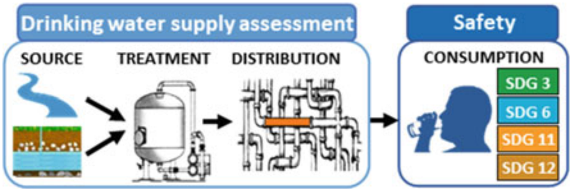
A Risk-Based Approach for Contaminants of Emerging Concern in Drinking Water Production and Distribution Chain

Beatrice Cantoni

Abstract Provision of safe drinking water (DW) is one of the major requisites for human health, related to four Sustainable Development Goals (SDGs) of the United Nation 2030 Agenda: SDGs 3 (Good health), 6 (Clean water and sanitation), 11 (Sustainable cities) and 12 (Responsible production and consumption). However, this is hindered by the presence, especially in highly-anthropized contexts, of contaminants of emerging concern (CECs) in DW, that may pose a risk for human health. The present study aims at developing a holistic framework to support both (i) decision-makers for CECs prioritization in DW regulation and (ii) water utilities for the selection of appropriate monitoring and treatment interventions for the optimization of DW supply system. In detail, a quantitative chemical risk assessment (QCRA), including uncertainties related to both exposure and hazard assessments, was developed. Then, it was combined with testing and modeling of CECs fate in treatment processes and in distribution network, obtaining a robust tool to achieve the above-mentioned SDGs.

Graphical Abstract



Keywords Activated carbon · Contaminants of emerging concern · Distribution networks · Drinking water · Fate predictive modeling · Pipe relining · Quantitative chemical risk assessment (QCRA)

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1 Introduction

In recent years, the presence of micropollutants in the aquatic environment has become an issue of growing global concern. Great attention is paid to the so-called Contaminants of Emerging Concern (CECs), that belong to several families of chemicals discharged from households (e.g. pharmaceutical active compounds, estrogens), agriculture (e.g. pesticides) and industrial processes (e.g. perfluorinated compounds, alkylphenols) [1]. CECs are currently not included in routine monitoring programs, although they are potentially hazardous, as some are persistent and biologically active, even being present at extremely low concentrations in the aquatic environments (ranging from ng L^{-1} to μg L^{-1}), that made them hard to detect and quantify [2]. Anthropic activities result in direct and indirect discharge of thousands of CECs in surface water and groundwater, used as drinking water (DW) sources. Few CECs have been introduced in the revision of the European Directive on DW, while a relevant number of compounds is still unregulated or just candidate for future regulations. Moreover, the revision of the European DW Directive promoted a shift in the current paradigm, pushing the preventive estimation of human health risk, in order to identify the main risk sources and prevent and minimize risks for the consumer throughout the whole supply system (from source to tap). Human health risk prediction consists of understanding whether CECs exposure concentrations exceed a tolerable health-based threshold, derived from toxicological studies. However, the use of a risk-based approach is not easy to be achieved for CECs due to several knowledge gaps. In particular, it is hard to evaluate CECs exposure levels in DW, firstly because of their low concentrations compared to the LOQ (Limit of Quantification) values of the analytical methods, which are in continuous refining; this results in monitoring databases characterized by high percentages of censored data, i.e. data below the LOQ. Moreover, uncertain estimation of CECs exposure levels in DW is also due to a lack of consolidated engineering knowledge about their fate throughout treatment processes in drinking water treatment plants and in drinking water distribution networks. Finally, high uncertainty is related also to CECs toxicity that hinders the prioritization of CECs to be included in the regulations and also the limit to be set.

The present work aims at filling current knowledge gaps in the field of risk assessment related to CECs in DW supply systems, and at developing an holistic risk assessment approach providing an effective tool to support: (i) decision-makers in evaluating the current human health risk and prioritizing CECs regulation, (ii) water utilities in planning affordable and effective upgrading, management and/or monitoring interventions for risk minimization throughout the whole water supply system.

Actually, the present research work is crucial to achieve provision of safe DW, that is one of the major requisites for human health, related to four of the 17 Sustainable Development Goals (SDGs) of the United Nation 2030 Agenda. In particular, **SDG 6** is completely devoted to "*Ensure availability and sustainable management of water and sanitation for all*", with targets related not only to accessibility and quantity of DW, but also to its quality, as reported in target 6.3 that aims by 2030, to improve water quality by reducing pollution, eliminating dumping and minimizing release of hazardous chemicals and materials. Secondly, the provision of safe DW has high impacts on the achievement of **SDG 3**, in order to "*Ensure healthy lives and promote well-being for all at all ages*". This is particularly clear when looking at target 3.9, aiming by 2030 at substantially reducing the number of deaths and illnesses from hazardous chemicals and air, water and soil pollution and contamination, and target 3.d, pointing at strengthening the capacity of all countries, in particular developing countries, for early warning, risk reduction and management of national and global health risks. Being the focus of this research the reduction of CECs in DW in highly-anthropized environments, this would help in "*Making cities and human settlements inclusive, safe, resilient and sustainable*", that is **SDG 11**. Finally, the proper management of the DW production and supply system and the consequent increase in citizens trust in tap water are two key elements to achieve **SDG 12**, "*Ensuring sustainable consumption and production pattern*". This is particularly linked to target 12.2 that promotes the achievement of sustainable management and efficient use of natural resources, and target 12.8 ensuring that people everywhere have the relevant information and awareness for sustainable development and lifestyles in harmony with nature.

2 Rationale and Methods

2.1 Problem Identification and System Conceptualization

To achieve the project goal of developing a holistic risk assessment approach to minimize health risk related to CECs in DW, the first step is the description of the system and the identification of the hazards that may influence tap water quality deterioration. The DW supply system was conceptualized in 3 elements before water is delivered to the point of use (i.e. the consumers): (i) DW sources (e.g. groundwater, surface water), (ii) drinking water treatment plants (DWTPs), and (iii) drinking water distribution network (DWDN). Thus, tap water deterioration can result from three main causes:

(1) Source contamination: the presence of CECs in source water can be due to direct or indirect discharges from different anthropic activities.
(2) Inefficiency of DWTPs towards CECs removal: currently the best available technology for CECs reduction, already present in DWTPs, consists in Granular Activated Carbon (GAC) filters. Failure of DWTPs towards CECs can be due to several factors, as low GAC-contaminant affinity ensuing low adsorption capacity, non-optimal configuration of the GAC filters in terms of dimensions and operation (in series or in parallel), and bad management with late exhausted GAC regeneration.
(3) Recontamination of treated DW through the DWDN: this could occur in case of contaminants release from pipes materials (metals, plastics) in contact with

water. This phenomenon depends on the nature of pipes materials, pipes maintenance and renovation management (e.g. substitution or relining) to prevent materials deterioration and on the effect of water aggressivity and corrosion potential on pipes.

As for the effects generated by the potential tap water contamination, a direct consequence may be the health risk for citizens, depending on the exposure levels and hazard characteristics of the analyzed CECs. On the other side, due also to the lack of knowledge and awareness of citizens about tap and bottled water quality and impacts, tap water deterioration brings citizen to drink less tap water and increase bottled water consumption. This choice has multiple negative effects, such as the exposure to health risks resulting by pollutants released by bottled water, environmental impacts due to the whole life cycle of bottled water and higher costs for citizens for drinking water.

2.2 Design of the Research

Based on the identified potential problems, experimental activity, at lab- and full-scale, advanced statistical methods and modelling techniques were combined to apportion the contribution of each element of the DW supply system in determining human health risk, in order to prioritize mitigation actions in view of an overall risk minimization. The work was structured in five tasks: (i) application of advanced method for censored CECs data analysis, (ii) development of a Quantitative Chemical Risk Assessment (QCRA) for CECs in DW supply, (iii) and (iv) predictive modelling of GAC adsorption performance towards PFAS and pharmaceuticals, respectively, and (v) modelling of bisphenol A (BPA) release from epoxy resins used for pipe relining and fate in DWDN. These tasks are interconnected according to the schematic overview given in Fig. 1 and briefly described in the following paragraphs.

3 Methods and Relevant Results

3.1 Application of Advanced Method for Censored CECs Data Analysis

Dealing with CECs, due to the low concentrations and the continuously refining of the analytical methods, monitoring databases are rich in censored data, meaning samples with concentrations below the LOQ of the analytical methods and that therefore cannot be quantified but only reported in the database as “<LOQ”. Censored data are traditionally eliminated or replaced with a value between 0 and LOQ, leading to erroneous estimations, but also to not fully exploit the information contained in the monitoring database. To face this constraint, the advanced Maximum Likelihood Estimation method for left-censored data (MLE_{LC}), that combines the values above

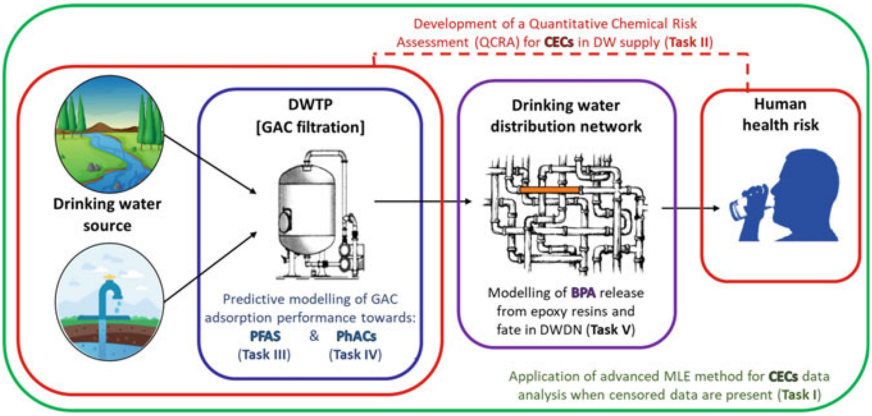

Fig. 1 Schematic overview of the project tasks and their focus on each part of the drinking water supply system

the LOQ with the information contained in the proportion of censored data, was here proposed and tested [3]. A field monitoring campaign was designed to evaluate CECs concentrations in addition to routinely monitored parameters in groundwater (GW) and DW in a highly urbanized area. A database was built with data of 19 contaminants (metals, volatile organic compounds, pesticides and perfluorinated compounds) in 5,362 GW and 12,344 DW samples, collected from 2012 to 2017 in 28 DWTPs of the monitored urbanized area. The $\mathrm{MLE}_{\mathrm{LC}}$ was applied to estimate the statistical distribution of CECs concentrations and results were compared to the traditional methods. Three applications for the comparison were selected, that are fundamental to predict the future raw water quality and to define intervention scenarios: evaluation of contaminants concentration time trend, estimation of treatment removal efficiency and risk assessment. Finally, a guideline was provided to select the data elaboration method to be preferred based on the comparison of the methods estimation errors, as a function of the percentage of censored data (from 0.3 to 99.0%) and the amplitude of concentration data range. This was made possible by the wide range of contaminants, the several DWTPs and the numerous sampling locations considered in this study.

An example of this task output is reported in Fig. 2, where the estimation error of the health risk likelihood (e_L) for the elimination and $\mathrm{MLE}_{\mathrm{LC}}$ methods is reported as a function of both the percentage of the censored data and the amplitude of data range, reported as the ratio between the LOQ and the concentration data 95th percentile.

The $\mathrm{MLE}_{\mathrm{LC}}$ method was demonstrated to be the most accurate method with estimation errors always below 20%. On the other hand, traditional elimination and substitution methods can lead to erroneous conclusions under- or overestimating the human health risk, especially for high percentages of censored data. This is of particular interest for CECs, often characterized by high censored percentages but with severe effects on human health also at very low concentrations. An accurate estimation of their risk is necessary to correctly plan the upgrading interventions of current

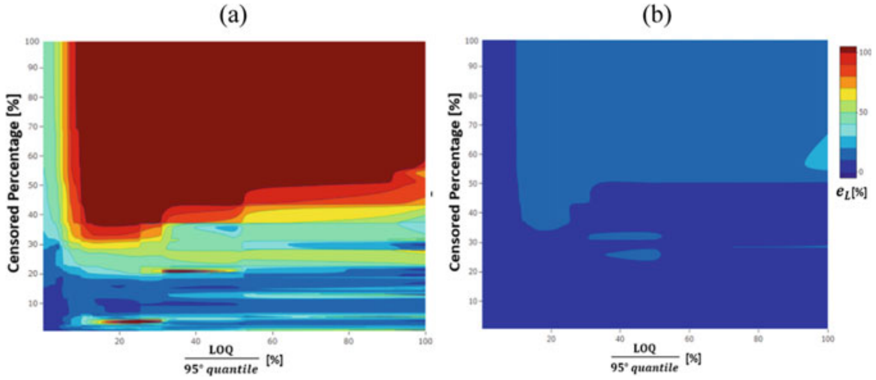

Fig. 2 Contour plot of the estimation error (e_L) of health risk as a function of the percentage of censored data and amplitude of data range for elimination **a** and $\mathrm{MLE}_{\mathrm{LC}}$ **b** methods [3]

DWTPs in order to meet the new regulatory limits proposed worldwide for CECs. In fact, both an underestimation or an overestimation of the exceedance probability could have important drawbacks in terms of underestimation of the risk or overestimation of the intervention needed, and the related costs, leading to potentially not precautionary or unsustainable intervention plans.

3.2 Development of a Quantitative Chemical Risk Assessment (QCRA) for CECs in DW Supply System

Currently, chemical risk assessment (CRA) in DW applications is deterministically performed and uncertainties are taken into account by selecting conservative point values, such as a high exposure concentration, or a lower bound estimate of the health-based guideline level [4]. Then, the ratio between the exposure concentration and the health-based guideline level point values is calculated as the deterministic benchmark quotient (BQ) that provides an indication of no risk or risk for BQ below or above 1, respectively [5]. However, evaluating the risk for CECs in DW is not easy due to several knowledge gaps and high uncertainties related both to their exposure levels and hazard characteristics. Therefore, in this study [6], a new probabilistic procedure, that is the quantitative chemical risk assessment (QCRA), was developed including in risk calculation the uncertainties in both exposure and hazard assessments, obtaining as output the BQ probabilistic distribution, useful to estimate the probability of BQ of exceeding threshold value of 1, $P(BQ > 1)$.

As for the exposure assessment, the probabilistic distribution of CECs concentration in DW was estimated based on their concentration in source water and simulating the breakthrough curves of GAC adsorption filters, through the Ideal Adsorbed Solution Theory (IAST) model. IAST model was implemented by AquaPriori, a Python based treatment simulation tool developed by KWR (Utrecht, NL), that was upgraded to accept distributions—instead of point values—for input parameters. The uncertainties in the CECs hazard assessment were included using the APROBA-Plus tool developed by the USEPA, as described by [4].

The model inputs and output uncertainties were evaluated by sensitivity and uncertainty analyses for each step of the risk assessment to identify the most relevant factors affecting risk estimation, highlighting future research needs to improve reliability of risk assessment. To stress the potential of this new QCRA approach, several case studies and GAC management options were considered, with a focus on BPA as an example CEC. The probabilistic risk quantified by the QCRA was compared to the deterministic one estimated by the traditional CRA. An example of how using the developed QCRA procedure to manage and optimize CECs treatment in the DWTP is provided in Fig. 3, where the risk estimation output of the deterministic CRA and QCRA are displayed as a function of two operating conditions of the GAC filters: (i) the Empty Bed Contact Time (EBCT), that is the time the DW is in contact with the GAC in the filter, and (ii) GAC regeneration time (BV_{REG}), that is the time when the exhausted GAC is treated to desorb contaminants and restore its adsorption capacity.

The EBCT has a negligible influence on the human health risk, compared to the regeneration time. Therefore, the EBCT seems not to be a relevant parameter through which optimize BPA removal by GAC process, suggesting that engineered intervention to increase EBCT does not imply any significant risk reduction. Both the deterministic BQ and the probability of exceeding BQ equal to 1 ($P(BQ > 1)$) decrease with the reduction of the regeneration time. However, using the deterministic approach (Fig. 3a), a regeneration time equal to 25,000 BV (bed volumes) would be selected as the optimal value under which there is no significant risk ($BQ_{DET} = 1$), while including all the uncertainties within the QCRA it comes out that there is still a 12.0% probability of exceeding BQ value equal to 1 (Fig. 3b) and the optimal

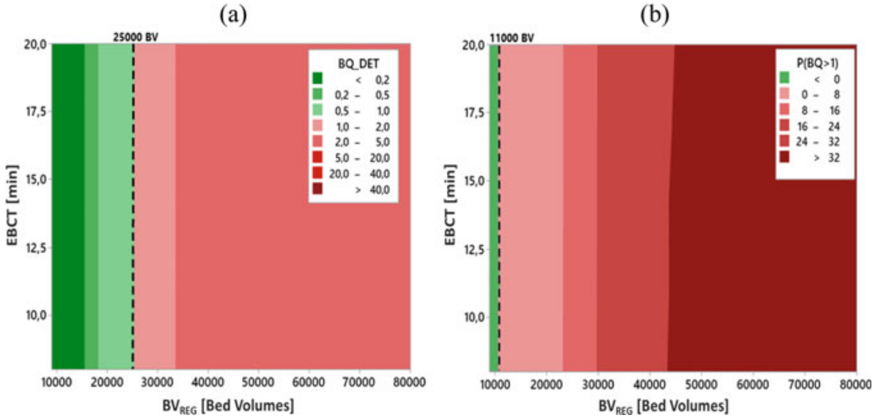

Fig. 3 Contour plot of BQ_{DET} **a** and $P(BQ > 1)$ **b** as a function of the empty bed contact time (EBCT) and the regeneration time (BV_{REG}) [6]

regeneration time of 11,000 BV would result in a virtual 0% P(BQ) > 1. Therefore, the deterministic BQ provides the less precautionary approach, which does not identify residual human health risk in specific cases. Actually, based on the deterministic BQ, costs associated to the regeneration time adopted as optimal threshold would lead to benefits lower than expected. In conclusion, the QCRA is more effective than deterministic CRA in evaluating the effect of each management option in risk minimization, permitting to select and prioritize the most appropriate interventions.

3.3 Modeling of GAC Adsorption Performance Towards CECs

Since with the QCRA it was found that modelling of GAC breakthrough curves has a relevant role in the accuracy of risk estimation [6], a thorough experimental work has been designed and performed to more accurately model GAC performance towards CECs, in particular pharmaceutical active compounds (PhACs) and perfluoroalkyl substances (PFAS) (for a detailed description see [7]). Four commercial GACs were tested by both isotherm batch experiments and rapid small-scale column tests (RSSCT), to calibrate CECs breakthrough curves. Experiments were performed on 8 PFAS and 8 PhACs in 3 water matrices, which were tap water and additional two synthetic matrices at lower dissolved organic carbon (DOC) and two levels of conductivity. As for activated carbon, 4 GACs were tested, differing for origin (bituminous or coconut-based), surface charge (neutral or positively charged), number of reactivation cycles (virgin and reactivated GACs) and porosity (micro- and mesoporous). Results were explored through multivariate analyses (i.e. factorial and cluster analyses) and used to calibrate a performance model able to predict the breakthrough curves as a function of CECs, activated carbon and water characteristics, and their interactions.

GAC performance can be related to the isotherm constant, K_F, and the RSSCT parameter BV_{50}, corresponding to the time at which the CEC reaches the 50% of the breakthrough curve, having a filter outlet concentration equal to half the inlet concentration (Cout/Cin = 0.5). Higher K_F and BV_{50} values indicate greater adsorption capacity of the adsorbent. In general, a good agreement among isotherm and RSSCT results was outlined, as suggested by data in Fig. 4, showing that for the majority of the analyzed PFAS, both K_F and BV_{50} increase from short-chain hydrophilic and marginally hydrophobic PFAS to medium-chain and hydrophobic PFAS, to PFOS.

In addition to the confirmation of literature evidences on the effect of compounds hydrophobicity on GAC adsorption capacity, it was found that GAC surface charge affects performance more than GAC porosity, therefore electrostatic interaction can be inferred as the main adsorption mechanism for PFAS. Consequently, GAC's surface charge should be checked prior to the GAC selection for each case-study. In fact, considering the pH of the water to be treated, positively-charged GAC should

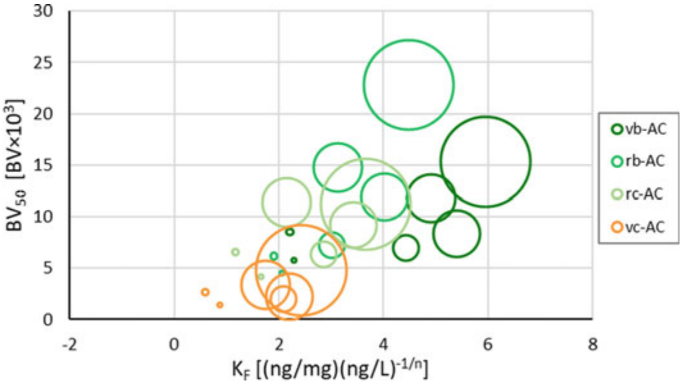

Fig. 4 Bubble chart for PFAS removal by adsorption on activated carbon (AC). Bubble centers are located according to the isotherm K_F and the RSSCT BV_{50}; bubble diameters are proportional to respective PFAS hydrophobicity [7]

be preferred rather than negatively-charged or neutral GAC, to fully exploit electrostatic attraction towards negatively-charged PFAS [8]. Moreover, the interaction between CEC hydrophobicity and GAC porosity was found to significantly affect performance. Therefore, GAC selection should also consider PFAS mixture in the source water to be treated.

Finally, a correlation was built between the reduction of UV absorbance at 254 nm (UVA_{254}), that is an easily measurable parameter also by on-line sensors, and CECs removal (Fig. 5), in order to evaluate whether UVA_{254} can be used as a proxy variable for CECs continuous on-line monitoring.

Actually, UVA_{254} removal is correlated to CECs removal, independently from the type of GAC and water matrix. Therefore, CECs-UVA_{254} correlations can be easily

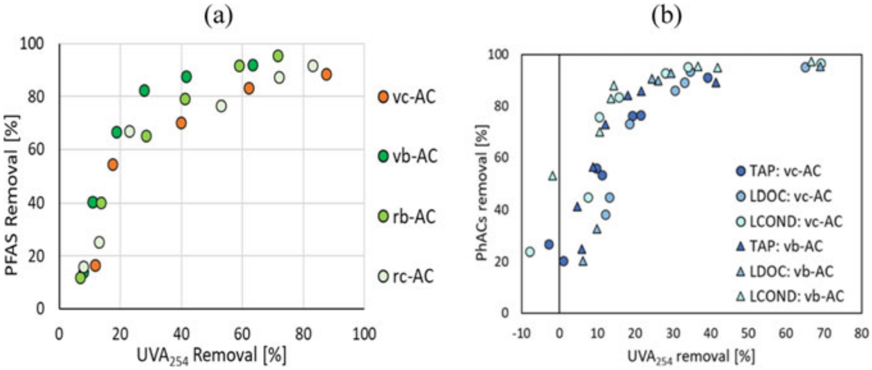

Fig. 5 Correlations between: **a** overall PFAS removal and UVA_{254} removal as a function of the four tested ACs [7], **b** overall PhACs removal and UVA_{254} removal as a function of two tested ACs and three water matrices

obtained with batch experiments, to be used in combination with UVA_{254} on-line monitoring data to predict the overall CECs breakthrough in real full-scale systems. This is an important tool to promptly identify possible system failures, which may result in human health risk, and to rapidly apply mitigation measures.

3.4 Modeling of BPA Release from Epoxy Resins and Fate in DWDN

Monitoring and management of DWDNs, including possible leaching from materials in contact with DW, have been stressed as crucial to avoid re-contamination of drinking water leading to a potential increase of human health risk. Recent scientific studies and regulations clearly highlighted the leaching of BPA from plastic materials used to renovate DWDNs pipelines as one of the major hazardous events, resulting in severe consequences for human health. Therefore, to complete the “from source to tap” holistic risk assessment, potential recontamination events in the DWDN should be properly studied. Here, a sound approach to evaluate BPA release from epoxy resins used to renovate pipelines, comprising experimental and modeling work, is reported [9].

Lab migration tests were performed on three commercial epoxy resins, characterized by different formulations, BPA content and installation techniques, put in contact with three water matrices: (i) tap water, (ii) deionized water, (iii) chlorinated tap water. The most critical resin was selected for a second set of experiments designed with the Design of Experiments (DoE) method, in order to build a BPA release model as a function of two water characteristics that were varied in a realistic range: (i) chlorine concentration, from 0 to 0.4 $mgCl_2$/L, (ii) water stability, described by the aggressivity index (AI) varying from 11.5 to 13.5. Tests lasted about 170 days to account for both short and long-term leaching. BPA migration over time (Fig. 6) was well described by a combination of two 1st-order kinetic models with an initial peak of leaching, a decrease and a second increase due to resins’ deterioration. The analysis of BPA migration trend over time, especially in the first week provides important insights on monitoring and management practices that water utilities should perform when renovating DWDN pipes with epoxy resins.

Looking at the effect of water conditions, an increase of residual chlorine leads to a decrease in BPA concentration in water. This is evident when comparing the trend in tap water without chlorine and with chlorine concentration at 1 mg/L, where the initial peak is absent. However, it is not BPA release to be reduce by increasing chlorine, but likely the BPA released is actually transformed in chlorination by-products, such as chloro-phenols and chloro-bisphenols [10]. Thus, it is important to monitor not only BPA but also its chlorination by-products in order to have a proper evaluation of the overall human health risk due to the mixture of chemicals. As for the water stability, aggressive waters display higher BPA release, but the effect is

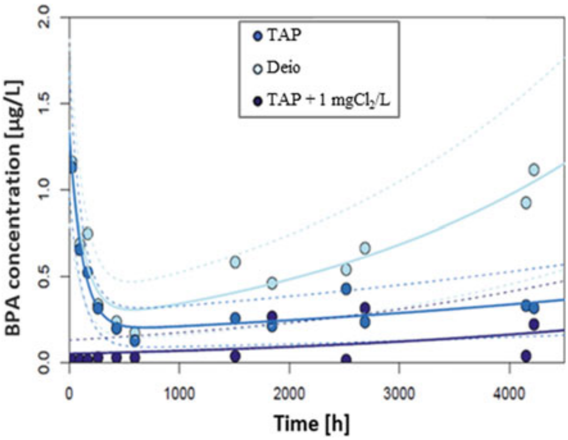

Fig. 6 BPA migration over time: dots correspond to experimental data as a function of the tested water matrix. Solid lines indicate the estimated models, dashed lines represent the models 95% confidence intervals [9]

more evident when looking at the total mass released, compared to the initial peak, for which the chlorine concentration has greater effect.

To evaluate the fate of BPA in the DWDN, the validated BPA release model was combined with the hydraulic model of a portion of a DWDN in a highly urbanized area, where two epoxy resins are installed. EPANET MSX software was used. A field monitoring campaign was also designed to measure BPA concentrations in locations nearby pipelines renovated with epoxy resins, for model validation. The model was successfully validated on full-scale monitoring data, demonstrating the occurrence of BPA leaching and the potential risk for consumers, especially if appropriate re-opening procedures are not adopted. The model allowed to simulate the current fate

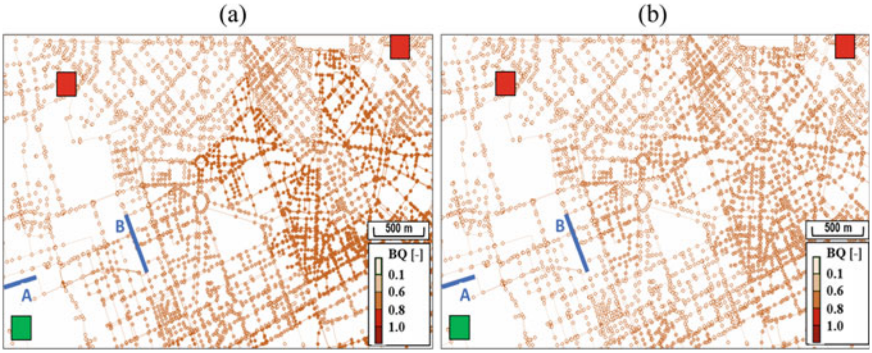

Fig. 7 Estimated BQ due to released BPA concentration in a portion of a DWDN where two pipes were renovated by epoxy resins relining (blue segments) as a function of the time from installation: **a** after 1 **day**; **b** after 30 days [9]

of BPA in the DWDN (Fig. 7), identifying the most vulnerable areas, with higher potential risk, described in terms of BQ; as a consequence, the combined model can be adopted to optimize monitoring and intervention plans, which can be site-specific customized to minimize human risk.

4 Conclusions

Drinking water supply systems should be managed by a risk-based approach to evaluate potential risks and devise preventive measures, in order to ensure the achievements of four of the 17 Sustainable Development Goals (SDGs) of the United Nation 2030 Agenda (SDGs 3, 6, 11 and 12).

The research work here described contributed to adapt and improve the application of risk assessment to control the spread of CECs in DW supply systems, overcoming some of the existing knowledge gaps and handling high uncertainties which characterize both CECs presence and toxicity and their removal. The work performed, integrating experimental and modeling features, has permitted to provide a supporting tool for:

- Water utilities, that need to verify whether their current control measures are adequate to meet new regulatory limits for CECs in DW and to elaborate intervention plans to effectively minimize human health risk. In this context, the QCRA can be used to apportion the contribution of each step of the supply system to overall risk minimization, to prioritize the interventions. The application of QCRA in combination with the simulation of GAC performance under different operating conditions allows optimizing GAC filters up-grade and management. Finally, several practical indications were provided both on the criteria for GAC and epoxy resins selection, and on planning monitoring campaigns to evaluate CECs removal by GAC filters and to minimize recontamination events for DW contact with materials in DWDNs.
- Decision makers, that need to prioritize CECs to be included in regulations, beside the high uncertainty involved in this process. In this context, the QCRA can be effectively used to assess the exposure and hazard of different CECs, including the related uncertainties, to evaluate which CECs pose the highest risk for human health. Moreover, the study on epoxy resins could provide important tools for decision makers to accurately regulate the characteristics of materials in contact with DW.
- Scientific community, that needs to fill the knowledge gaps and reduce the uncertainties related to CECs exposure in DW and resulting health effect. In this context, this work provided indications on how to reduce estimation errors in different applications when analyzing databases characterized by high percentages of censored data. Additionally, the sensitivity and uncertainty analyses applied in combination to the QCRA can be useful to identify future needed research investigations and directions to reduce uncertainties in risk estimation. Finally, this

work showed how to combine experimental tests at different scales, prediction modelling tools and field monitoring to reduce uncertainties related to CECs fate in GAC system and DWDN.

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