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Evaluation and comparison of centralized drinking water softening technologies: Effects on water quality indicators

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ABSTRACT

Drinking water softening is often implemented to increase consumer convenience e.g. by reducing lime scaling and soap use. Softening reduces hardness, but changes also the overall mineral composition of the water, depending on the technology. A broad spectrum of effects from softening has to be considered in relation to e.g. health and corrosion when selecting softening technology and design, otherwise adverse effects may be overlooked in the attempt to increase consumer convenience. We here provided a framework for evaluating softening technologies using water quality indicators for lime scaling, soap use, corrosion, human health, taste and removal of contaminants. None of the evaluated softening technologies scored positive on all the included water quality indicators. Precipitation technologies (lime/soda-ash softening and pellet softening) reduce the predicted copper and lead release, but negatively affect stainless steel corrosion expressed by the Larson Ratio. Pellet softening does not remove magnesium, which may limit the achievable softening depth, but maintains a protective effect against cardio-vascular diseases. Strong-acid cation exchange is not expected to affect the included corrosion indicators, whereas the effects from membrane separation (nanofiltration and reverse osmosis) and weak-acid cation exchange depend on the specific source water and process design. All the evaluated technologies reduce hardness, calcium carbonate precipitation potential (CCPP) and atopic eczema, but have potential adverse effects on dental carries (expressed by DMF-S). Our framework provides a better understanding of softening and can prepare water utility planners and managers for better decisions that balance the positive and adverse effects from drinking water softening.

1. Introduction

Centralized drinking water softening can provide socioeconomic and environmental benefits in areas with hard drinking water (Beeftink et al., 2021; Godskesen et al., 2012; van der Bruggen et al., 2009). Historically, softening has been implemented to reduce copper (Cu) and lead (Pb) released from pipe materials and thereby comply with drinking water quality guidelines (Mons et al., 2007). Nowadays, softening is often motivated in improvement of consumer convenience by reducing adverse effects from hard water such as increased soap use and lime scaling in household installations and appliances (Hofman et al., 2007; Mons et al., 2007). Thus, the resulting water quality and softening depth (i.e. hardness removal) is not defined by specific guidelines, but by the motivation for improvement of the water characteristics, by the effects in the distribution system, and by the consumers.

Drinking water softening technologies reduces the water hardness, which in practice is defined as the sum of calcium (Ca^{2+}) and magnesium (Mg^{2+}) ions (de Moel et al., 2006; Loewenthal and Marais, 1976). Softening technologies not only reduce hardness, but they also alter the overall mineral composition of the drinking water differently, depending on the technology (Mons et al., 2007; Ruhland and Jekel, 2004). Consequently, the effects from softening are beyond hardness removal (Tang et al., 2019b) and may also include effects on e.g. corrosion, health and taste. If hardness removal is the only aspect considered when designing the softening process, adverse effects may be overlooked compromising the overall benefits from softening.

It is very difficult to quantify how water treatment processes affect e.g. lime scaling or corrosion from the individual water quality

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parameters, but it can be predicted by water quality indicators (or indices). Indicators integrate a broad range of effects and are often applied in decision support analyses such as multi-criteria decision analysis to account for technical (e.g. performance), environmental (e.g. carbon footprint and energy use), economic (e.g. net present value and construction costs), and sociocultural (e.g. cultural acceptance) aspects (Beeftink et al., 2021; Godskesen et al., 2018; Hajkowicz and Collins, 2007; Hamouda and Huck, 2010). Considering all the above aspects allows for a broad evaluation of e.g. environmental and economic sustainability of the treatment process (Godskesen et al., 2018; Marques et al., 2015), but can also increase the complexity and data requirement (Hamouda et al., 2014). Unfortunately, in the desire to overcome this complexity the evaluation of water quality is sometimes limited to a few indicators such as “removal efficiency” for a single water quality parameter or pollutant (e.g. Hamouda and Huck, 2010; Santos et al., 2016), thereby neglecting the effects on the overall water quality.

So far, literature has either focused on the individual drinking water softening technologies (e.g. Höll and Hagen, 2002; Shahmansouri and Bellona, 2015; van der Bruggen et al., 2001) or reviewed the removal of specific, individual water quality parameters by various treatment processes (e.g. Kapoor and Viraraghavan, 1997; Karunanithi et al., 2019; Zhang et al., 2015). Only few papers have addressed the selection of softening technology and they only include few technologies and water quality effects (Bergman, 1995; Ruhland and Jekel, 2004; Sombekke et al., 1997; Tang et al., 2019b; Thompson and Azar, 1999). To our knowledge, a framework does not exist to evaluate softening technologies based on a broad spectrum of water quality indicators. Such a framework can provide decision support when choosing softening technology by achieving the full potential of softening while limiting adverse consequences on e.g. corrosion and human health, ultimately optimizing the overall water quality after softening.

Our study aimed to provide a framework for evaluation and comparison of softening technologies and their effects on soap use, corrosion, lime scaling, taste and health using selected water quality indicators. To fulfill the aim we 1) identified relevant indicators for water quality and their targets, 2) identified softening technologies currently used in full-scale, and 3) evaluated how each softening technology affects our chosen water quality indicators.

2. Materials and methods

2.1. Evaluation method

To evaluate the softening technologies and their consequences experienced by the consumers we developed a framework with four steps (Fig. 1). We distinguished between *water quality parameters* and *water quality indicators*. Consequences of softening such as corrosion or human health effects can be predicted using *water quality indicators* (e.g. Larson Ratio and CCPP), whereas *water quality parameters* refer to ionic species and other drinking water components required to calculate the indicators (e.g. Ca^{2+} or Mg^{2+} concentrations).

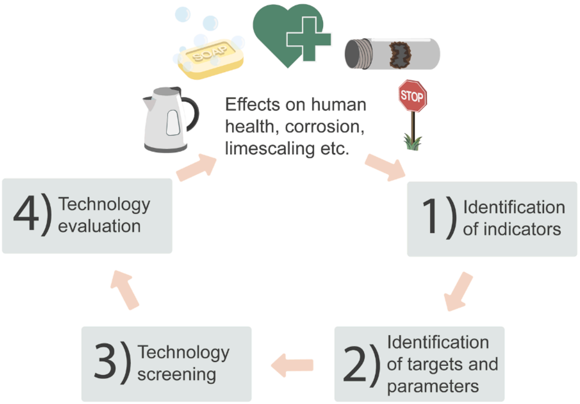

Fig. 1. Conceptual representation of stepwise method for evaluating the effects from drinking water softening technologies on e.g. corrosion, soap use, lime scaling and health.

In the first step of the framework, we identified relevant indicators (Section 2.2), as well as their targets and the water quality parameters required to quantify them (step 2, Section 3.1). In the third step, we identified softening technologies currently used in full-scale softening and screened for how they change the water quality parameters of importance for each indicator (Section 3.2 and 3.3).

In the final step of the framework (“technology evaluation”), we evaluated how each of the softening technologies affect the water quality indicators based on the changes to the water quality parameters (Section 3.4). For consistency, we describe a parameter change that brings an indicator further away from the identified target as an “adverse effect”. On the other hand, a “positive effect” describes a parameter change that brings the indicator closer to the target (Table 1).

Based on the evaluation of the individual parameters, the overall effect on the water quality indicator was assessed (Table 1). If a technology did not change the parameters, the overall effect was categorized as “no effect”. If a technology had only adverse changes to the parameters, the overall effect was characterized as “adverse” and similarly for positive effects. If the technology provided a mix of positive and adverse changes to the parameters, the overall effect was characterized as “indecisive”.

If a technology only changed a water quality parameter less than 15 % from its initial value, we considered that there would be no relevant water quality effects.

2.2. Water quality indicators

We selected a range of existing water quality indicators to evaluate a broad spectrum of consequences from softening (Table 2). Softening is implemented for various reasons, e.g. to reduce soap use, lime scaling or corrosion of copper and lead from pipe materials to the drinking water (Mons et al., 2007), and consequently, we included indicators to quantify these effects (Table 2). We included the Larson ratio, commonly used to assess the corrosion potential of drinking water. We included the health effects selected by Rygaard et al. (2009) (dental caries, cardiovascular diseases and atopic eczema) who valuated the effects from water quality changes due to reverse osmosis. Finally, we included water taste and the technologies’ ability to act as a barrier towards organic and inorganic contaminants in the evaluation (Table 2). We categorized the different water quality indicators based on their consequences (e.g. corrosion, health and taste) in a drinking water system.

Water quality parameters required to calculate the indicators and their target values were identified in the cited literature. Furthermore, we identified if minimizing or maximizing the parameter concentrations brings the indicator closer to the target value.

2.3. Technology screening

DTU Findit that encompasses databases such as Web of Science, Scopus and more (DTU Library, 2021), was used to identify technologies

Table 1
Conceptual representation of softening technology evaluation for each water quality indicator.

Technology	Parameter A *Removal = good*	Parameter B *Addition = good*	Overall effect
Technology 1	No effect	No effect	No effect
Technology 2	Adverse effect	No effect	Adverse
Technology 3	Positive effect	No effect	Positive
Technology 4	Adverse effect	Positive effect	Indecisive

Table 2
Selected consequences and corresponding water quality indicators that are potentially affected by drinking water softening.

Consequences		Indicator	Calculation method/definition	Reference
Soap use and limescale	Soap use	Hardness	$\sum[\mathrm{Ca}^{2+}]+[\mathrm{Mg}^{2+}]$	(de Moel et al., 2006)
	Limescale	Calcium Carbonate Precipitation Potential (CCPP)	Δcalcite to reach equilibrium, e.g. calculated in PHREEQC	(Tang et al., 2021)
Corrosion	Steel corrosion	Larson Ratio	$\frac{[\mathrm{Cl}^-]+2[\mathrm{SO}_4^{2-}]}{[\mathrm{HCO}_3^-]}$ 2)	(Larson and Skold, 1958)
	Copper release	Predicted copper release (Cu-release) [mg/L]	$0.88+0.015\mathrm{Alk}$ (pH 7.0) (3) $0.22+0.0055\mathrm{Alk}$ (pH 8.0) (3)	(Edwards et al., 1996)
	Lead release	Predicted lead release (Pb-release) [µg/L]	$1.027^{(\mathrm{T}-25)}\mathrm{Alk}^{0.677}\mathrm{pH}^{-2.726}[\mathrm{Cl}^-]^{1.462}[\mathrm{SO}_4^{2-}]^{-0.228}$ 3)	(Imran et al., 2006)
Health	Caries	Decayed Missing Filled Surfaces (DMF-S)	$\exp\left(1.05-\frac{0.18([\mathrm{F}^-]-0.33)}{0.25}-\frac{0.11([\mathrm{Ca}^{2+}]-83.50)}{25.63}\right)$ 3)	(Bruvo et al., 2008)
	Atopic eczema	Calcium concentration	$[\mathrm{Ca}^{2+}]$	(Mcnally et al., 1998)
	Cardio-vascular diseases	Magnesium concentration	$[\mathrm{Mg}^{2+}]$	(Kozisek, 2020)
Taste	Taste	None1)	-	(Platikanov et al., 2013)
Barrier against contaminants	Organic contaminants	Removal efficiency	Evaluation of overall potential for removal	
	Inorganic contaminants	Removal efficiency	Evaluation of overall potential for removal	

1) No indicator exists for evaluating changes to drinking water taste. Platikanov et al. (2013) identified ions with a positive and negative effect on taste that we used in our study.
2) All concentrations in mmol/L
3) All concentrations in mg/L, alkalinity in mg CaCO_3/L

for centralized drinking water softening and their effects on water quality. Search words included general terms (e.g. “water softening” and “hardness removal”), specific technologies (e.g. “cation exchange” and “pellet softening”), groups of water quality parameters (e.g. “divalent ion removal”) and specific water quality parameters (e.g. “sulphate removal drinking water”). The search included the entire paper and no restrictions on year publication, although recent publications (2010 and onwards) were prioritized over older publications.

We limited the literature search to:

- Technologies currently commercially available for full-scale centralized drinking water softening, excluding emerging technologies and technologies for pre- and post-treatment.
- Technologies that remove hardness from the water, excluding technologies claiming to physically reduce lime scaling in households using e.g. ultrasound, electromagnetism and electrostatic forces
- Studies that include water samples from actual drinking water systems, excluding studies with synthetically prepared water solutions with a limited number of ions

The latter point was necessary because membrane treatment of synthetic water compositions may vary greatly from natural waters, where e.g. the interaction between ions and ionic strength affect the rejection of ions (Santafé-Moros et al., 2005).

Some studies report water quality changes as concentrations before ($C_{\text{before softening}}$) and after softening ($C_{\text{after softening}}$). When appropriate (see Section 3.3), we converted these concentration changes to a percent removal/rejection (Eq. 1).

$$\text{Percent removal} = \frac{C_{\text{before softening}} - C_{\text{after softening}}}{C_{\text{before softening}}} \cdot 100\ \% \tag{1}$$

Table 3
Selected indicators, their qualitative targets and the water quality parameters required to calculate the indicators, including if they should be minimized or maximized to approach the indicator target.

Consequence	Indicator	Indicator target	Parameters		Reference
			Minimized to meet target	*Maximized to meet target*	
Soap use	Hardness	As low as possible	Ca^{2+}, Mg^{2+}	-	(de Moel et al., 2006)
Limescale	CCPP	Slightly positive ($\mathrm{CCPP}_{90} < 0.6$ mmol/L)	Ca^{2+}, pH 1), temperature, HCO_3^- 2)	-	(Tang et al., 2021)
Steel corrosion	Larson Ratio	As low as possible	SO_4^{2-}, Cl^-	HCO_3^-	(Larson and Skold, 1958)
Copper release	Cu-release	As low as possible	HCO_3^-	pH	(Edwards et al., 1996)
Lead release	Pb-release	As low as possible	Temperature, HCO_3^-, Cl^-	pH, SO_4^{2-}	(Imran et al., 2006)
Caries	DMF-S	As low as possible	-	Ca^{2+}, F^-	(Bruvo et al., 2008)
Atopic eczema	Calcium concentration	As low as possible	Ca^{2+}	-	(Mcnally et al., 1998)
Cardio-vascular diseases	Magnesium concentration	20 – 40 mg/L	-	Mg^{2+} 3)	(Kozisek, 2020)
Taste	-	-	Na^+, K^+, Cl^-	Ca^{2+}, Mg^{2+}, SO_4^{2-}, HCO_3^- 4)	(Platikanov et al., 2013)
Contaminants	Organic contaminants	Below guideline			
	Inorganic contaminants	Below guideline			

1) Low pH can lead to negative CCPP, which is undesired
2) For more accurate calculation of the CCPP, the concentrations of Mg^{2+}, Na^+, K^+, Cl^-, PO_4^{3-}, SO_4^{2-} and NO_3^- must also be known. These ions affect the ionic strength, which has little effect on the CCPP compared to Ca^{2+}, pH and HCO_3^- (Tang et al., 2021).
3) Mg should not exceed 150 mg/L (Rosborg and Kozisek, 2019)
4) TDS should not exceed 400 mg/L (Platikanov et al., 2013)

3. Results

3.1. Identification of indicator targets and relevant water quality parameters

To evaluate if water quality changes were positive or adverse, we identified a qualitative target (e.g. “as high as possible”) for each of the indicators presented in Table 2 (i.e. step 2 in Fig. 1, Table 3). The targets only apply for the specific indicator and may represent a less optimal target for other impacts unrelated to the softening technology. For instance, pH should be as high as possible to avoid copper corrosion, but a high pH reduces the efficiency of chlorine disinfection (Lahav and Birnhack, 2007), which was not included in our evaluation.

In addition, we identified water quality parameters required to calculate the indicators and if the concentrations of these parameters should be minimized or maximized in hard water to approach the indicator targets (Table 3):

- **Hardness,** and thereby the concentrations of Ca^{2+} and Mg^{2+}, should be as low as possible to reduce soap use (Mons et al., 2007).
- **CCPP** is mainly affected by the concentrations of Ca^{2+} and HCO_3^- as well as pH and temperature (APHA/AWWA/WEF, 2017), which are the parameters included in this study; however, CCPP is most accurately calculated when a range of further ionic species are included (as discussed in Tang et al., (2021)). CCPP should be as low as possible to reduce lime scaling. However, it should be slightly positive to avoid water that is $CaCO_3$-dissolving, which can lead to corrosion (Lahav and Birnhack, 2007).
- **The Larson Ratio** should be as low as possible to prevent steel corrosion (Larson and Skold, 1958). The Larson Ratio decreases with increasing HCO_3^- concentration or decreasing concentrations of Cl^- and SO_4^{2-}. On the other hand, increasing SO_4^{2-} concentration may also reduce Pb-release from pipe materials showing that some water quality parameters have contradictory effects on corrosion (Table 3).
- **Copper (Cu)-release** from especially new pipe materials increases with decreasing pH and increasing alkalinity and should be as low as possible (Edwards et al., 1996).
- **Lead (Pb)-release** from pipes can be predicted from the water alkalinity, temperature, pH, Cl^- and SO_4^{2-} concentrations (Imran et al., 2006). Temperature, alkalinity and Cl^- should be minimized and pH and SO_4^{2-} should be maximized for the Pb-release to be as low as possible.
- **Decayed, Missing, and Filled tooth Surfaces (DMF-S)** caused by caries among Danish school children was related to the Ca^{2+} and F^- concentration in drinking water (Arvin et al., 2017; Bruvo et al., 2008). DMF-S increases with decreasing concentrations of Ca^{2+} and F^-.
- **Ca^{2+}** has been associated with atopic eczema among children (Mcnally et al., 1998) and should be minimized.
- **Mg^{2+}** likely has a protective effect towards cardiovascular diseases (Kozisek, 2020) and should for that purpose be maximized.
- **Taste:** We were unable to identify an indicator for predicting water taste. Platikanov et al. (2013) identified ions with a positive effect (Ca^{2+}, Mg^{2+}, SO_4^{2-} and HCO_3^-) and with a negative effect (Na^+, K^+ and Cl^-) on drinking water taste using trained panellists. The total dissolved solids (TDS) also affected drinking water taste with preferred values around 200-400 mg/L (Platikanov et al., 2013), which was not considered further, since TDS after softening depends on the source water TDS that may either be below, within or above the optimal range.
- **Organic and inorganic contaminants** are unwanted. We considered the overall potential of the technologies to act as barriers towards pesticides and heavy metals.

3.2. Technologies for centralised drinking water softening

Technologies for full-scale drinking water softening falls into three types depending on the mechanism for hardness removal (Fig. 2):

- Precipitation technologies: hardness ions are removed as solid minerals due to precipitation
- Ion exchange technologies: hardness ions are exchanged with other ions (counter ions) into the water
- Membrane separation technologies: hardness ions are separated from the water using semi-permeable membranes

The technologies can be further subdivided within each mechanism: Precipitation can e.g. be divided into pellet softening and lime/soda-ash softening (Fig. 2). Different choices of precipitation chemical, type of ion exchange resin and type of membrane form the softening technology. The treatment process and design affects the reactions for hardness removal and accordingly the final water quality in terms of e.g. addition of ions due to chemical addition (Table 4).

The chemical reactions reflect the main reaction for hardness removal and parameters directly related to hardness removal, but the overall changes of the water quality also depend on the process design, chemical equilibria limitations, membrane type, other ions precipitating with $CaCO_3$ etc. (Tang et al., 2019a; van der Bruggen and Vandecasteele, 2003; van Dijk and Wilms, 1991). Consequently, the chemical reactions in Table 4 only predict the major changes to water quality actually observed in full-scale softening.

3.3. Changes to water quality parameters

To evaluate the changes to the selected indicators (Table 3) in the third step of our framework (Fig. 1), we included changes to water quality observed in (i.e. from chemical water analyses) pilot-scale and full-scale drinking water softening in literature (Supporting Information A). Literature reporting of changes to water quality varies depending on the softening technology (Fig. 3).

In the precipitation technologies and weak-acid cation exchange, limitations in chemical equilibria or the process design itself establishes a de facto achievable minimum concentration (Fig. 3). For instance, if pH is too high in pellet softening, $CaCO_3$ will precipitate in the water phase (homogenous nucleation) and not onto pellets, resulting in a minimum achievable concentration (van Dijk and Wilms, 1991).

In membrane separation the removal of ions is typically quantified by the “rejection”, which is the percentage removal from the feed/influent water stream (Table 3). The removal of ions by membrane separation depends on e.g. the membrane characteristics and feed water composition (Shahmansouri and Bellona, 2015). Particularly nanofiltration membranes are highly selective and rejects divalent ions to a higher degree than monovalent ions (Nasr et al., 2013). Consequently, the reported rejections for a single water quality parameter may vary from case to case.

In ion exchange, ions released from the resin into the water (i.e. counter ions) are added to the water in molar concentrations equivalent to the ions adsorbed to the resin (Clifford, 1999; Höll and Hagen, 2002). Ions are also added to the water in the precipitation technologies in concentrations equivalent to the chemical dosage (de Moel et al., 2006). We report equivalent effects on water quality parameters as the molar change based on either the softening depth ($X_{\text{softening depth}}$) or chemical dosage ($X_{\text{chemical added}}$).

3.3.1. Hardness removal

Some of the included softening technologies remove total hardness, whereas others only remove carbonate hardness. Hu et al. (2018) reported a Ca^{2+} removal efficiency of 90 % in a new pellet reactor design circulating the water and concluded that it was more efficient than the full-scale pellet softening in the Netherlands reported by Hofman et al.

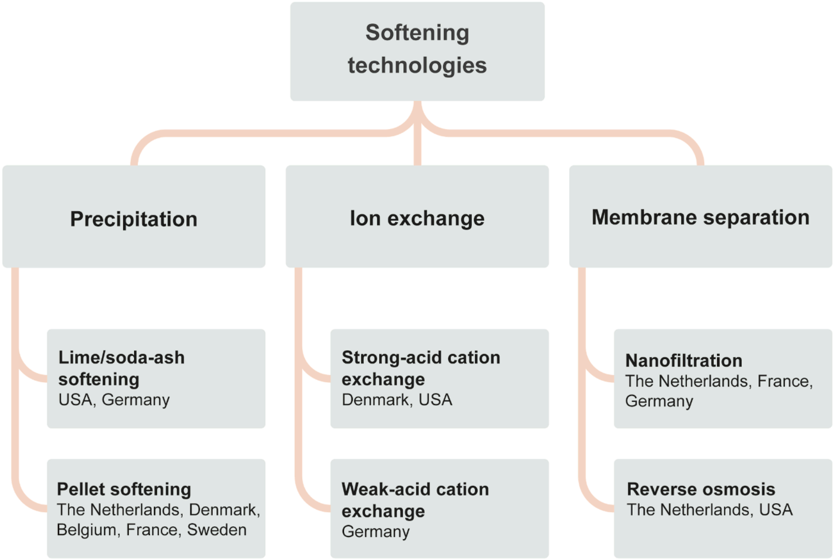

Fig. 2. Technologies for centralized drinking water softening grouped by the mechanism for hardness removal including examples of countries with full-scale application. Data from: (Bakshi et al., 2021; Höll and Hagen, 2002; Mons et al., 2007; Sydvatten, 2016; Tårnby Forsyning, 2017).

Table 4
Softening technologies including a brief description of the main principle for hardness removal, main design options and chemical reaction for hardness removal if applicable (Benefield and Morgan, 1999; Clifford, 1999; de Moel et al., 2006; Höll and Hagen, 2002; Taylor and Wiesner, 1999).

Softening technology	Principle for hardness removal	Main design option affecting water quality	Reaction
Lime/soda-ash softening	Precipitation and flocculation of calcium and magnesium removed as sludge. pH is increased by adding a chemical base (either $Ca(OH)_2$, Na_2CO_3 or a combination of the two).	Calcium hydroxide (lime, $Ca(OH)_2$)	$Ca^{2+}+2HCO_3^- + Ca(OH)_2 \rightarrow 2CaCO_3\downarrow + 2H_2O$ $Mg^{2+}+2HCO_3^- + 2Ca(OH)_2 \rightarrow 2CaCO_3\downarrow + Mg(OH)_2\downarrow + 2H_2O$
		Sodium carbonate (soda-ash, Na_2CO_3)	$CaSO_4 + Na_2CO_3 \rightarrow CaCO_3\downarrow + Na_2SO_4$
Pellet softening	Precipitation onto seeding material in a fluidized bed reactor with $CaCO_3$ removed as pellets. pH is increased by adding a chemical base as above.	Sodium hydroxide (caustic soda, NaOH)	$NaOH + HCO_3^- + Ca^{2+} \rightarrow CaCO_3\downarrow + Na^+ + H_2O$
		Calcium hydroxide (lime, $Ca(OH)_2$)	$Ca^{2+}+2HCO_3^- + Ca(OH)_2 \rightarrow 2CaCO_3\downarrow + 2H_2O$
Ion exchange	Exchange of hardness ions with sodium (strong-acid ion exchange) or $H_2O + CO_2$ (carbon dioxide regenerated weak-acid ion exchange). Regenerating of the ion exchange resin using e.g. NaCl or CO_2	Strong-acid ion exchange	$\begin{matrix}Ca^{2+}\\Mg^{2+}\end{matrix} + Na_2 - R_c \rightarrow \begin{matrix}Ca - R_c\\Mg - R_c\end{matrix} + 2Na^+$
		Weak-acid ion exchange (carbon dioxide regenerated)	$\begin{matrix}R_c - (COOH)_2\\+\ Ca^{2+}+SO_4^{2-}\\R_a - (HCO_3^-)_2\end{matrix} \leftrightarrow \begin{matrix}R_C - (COO^-)_2Ca^{2+}\\+\ 2H_2O\ +\ 2CO_2\\R_a - SO_4^{2-}\end{matrix}$
Membrane separation	Separation of hardness ions by a semi-permeable membrane allowing water and other water constituents to pass into the permeate stream.	Nanofiltration or reverse osmosis	$Rejection = 1 - \frac{C_{permeate}}{C_{feed}}$ Where C = solute concentration

(2006) that removed about 50 % of the Ca^{2+}. Hu et al. (2018) softened water with a relatively high temperature (18-21°C), which increases $CaCO_3$ precipitation (van Schagen et al., 2008), and did not consider that removal in conventional Dutch pellet reactors also was limited by targets set for minimum Ca^{2+} in the effluent (Hofman et al., 2007). This example illustrates the importance of reporting water quality changes in transparent and comparable manners.

Nanofiltration, reverse osmosis and strong-acid cation exchange remove nearly all hardness from drinking water. In practice, a water stream is typically by-passed the softening process and is mixed with the softened water to reach the target hardness. Lime/soda-ash softening can be operated at higher pH (> 10.8) than pellet softening and can consequently remove Mg^{2+} as $Mg(OH)_2$ in addition to Ca^{2+} (Benefield and Morgan, 1999; Hammer and Hammer, 2008). For water types with high Mg^{2+} hardness, it may not be possible to achieve the desired softening depth with the precipitation technologies operated at $pH < 10.8$. In weak-acid cation exchange, the regeneration frequency, and thereby water consumption, increases with increasing softening depth, which limit the feasible softening depth.

3.3.2. *Effects on pH, alkalinity and temperature*

The softening technologies may reduce, increase or have no effect on pH, and decrease or have no effect on alkalinity (Table 6). None of the technologies affect the water temperature. pH increases during lime/soda-ash softening and pellet softening due to the addition of a chemical base, although the removal of Ca^{2+} as $CaCO_3$ partially decreases pH (Table 4). When Ca^{2+} is removed as $CaCO_3$, ${CO_3}^{2-}$ is removed from the water, which reduces the alkalinity. Depending on the softening chemical, ${HCO_3}^-$ and Ca^{2+} are removed in a 1:1 or 2:1 molar ratio (Table 4). Thus, use of $Ca(OH)_2$ as the chemical base would maximize the removal of ${HCO_3}^-$, and use of Na_2CO_3 would avoid ${HCO_3}^-$ removal if necessary.

$CO_2(g)$ is not rejected by reverse osmosis and nanofiltration membranes, resulting in pH reduction. Alkalinity is nearly completely removed during reverse osmosis and partially removed by nanofiltration

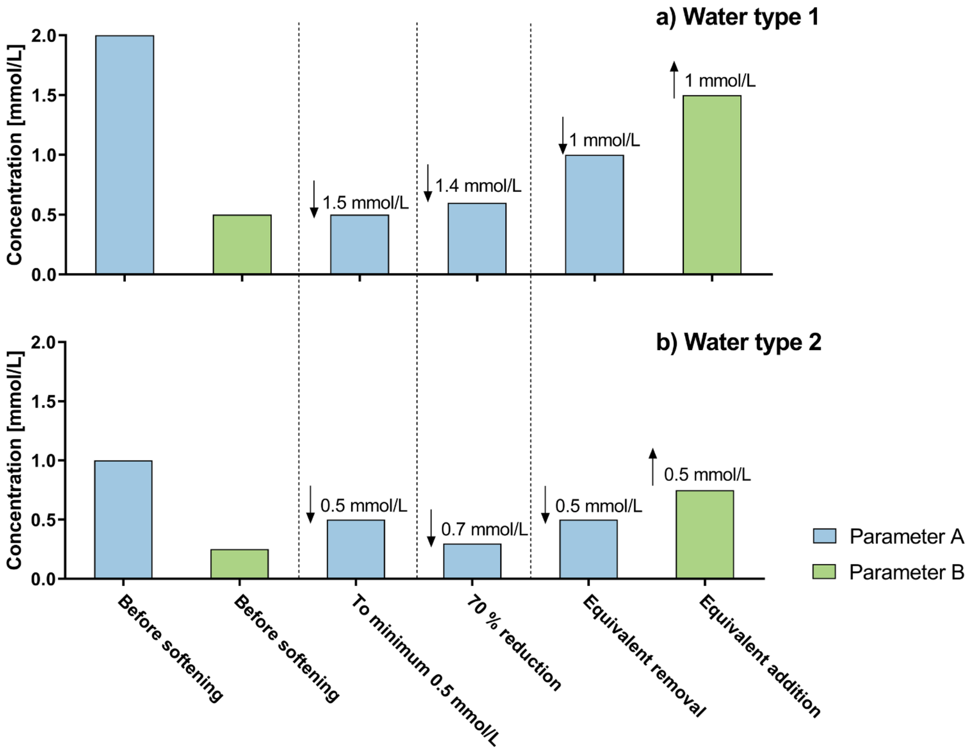

Fig. 3. Conceptual drawing of changes to water quality as a result of softening. Changes can be to a minimum concentration, a percent removal or equivalent removal and addition of ions.

(Table 6).

In weak-acid cation exchange, Ca^{2+} and Mg^{2+} are exchanged with H^+ that is removed from the water with HCO_3^- as CO_2. The removal of CO_2 from the water increases pH and decreases the alkalinity (Höll and Hagen, 2002). On the other hand, strong-acid cation exchange has no influence on neither pH nor alkalinity (Clifford, 1999).

3.3.3. *Effects on other ions (Na^+, K^+, Cl^-, F^- and SO_4^{2-})*

The WHO has not recommended guideline concentrations for K^+ or SO_4^{2-} due to a low risk towards human health at concentrations typically observed in drinking water (WHO, 2017). Consequently, removal of K^+ and SO_4^{2-} from drinking water has only little attention in literature.

Some ions can crystallize with $CaCO_3$ during precipitation and can be removed from the water together with hardness, whereas others, typically not forming carbonate minerals, remain in the water (Tang et al., 2019a). Tang et al. (2019a) observed < 4 % removal of K^+ during pellet softening with NaOH. We were unable to find data for the other precipitation technologies, but expect comparable results due to the similar mechanisms. During pellet softening with NaOH and soda-ash softening, Na^+ is added to the water in concentrations equivalent to the chemical dosage (Table 7). The Na^+ guideline concentration can be exceeded if the Na^+ in the source water is high, which can limit the chemical dosage, and thereby softening depth, or need for alternative technologies or process design.

Lime softening is reported to partially remove F^- as calcium fluoride (CaF_2). Due to the high solubility of CaF_2, the theoretical minimum concentration of F^- after softening is 7.5 mg/L (Ayoob et al., 2008), which is above the maximum guideline of 1.5 mg/L (WHO, 2017), and thereby not relevant for evaluation of the effects on dental caries expressed by DMF-S.

Na^+, K^+, Cl^- and F^- are monovalent ions that are nearly completely removed by reverse osmosis and partially removed by nanofiltration (Table 7). The removal depends e.g. on the feed water composition. E.g., Nasr et al. (2013) observed decreased F^- removal in nanofiltration with increasing Ca^{2+} feed concentrations, indicating that F^- removal is reduced in hard water types. SO_4^{2-} is divalent and can be nearly completely removed by both nanofiltration and reverse osmosis (Table 7).

In strong-acid cation exchange, Na^+ is exchanged from the resin into the drinking water, increasing the concentration in a 2:1 molar ratio with the divalent ions Ca^{2+} and Mg^{2+} (Berrios et al., 2014). No anions are removed from or added to the water during the process (Table 7). If weak-acid cation exchange is combined with strong-base anion exchange, anions are also removed with hardness. The strong-base anion exchange resin has higher affinity for divalent SO_4^{2-} compared to monovalent NO_3^- and Cl^-. Thus, the removal of NO_3^- and Cl^- is reduced at high SO_4^{2-} concentrations (Höll and Hagen, 2002; Kapoor and Viraraghavan, 1997)

3.3.4. *Effects on organic and inorganic contaminants*

Groundwater and surface water can contain various organic and inorganic contaminants which are harmful to human health and must be removed from the water (Rosborg and Kozisek, 2019). Removing the contaminants with water hardness in a single process step can be desirable from an economic perspective (van der Bruggen et al., 2001).

Both nanofiltration and reverse osmosis can reject organic contaminants, but the removal efficiency is complex and depends both on the compound, the water composition and the membrane characteristics (Bellona et al., 2004). Thus, the membrane type should be carefully selected for removal of the target compound (Plakas and Karabelas, 2008). Nanofiltration membranes are especially promising for removing organic contaminants together with water hardness and can be designed to remove organic compounds, but only partially remove water hardness to avoid remineralization after the process (van der Bruggen and Vandecasteele, 2003).

An investigation of removal of 12 pharmaceuticals and three oestrogens during pellet softening revealed only little removal of the pharmaceuticals (< 5.7 %), whereas the highly hydrophobic oestrogens were removed by 57-60 %, most likely due to sorption onto the pellets

(Maeng et al., 2016). This indicates that organic compounds may be removed during pellet softening due to sorption, but otherwise remain in the water. Similarly, lime softening has only limited removal of organic compounds compared to membrane softening (Bergman, 1995), with sorption and potentially co-precipitation being the main mechanisms for removal (Liao and Randtke, 1986).

Softening technologies that combine weak-acid cation exchange and anion exchange can potentially remove organic compounds, since anionic resins are known to remove organic contaminants (Hsu and Singer, 2009). However, the effects on specific compounds are unknown.

Nanofiltration and reverse osmosis may remove heavy metals such as arsenic (Choong et al., 2007) from drinking water together with hardness. The removal of a specific ions depends on e.g. the membrane type, pH as well as operating conditions such as trans-membrane pressure and crossflow velocity (Choong et al., 2007). Lime softening can be adapted to also remove arsenic from the water, but requires the addition of chlorine for oxidation of arsenic and pH > 10.5 (Choong et al., 2007). In pellet softening carbonate forming ions (e.g. nickel, iron and strontium) were partially removed, whereas ions not forming carbonates (arsenic and potassium) were not removed (Tang et al., 2019a). Ion exchange can remove other ions than hardness and has higher affinity for multivalent than monovalent ions (Barrios et al., 2008). If the resin has high affinity for a specific ion, it may decrease the efficiency of the regeneration process (Snoeyink et al., 1987).

Table 6
Observed effects on pH and alkalinity during drinking water softening. $X_{\text{chemical added}}$ = moles chemical added. ↓ = decrease. ↑ = increase, - = no change in concentration.

Technology	pH	Alkalinity	Reference(s)
Lime softening	↑ 1)	↓2$X_{\text{chemical added}}$	(Benefield and Morgan, 1999)
Soda-ash softening	↑ 1)	-	(Benefield and Morgan, 1999)
Pellet softening (Ca(OH)$_2$)	↑ 1)	↓2$X_{\text{chemical added}}$	(Benefield and Morgan, 1999)
Pellet softening (NaOH)	↑ 1)	↓ $X_{\text{chemical added}}$	(Tang et al., 2019a; van Dijk and Wilms, 1991)
Nanofiltration	↓ 2)	↓ 39-83 %	(Nasr et al., 2013; Saitua et al., 2011)
Reverse osmosis	↓ 2)	↓ 99 %	(Biesheuvel et al., 2020)
Strong-acid cation exchange	-	-	(Clifford, 1999; Hammer and Hammer, 2008)
Weak-acid cation exchange	↑ 0,2-0,6 pH units 2)	↓39-64 % 3)	(Höll and Hagen, 2002)

1) Depends on softening depth and chemical dosage
2) Depends on softening depth and subsequent stripping of CO_2
3) Depends on softening depth, reported as m-alkalinity

3.4. Effects on water quality indicators

The influence of softening technologies on the selected water quality indicators was evaluated from the observed changes to the water quality parameters (Table 5, Table 6 and Table 7) in the fourth and final step of our framework (Fig. 1). The evaluation of each indicator is available in the Supporting Information B. The evaluation included only the overall effect on the indicators and not absolute and relative changes, since this depends on the specific water type and process design. For example, the Larson Ratio calculated from the concentrations of Cl^-, SO_4^{2-} and HCO_3^- should be as low as possible to avoid corrosion in steel pipes and installations (Table 3). Increasing concentrations of Cl^- and SO_4^{2-} increase the Larson Ratio, thereby affecting the indicator adversely, whereas increasing HCO_3^- decrease the Larson Ratio, and subsequently the corrosion potential. Lime softening and pellet softening do not affect SO_4^{2-} or Cl^- (Table 7), but reduce HCO_3^- (Table 6) and will consequently increase the Larson Ratio, which is an overall adverse effect (Table 8). Soda-ash softening and strong-acid cation exchange have no effect on HCO_3^-, SO_4^{2-} and Cl^- (Table 7 and Table 6) and thus no effect on the Larson Ratio. Finally, nanofiltration, reverse osmosis and weak-acid cation exchange can remove both SO_4^{2-}, Cl^- and HCO_3^- from the water. Consequently, the overall effect on the Larson ratio depends on the specific water type and process design, and is therefore indecisive (Table 8).

CCPP depends highly on pH (Tang et al., 2021), and in practice nanofiltration and reverse osmosis reduces pH (and thereby CCPP) which often result in undesired, negative CCPP values, requiring post-treatment (e.g. Metsämuuronen et al., 2014; Tang et al., 2019b). Thus, even though CCPP is reduced, the negative values often require post-treatment and it would be misleading to categorize them as an overall positive effect. To avoid this, we did not include pH in the evaluation of CCPP.

The precipitation technologies tend to affect indicators similarly, independent of the technology (Table 9). Lime and soda-ash softening can potentially remove Mg^{2+} and thereby adversely affect the risk of cardio-vascular diseases, which is not affected by pellet softening. The precipitation technologies all increase the Larson Ratio and hence have an adverse effect on steel corrosion, but a positive effect on both Cu and Pb release that are decreased (Table 9). Thus, the distribution system materials should be considered when designing a softening process based on precipitation.

All softening technologies have positive effects on soap use (hardness reduction), the potential for lime scaling (CCPP reduction) and atopic eczema (Ca^{2+} removal), but potentially adverse effects on dental health

Table 5
Observed hardness removal for softening technologies in full-scale and pilot-scale installations. ↓ = decrease.

Technology	Hardness removed	Observed removal		References
		Ca^{2+}	Mg^{2+}	
Lime softening	Carbonate hardness	↓ to 12-20 mg /L	↓ to 10 mg/L	(Benefield and Morgan, 1999; Hammer and Hammer, 2008)
Soda-ash softening	Total hardness	↓ to 12-20 mg /L	↓ to 10 mg/L	(Benefield and Morgan, 1999; Hammer and Hammer, 2008)
Pellet softening (Ca(OH)$_2$)	Carbonate hardness	↓ to 20 mg /L	↓ to 0-10 %	(KWR, 2003; Ruhland and Jekel, 2004)
Pellet softening (NaOH)	Carbonate hardness	↓ to 20 mg /L	↓ to 0-12 %	(KWR, 2003; Ruhland and Jekel, 2004; Tang et al., 2019a)
Nanofiltration	Total hardness	↓ 43-99 %	↓ 48-99 %	(Gorenflo et al., 2003; van der Bruggen et al., 2001; van der Bruggen and Vandecasteele, 2003; Wesolowska et al., 2004)
Reverse osmosis	Total hardness	↓ 100 %	↓ 100 %	(Biesheuvel et al., 2020)
Strong-acid cation exchange	Total hardness	↓ > 98 %	↓ > 98 %	(Hammer and Hammer, 2008)
Weak-acid cation exchange	Carbonate hardness or total hardness[1]	↓ to 66-78 mg/L	↓ to 11-18 mg/L	(Höll and Hagen, 2002)

[1] Total hardness if combined with strong-base anion exchange

Table 7
Observed changes to Na^+, K^+, Cl^-, F^- and SO_4^{2-} during full-scale and pilot-scale drinking water softening. $X_{\text{chemical added}}$ = moles chemical added. X_{hardness} = water hardness removal in moles. ↓ = decrease. ↑ = increase. - = no change in concentration. NA = data not available.

Technology	Na^+	K^+	Cl^-	F^-	SO_4^{2-}	Reference
Lime softening	-	NA	-	↓ 7.5 mg/L	-	(Ayoob et al., 2008; Benefield and Morgan, 1999; Kyser and Doucette, 2018)
Soda-ash softening	↑$2X_{\text{chemical added}}$	NA	-	NA	-	(Benefield and Morgan, 1999; Kyser and Doucette, 2018)
Pellet softening ($Ca(OH)_2$)	-	NA	-	NA	-	(de Moel et al., 2006; KWR, 2003)
Pellet softening (NaOH)	↑ $X_{\text{chemical added}}$	↓ 0-4 %	-	NA	-	(KWR, 2003; Tang et al., 2019a)
Nanofiltration	↓ 9-84 %	↓ 53 %	↓ 7-82 %	↓ 40-99 %	↓ 9- >99 %	(Nasr et al., 2013; Saitua et al., 2011; Santafé-Moros et al., 2005; Tahaikt et al., 2008; van der Bruggen et al., 2001)
Reverse osmosis	↓ 97 %	↓ 98 %	↓ > 99 %	↓ 85-95 %	↓ > 99 %	(Biesheuvel et al., 2020; Karunanithi et al., 2019)
Strong-acid cation exchange	↑ $2X_{\text{hardness}}$	-	-	-	-	(Berrios et al., 2014)
Weak-acid cation exchange	-	-	↓ 3-13 %	-	↓ 33-81 %	(Höll and Hagen, 2002)

(DMF-S) (Table 9), which is mainly due to the removal of Ca^{2+}.

The membrane separation processes remove many different ions, but the specific removal may vary, and consequently, it was not possible to predict the effects on steel corrosion (Larson Ratio), Pb-release, Cu-release or taste. The membrane technologies have adverse effects on cardiovascular diseases due to Mg^{2+} removal (Table 9).

The ion exchange technologies are based on the same mechanism, but affect the water quality indicators differently. Strong-acid ion exchange only affects a few ionic species resulting in no effect on the three corrosion indicators (Table 9). The effects from weak-acid cation exchange on the corrosion indicators were indecisive.

The softening technologies affect the remaining indicators differently, illustrating that the effects on the overall water quality is technology specific. Furthermore, none of the softening technologies have a positive effect on all of the included indicators, indicating that the softening technology choice must balance adverse and positive effects.

4. Discussion

Nowadays, softening is often implemented to increase consumer convenience by reducing lime scaling and soap use in households. We found that all the technologies reduce lime scaling (expressed by the CCPP) and soap use (expressed by hardness), but also alters the overall mineral composition of water affecting corrosion, health and taste predicted by indicators. We included eight indicators as well as taste and contaminant removal, but further indicators may be relevant to include, depending on e.g. the water type, distribution pipe materials and national standards for evaluating water quality. An evaluation of a broad spectrum of water quality indicators provided information on both positive and adverse effects from softening, which can support decisions when designing the softening process. To our knowledge, such framework has not previously been provided.

Table 8
Effects from the softening technologies on SO_4^{2-}, Cl^- and HCO_3^- (from Table 6 and Table 7) and overall effect on the Larson Ratio and potential for steel corrosion.

Technology	SO_4^{2-}	Cl^-	HCO_3^-	Overall effect on Larson Ratio
	Removal = good	*Removal = good*	*Addition = good*	
Lime softening	No effect	No effect	Adverse	Adverse
Soda ash softening	No effect	No effect	No effect	No effect
Pellet softening ($Ca(OH)_2$)	No effect	No effect	Adverse	Adverse
Pellet softening (NaOH)	No effect	No effect	Adverse	Adverse
Nanofiltration	Positive	Positive	Adverse	Indecisive
Reverse osmosis	Positive	Positive	Adverse	Indecisive
Strong-acid cation exchange	No effect	No effect	No effect	No effect
Weak-acid cation exchange	Positive	No effect	Adverse	Indecisive

The evaluation of softening technologies using water quality indicators can be further improved by:

- **Quantify indicators for each specific case:** We considered the overall positive or adverse effects from the softening technologies on the selected indicators. In practice, the indicators should be calculated based on the actual water type in question to compare the absolute effects from each technology. If possible, the indicators should be compared to targets that water quality managers may set for each case.
- **Consideration of the entire treatment train:** We evaluated the softening technologies as isolated units. However, in practice the softening technologies are part of a treatment train consisting of also e.g. filtration, aeration and pH adjustment that also affect the mineral composition of the drinking water (Tang et al., 2019b). Moreover, pre- and post-treatment may be necessary before and after softening to e.g. remove ionic species that can foul the softening unit or adjust pH after softening (Berrios et al., 2014; Tang et al., 2019b). The effects experienced in the distribution networks and by the consumers depend on the water quality leaving the drinking water treatment plant, which should form the basis when predicting the effects in specific cases.
- **Better understanding of relations between indicators and consequences:** Indicators are not perfect representations of the consequences they predict. For instance, corrosion covers a range of complex chemical and physical processes that are difficult to predict (Loewenthal et al., 2004). Likewise, the relationship between magnesium and cardio vascular diseases is not fully understood (WHO, 2017), resulting in only indicative suggested concentrations (Rosborg and Kozisek, 2019). We suggest that future research focus on the understanding between theoretical indicators and the phenomena they predict. Implementation of softening will especially benefit from improved understanding of the relationship between CCPP and lime scaling to maximize the positive effects from softening (Tang et al., 2021).

Historically, water treatment technologies have been evaluated based on technical and economic aspects, but nowadays environmental

Table 9

Typical effects from softening technologies on water quality indicators. + = positive effect compared to no water softening. – = Adverse effect compared to no water softening. ? = effect depends on the specific water type and process design. 0 = no effect compared to no water softening. NF = nanofiltration. RO = Reverse osmosis.

		Lime/soda-ash softening		Pellet softening		Membrane separation		Cation exchange	
Consequence	Indicator	*$Ca(OH)_2$*	*Na_2CO_3*	*$Ca(OH)_2$*	*NaOH*	*NF*	*RO*	*Strong-acid*	*Weak-acid*
Soap use	Hardness	+	+	+	+	+	+	+	+
Limescale	CCPP	+	+	+	+	+	+	+	+
Steel corrosion	Larson Ratio	–	–	–	–	?	?	0	?
Copper corrosion	Cu-release	+	+	+	+	?	?	0	?
Lead release	Pb-release	+	+	+	+	?	?	0	?
Caries	DMF-S	–	–	–	–	–	–	–	–
Atopic eczema	[Ca]	+	+	+	+	+	+	+	+
Cardio-vascular diseases	[Mg]	?[1]	?[1]	0	0	–	–	–	–
Taste	-	–	–	–	–	?	?	–	–

[1] If operated at $pH > 10.8$, lime and soda-ash softening also remove Mg^{2+} with an adverse effect on cardio-vascular diseases.

and societal effects are increasingly considered as well (Hamouda et al., 2009). Evaluation of eight water quality parameters commonly measured in drinking water in addition to water hardness (alkalinity, pH, temperature, SO_4^{2-}, Cl^-, F^-, Na^+ and K^+) allowed for a broad evaluation of effects on corrosion, health and taste contributing to better understanding of softening in an overall systems perspective. Our framework can be expanded with economic, environmental, technical and societal indicators already used for decision support to further optimize the implementation of softening. Water utilities can use this systematic approach and overview to make informed decisions, weight positive and adverse effects from softening, and justify the choice of softening technology to consumers and other stakeholders.

5. Conclusions

We evaluated the effects from centralized softening technologies on selected water quality indicators within soap use (hardness) and limescale (CCPP), corrosion (Larson Ratio and predicted Pb and Cu release), health (DMF-S, Ca^{2+} and Mg^{2+}), taste and the removal of contaminants using existing water quality indicators, and conclude that:

- Softening technologies can be divided into three types, depending on the hardness removal mechanism that determines the changes to the overall water quality and whether hardness can be fully or only partially removed from the water: Precipitation (lime, soda-ash and pellet softening), membrane separation (nanofiltration and reverse osmosis) and ion exchange (strong-acid and weak-acid cation exchange) technologies.
- All the included technologies have a potential positive effect on hardness reduction, CCPP and atopic eczema, but potential adverse effects on dental carries (expressed by DMF-S), due to reduction of the Ca^{2+} concentration. The technologies are expected to have an overall adverse effect on the water taste, except reverse osmosis and weak-acid cation exchange where it depends on the specific water type and softening depth.
- The precipitation technologies have a positive effect on predicted Cu- and Pb-release, but an adverse effect on steel corrosion expressed by the Larson ratio. Membrane separation and weak-acid cation exchange can have either an adverse, no or positive effect on the corrosion indicators, whereas strong-acid cation exchange is not expected to affect the included corrosion indicators.
- Both ion exchange and precipitation has the potential for removing some heavy metals, but especially membranes can potentially remove organic and inorganic contaminants thereby providing softening and contaminant removal in a single process step.

None of the included softening technologies result in only positive effects on the included water quality indicators. Better understanding of potential adverse effects from softening allows for planning of preventive measures. Water utilities can use our framework to balance the positive and adverse effects from softening, and thereby optimizing decision support and the implementation of softening.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

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