consequently detailed mechanistic investigations are critical for the evaluation of treatment process efficiencies.

Despite the available recent literature which indicates an increasing interest in future applications of AOPs for the removal of T&O, there are many issues that should be clarified before their practical application. Since conventional treatments are already implemented in DWTPs, the potential improvement of their infrastructures is a key aspect that should also be investigated by the scientific community and water

suppliers. Aiming at the highest efficiency, the selection of the appropriate technologies potentially depends on the application scale (large-, small-scale, point-of-use), as well as on the quality of the source water. Concerning the quality of source water, NOM can potentially have a detrimental effect on treatment efficiency depending on its content and composition. Additionally, NOM can act as precursor of transformation products (TPs) or as photosensitizer.

Many gaps and challenges must be addressed to achieve improvements in the T&O treatment efficiencies. Detailed studies should be conducted under realistic conditions, i.e., using drinking water as matrix and at environmentally relevant concentrations, near the OTC of the compounds. Other aspects deserving attention are the integration of conventional and/or AOPs, the evolution of T&O after each treatment step, and the elucidation of TPs formed during AOPs. In addition, the impact of T&O, their TPs, and TPs from NOM to human health should also be considered. The safe application of any treatment pre-supposes the avoidance and minimization of TPs and a comprehensive cost estimation, rendering pilot-scale studies necessary before full-scale implementation. All these tasks are fundamental to improve the overall process and to have a better understanding of their applicability under real conditions. It is necessary to keep in mind, the stronger oxidation process, the higher risk of toxic compounds formation.

6. Closing the gaps: the WaterTOP network

The state-of-the-art evolved to define and resolve sensory issues in water, requires the integration of four areas: (a) sensory analysis to describe the issue; (b) chemical analysis to determine the identity and concentration of T&O; (c) assessment of the associated health risks and (d) control and treatment strategies to mitigate the problem. This integrated approach requires expertise and contributions from different scientific disciplines as well as interaction with end-users to raise awareness and widen applications of the best available tools and techniques.

WaterTOP (COST Action CA 18225) is a pan-European and international network of experts, end-users and stakeholders aiming to promote research and increase capabilities and capacities in the field of water T&O, applying the integrated approach (Figure 3). In particular, the network aims to consolidate the largely fragmented

existing knowledge and to exploit cross-sector transfer of expertise from other sectors such as food flavor analysis, for the benefit of water supplies.

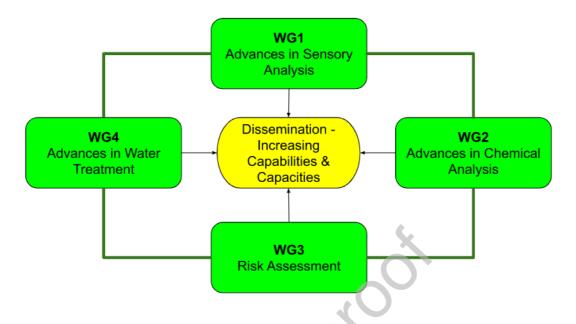


Figure 3: Structure of the WaterTOP network. WG: Working Group. Source: CA18225 MoU.

WATERTOP further aims to close the existing gaps in the field to increase the use of T&O for diagnosis of water quality-related problems. The main gaps identified by the network show the need for: (a) advances and harmonization of sensory and analytical methods including sensors; (b) better understanding of the effects and hazards of T&O; (c) improvement of the efficiency of water treatment to remove T&O; and (d) integration of T&O in the context of WSPs.

WaterTOP funds collaborative research projects (Short-Term Scientific Missions, STSM), training schools and workshops, aiming to train and develop the next generation of water quality researchers and employees. Products and outcomes of the work carried out are disseminated mostly as open-access publications and using the network's website and social media.

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Perspective

Water taste and odor (T&O): challenges, gaps and solutions from a perspective of the WaterTOP network

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Declaration of interests

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