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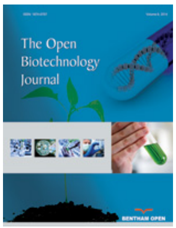

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REVIEW ARTICLE

Water an Eco-Friendly Crossroad in Green Extraction: An Overview

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Abstract: In this review, the function of water and the increasing role of water as a green solvent and co-extractant based on its application in ancient times and the need of environmental thinking have been discussed. A brief summary of various extraction methods for natural products, the application of conventional and innovative processes, based on water and future insights and perspectives considering water as an eco-friendly crossroad in green extraction have been reported. Taking into account also the issue of wastewater, this paper calls for-more effective use of water as a finite resource.

Keywords: Water, Green solvent, Extraction, Wastewater, Natural products, Eco-friendly.

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1. INTRODUCTION

Water is the most essential compound for the human body and life, in general [1]. It accounts for about 75% of the baby's body weight. In adults, water content represents approximately 60-65% and goes down to 55% with age [2, 3]. The intracellular space constitutes the largest human body compartment, holding approximately two-thirds of body fluid, therefore changes in water homeostasis predominantly affect cells; water excess leads to cellular swelling, and water deficit leads to cellular shrinkage. Due to the homeostatic mechanisms responding to the state of body water, its amount remains remarkably stable despite a huge range of water intake and a multitude of routes for water loss, including the respiratory and gastrointestinal tract, skin, and the kidneys [4].

The physical characteristics of water (Table **1**) make it compatible with living beings and determine its broad application as an odorless and colorless liquid with a density of 997 kg/m^3.

The structure of water consists of two hydrogen and one oxygen atoms (Fig. **1**). This is a polar molecule which means that there is an uneven distribution of electron density. Polar liquids, such as water, are excellent solvents able to participate in solutions capable of participating in solutions by reacting with other polar substances or ionic materials [5]. Thanks to its polarity, water can form electrostatic interactions (charge-based attractions) with other polar molecules and ions. Nonpolar molecules, like fats and oils, do not interact with water or form hydration shells. The Snyder's polarity index of water is 9.0, as compared to ethanol - 5.2 and ethyl acetate- 4.3 [6] arranging the common solvents based on the polarity, from least polar to most polar as follow: hexane < chloroform < ethylacetate < acetone < methanol < water. In chemistry, water is commonly used in mixtures with other solvents which change the polarity of the final solution. As a consequence, the ability to the dissolution of compounds changes [7].

2. WATER AS EXTRACTANT OF BIOLOGICALLY ACTIVE COMPOUNDS

Water is recognized as safe *a priori* (lit. "from the earlier"). It is an environmentally friendly compound and does not harm the environment whether in its production, use or disposal.

Biologically Active Compounds (BAC) are phytochemicals naturally found in nature - in plants, microbes, endophytic fungi, *etc*. The typical BAC in plants are produced as secondary metabolites, generated through various biological pathways in secondary metabolism processes, and play an important role in protecting plants from biotic or abiotic stress [9, 10].

In order to recover the valuable phytochemicals, efforts for their extraction have to be made. The researchers applied various extraction procedures aiming an appropriate and effective extraction. Therefore, the first step in the analysis of

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biological activity is the extraction of the compounds from the matrix and it is an appropriate way to transfer the beneficial compounds in an acceptable form for the researchers and consumers. With this point of view, the choice of extraction technique applied is of a critical matter, important for separating the desired natural products from the raw materials. Extraction methods may be carried out by using solvent, distillation pressing and sublimation according to the extraction principle, but the solvent extraction is the most widely used technique.

Nowadays, the extraction methods are widely explored in order to expand their efficiency by optimizing several parameters [11, 12]. The known and broadly used extraction procedures could be divided into two groups - conventional extraction methods and the modern one. The first includes maceration, percolation, reflux extraction, soxhlet extraction, Liquid-Liquid Extraction (LLE), solid-liquid extraction and mechanical shaking and usually organic solvents in large volumes for a long extraction time are involved. A major drawback is a fact that the obtained extracts often still require more concentration and purification steps, thus prolonging the entire analysis time. On the other hand, modern extraction methods include Microwave-Assisted Extraction (MAE), Accelerated Solvent Extraction (ASE), also known as Pressurized Liquid Extraction (PLE), and Supercritical Fluid Extraction (SFE), solid-phase extraction and micro-extraction, and surfactant-mediated techniques, which possess certain advantages. These are the reduction in organic solvent consumption and in sample degradation, elimination of additional sample clean-up and concentration steps before chromatographic analysis, improvement in extraction efficiency, selectivity, and kinetics of extraction. In addition, modern methods are suitable for automation and their usage for the extraction of plant materials is favored (Table **2**) [13].

A good solvent is characterized by its optimal extraction capacity and its potential to keep the stability of the chemical structure of the desired compounds [54]. Water is broadly applied as extractant of biologically active compounds [24, 55 - 61] and is commonly used in combination with alcohols [62 - 64]. Water, as a sole extractant, is applicable for the extraction of ions from plants and atmosphere [65, 66], as a mobile phase in compounds separation [67 - 69] and in conducting microwave and ultrasonic synthesis [70 - 72]. Water is used as a solvent in organic synthesis thus the chemical transformations occur mainly in an aqueous environment and the water-promoted reactions are numerous [73]. Furthermore, water is the medium in which gamma rays are applied with respect to water-soluble food components preservation [74] and transformations of organic matter induced by UV photolysis, hydroxyl radicals, chlorine radicals, and sulfate radicals in aqueous-phase UV-based advanced oxidation processes are conducted [75].

Fig. (1). Structure of water.

Table 1. Some physical characteristics of water [8].

Parameter	Unit	Value
GENERAL INFORMATION		
Name	Water	
CAS ♯	-	7732-18-5
IUPAC Name	-	dihydrogen oxide
Empirical formula	-	H_2O
Molecular mass	daltons	18.012
EC Number	-	231-791-2
PHYSICAL PROPERTIES		
State	-	liquid
Odor	-	odorless
Color	-	colorless
Boiling point	°C	100
Freezing point	°C	0
Density	kg/m^3	997
Refractive index at 20°C	-	1.333
Molar volume	cm^3/mol	18.0
Viscosity	mPas(cP)	0.89
Viscosity temperature	°C	25
Specific gravity	g/cm^3	1
Specific heat at 25°C	kJ/K.mol	75.31

2.1. Water-Based Extraction Methods

The extractability of biologically active compounds also depends on the technique employed. Several methods use water as a sole extractant, such as decoction, infusion and hydrodistillation. However, various techniques both conventional and modern are applicable to water as a solvent.

Decoction as a water-based extraction technique conducted by boiling is mostly suitable for extracting heat-stable compounds and hard plant materials. Georgieva *et al*. [24] obtained water extracts of *Achillea millefolium* by applying different extraction techniques aiming comparative evaluation. Decoction, in particular, was reported as the most potent one. Popova *et al*. [22] investigated *Melissa officinalis* antioxidant potential by comparing decoction with an infusion. The extraction was conducted by boiling plant material to dissolve the chemicals of the material in water for 30 minutes. Guimarãe *et al*. [25] investigated the bioactivity and characterized the organic acids and phenolic compounds of wild German chamomile by conducting decoction with boiled water for 5 min and left the sample at room temperature for 5 min. Ennaifer *et al*. [23] conducted decoction in the range from 8 to 20 min in an attempt to obtain a new water-soluble polysaccharide and antioxidant-rich extract of *Pelargonium graveolens*.

Infusions of medicinal plants were obtained by pouring the plant material with boiling water and let the mixture cool down for 20 min [55]. Infusions of six mistletoe tea bags extracted with water for 5 min while moving the tee bags up and down were investigated by Jäger *et al*. [19].

Using the “green solvent” water, Lante *et al*. [57] proposed a novel eco-friendly extraction procedure of isoflavones from soybean seeds comprising sonication and Saravana *et al*. [38] obtained seaweed polysaccharides from *Saccharina japonica* by subcritical water extraction. Hydrosols as co-products during water or steam distillation of plant material are the topic of the Lante and Tinello [56] research paper, while Zocca *et al*. [61] investigated the effects of *Brassicacaea* processing water in controlling enzymatic browning due to polyphenol oxidases from different plant sources.

Munir *et al*. [43] reported Subcritical Water Extraction (SWE) as a modern and advantageous extraction technique that can be an excellent alternative of the organic solvent when extracting bioactive compounds from onion skin. In addition, Todd and Baroutian [44] acknowledged the method as especially applicable in the extraction of medically and commercially important phenolic compounds from food and food by-products. Zakaria *et al*. [42] discussed the applications of SWE in maximizing the recovery of *Chlorella sp.* phenolic content and antioxidant activity and outlined that the technique improved mass transfer rate and preserved the biological potency of the extracts. Awaluddin *et al*. [45] used subcritical water technology for the extraction of biochemical compounds from *Chlorella vulgaris* and conducted optimization using central composite design under varying process conditions of temperature (180-374 °C), extraction time (1-20 min), biomass particulate size (38-250 μm), and microalgal biomass loading (5-40 wt. %). In addition, SWE of high-value-added compounds of grape pomace was studied [38]. The author established that coupling the subcritical water with membrane technologies offers an innovative solution for the recovery of bioactive compounds.

Marić *et al*. [32] conducted ultrasound, microwave and enzyme extractions using water in an attempt to increase the pectin yield and quality, and reducing extraction time, temperature, use of toxic solvents and strong acidic conditions for pectin recovery from plant food wastes and by-products. Petkova *et al*. [46] performed microwave-assisted extraction with water at 700 W power and frequency 2450 MHz for 5 min with the intention to evaluate the content of biologically active compounds (phenolic, flavonoids and fructans) and antioxidant activity of medicinal plants. In addition, Mihaylova *et al*. [47] investigated Bulgarian medicinal plants and subjected the plant material to 2450 MHz frequency waves for 30 s at 800 W output power. The used solvent was water. Furthermore, microwave-assisted water extraction of plant compounds was discussed and recommended as advantageous over conventional methods of extraction in respect of natural compounds. The potential of the technique for industrial applications was outlined as well [76, 77].

Table 2. A brief summary of various extraction methods for natural products [14, 15].

Method	Solvent	Temperature	Pressure	Time	Volume of Organic Solvent Consumed	Polarity of Natural Products Extracted
Soxhlet extraction [16, 17]	Methanol, ethanol, or mixture of alcohol and water	Under heat depending on solvent used	Atmospheric	Long	Moderate	Dependent on extracting solvent
Maceration [17 - 19]	Methanol, ethanol, or mixture of alcohol and water	Room temperature	Atmospheric	Long	Large	Dependent on extracting solvent
Percolation [20, 21]	Water, aqueous and non-aqueous solvents	Room temperature, occasionally under heat	Atmospheric	Long	Large	Dependent on extracting solvent
Decoction [22 - 26]	Water	Under heat	Atmospheric	Moderate	None	Polar compounds
Infusion [19, 26, 27]	Water	Under heat	Atmospheric	Short	Moderate	Polar compounds
Ultrasound-assisted extraction [28 - 32]	Methanol, ethanol, or mixture of alcohol and water	Room temperature, occasionally Under heat	Atmospheric	Short	Moderate	Dependent on extracting solvent
Pressurized liquid extraction [33 - 35]	Water, aqueous and non-aqueous solvents	Under heat	High	Short	Small	Dependent on extracting solvent
Reflux extraction [26, 36]	Aqueous and non-aqueous solvents	Under heat	Atmospheric	Moderate	Moderate	Water, aqueous and non-aqueous solvents
Supercritical fluid extraction [37 - 40]	Supercritical fluid (usually S-CO_2), sometimes with modifier	Near room temperature	High	Short	None or small	Nonpolar to moderate polar compounds
Subcritical water extraction [41 - 45]	Water	Under heat	High	Moderate	Small	Nonpolar to moderate polar compounds
Microwave assisted extraction [32, 46, 47]	Water, aqueous and non-aqueous solvents	Room temperature, occasionally under heat	Atmospheric	Short	None or moderate	Water, aqueous and non-aqueous solvents
Enzyme assisted extraction [32, 48 - 51]	Water, aqueous and non-aqueous solvents	Room temperature, or heated after enzyme treatment	Atmospheric	Moderate	Moderate	Water, aqueous and non-aqueous solvents
Hydro distillation and steam distillation [52, 53]	Water	Room temperature	Atmospheric	Long	None	Essential oil (usually non-polar)

Segovia *et al*. [31] recover polyphenols of avocado seeds with ultrasound-water extraction at different temperatures (20-60 °C) for 45 min and 40 kHz by column extractor. Lante and Friso [59] have demonstrated the use of ultrasound to extract catechins from green tea leaves with improved epigallocatechin-3-gallate yield, whereas Jäger *et al.* [19] subjected on maceration with water six mistletoe tea bags comparing the technical efficiency with infusion.

Hot water percolation was reported as a rapid soil extraction method conducted with hot water (102-105 °C) at 120-150 kPa pressure [20]. In addition, Hardouin *et al.* [49] applied enzyme-assisted extraction for the production of antiviral and antioxidant extracts from the green seaweed *Ulva armoricana* whereas endo-protease treatments significantly increased the extraction yields. Enzymatic release of phenolic compounds from pomace remaining from black currant (*Ribes nigrum*) when using commercial pectinolytic enzyme preparations and protease treatment was reported as well [50].

Nasardin *et al*. [52] investigated the hydrodistillation technique by obtaining agarwood oil and Richter and Schnellenber [53] recovered essential oils of aromatic plants using the same method, which is one of the oldest and easiest methods [78] for the extraction of essential oils.

Dhanani *et al*. [26] explored refluxing as a conventional method for *Withania somnifera* extraction with water for about 5 h at 100 °C. Ghasemzadeh and Jaafar [36] conducted optimization of the conditions for the reflux extraction of Pandan (*Pandanus amaryllifolius* Roxb.) in order to achieve a high content of total flavonoids, total phenolics, and high antioxidant capacity in the extracts.

Bocian and Kzreminska [68] followed the idea of green chemistry and used water as a mobile phase in compounds separation by liquid chromatography using polar-embedded stationary phases. Hadjikinova *et al*. [67] used water as a mobile phase by the development and validation of an HPLC-RID method for the determination of sugars and polyols.

Polo *et al*. [72] synthesized pyrazolo[3,4-*b*] pyridine derivatives trough one-pot condensation of 3-methyl-1-phenyl-1*H*-pyrazolo-5-amine (1), formaldehyde (as parafor-maldehyde) (2) and β-diketones (3) under microwave irradiation in aqueous media catalyzed by $InCl_3$.

Walinga *et al*. [65] dried plant material was subjected to water extraction by shaking for 30 min at ambient temperature. The filtrate was used for particular/specific analyses of Cl^-, NO_2^-, NO_3^- and SO_4^{2-}. Farren *et al*. [66] used water extraction for chemical characterisation of water-soluble ions in aerosol over the East coast of peninsular Malaysia. The authors evaluated the content of Cl^-, NO^{-2}, NO^{-3}, PO_3^{-4}, SO_2^{-4}, CH_3SO^{-3}, $C_2O_2^{-4}$, Na^+, NH^{+4}, K^+, Mg^{2+} and Ca^{2+} by ion chromatography system.

Lachos-Perez *et al*. [39] studied hydrolysis in subcritical water performance of sugarcane bagasse as an approach to solid residues characterization. The authors reported the highest reducing sugar yields at a temperature above 200 °C, with the highest reducing sugar yield reaching 15.5%.

2.2. Other Extraction Methods with Water as Co-Extractant

However, water has some limitations to being converted to a universal sustainable alternative for solvent extraction processes, such as the low solubility of apolar compounds and the energetic requirements to concentrate products [76, 79].

Therefore, Corrales *et al*. [28] extract anthocyanins from grape by-products with an ethanol-water mixture at 35 kHz, 70 °C for 1 h. Dahmoune *et al*. [29] used ultrasound power of 200 W and 24 kHz to obtain phenolic compounds from *P. lentiscus* leaves at 25 °C with water-ethanol mixture and Samavardhana *et al*. [30] obtained phenolic compounds and flavonoids in particular at 40 kHz, 320 W with 60% ethanol for 30 min.

Attempting to evaluate the antioxidant activity and the content of total phenolic, flavonoids and fructan content of eight herbs; de Hoyos-Martínez *et al*. [58], on the other hand, summarized and compared different tannins extraction methods and Pojić *et al*. [80] outlined the concept of zero food waste when associating the isolation of valuable proteins from sustainable sources and eco-innovative technologies.

Mihaylova and Shalow [18] conducted maceration with buffers as pretreatment in order to obtain quercetin from *S. japonica* followed by triple water-ethanol extraction at 70 °C, while Bandar *et al*. [17], on the other hand, investigated the influence of various factors on maceration process of *U. dioica* resuming the significant effect of solvent and time on the amount of the extracted compounds.

A mixture of catalyst altered water in combination with distilled water is discussed in a patented method for making herbal extracts using percolation [21].

Mihaylova *et al*. [35] obtained pressurised-liquid *Allium ursinum* extract with ethanol/water solution (85:15, v/v) containing 0.1M HCl (pH 3.5) evaluating the antioxidant potential. The same technique was applied as an innovative green technology for the investigation of six algae species from the Northwest of Spain by using five solvents of different polarities (hexane, ethyl acetate, acetone, ethanol and ethanol: water 50:50) at three temperatures (80, 120 and 160 °C) [33]. Furthermore, anthocyanins from the freeze-dried skin of a highly pigmented red wine grape were extracted with acidified water and acidified 60% methanol as solvents at 50 °C, 10.1 MPa, and 3 x 5 min extraction cycles [34].

Gligor *et al*. [51] recommended the enzyme-assisted extraction methods to increase the accessibility of polyphenolic compounds summarising a bunch of examples conducted at different optimal reaction conditions. Enzymes have been considered a useful tool for recovering biological actives substances from plants and stevioside from *Stevia rebaudiana* [48].

Kumoro *et al*. [16] investigated the effects of solvent properties (water, organic solvents and organic aqueous mixtures) on the conventional extraction using the Soxhlet method aiming a high extraction yield of *A. paniculata*. Bandar *et al*. [17] studied the same method for the extraction of bioactive compounds from *Urtica dioica* using different solvents (hexane, dichloromethane, acetone, ethanol and water) and established influence of the solvent type and the extraction time on the extraction process efficiency.

As a green approach, Rao *et al*. [71] conducted a reaction of 2-cyanothiomethylbenzimidazole 1 with aromatic aldehydes in water, under ultrasonic irradiation for 10-13 min. Mahdavinia *et al*. [70] reported a rapid and efficient synthesis of various aryl-14-*H*-dibenzo[*a,j*]xanthenes with excellent yields using ultrasonic irradiation and the condensation of an aldehyde and β-naphthol.

Subcritical water hydrolysis was applied to obtain antioxidant and antimicrobial hydrolysates from tuna skin and isolated collagen [40]. Different temperatures (150-300 °C) with pressure (50-100 bar) and reaction time (5 min) were employed to find the optimum condition. The degree of hydrolysis was highest at 250 °C for both Skin Hydrolysate (SH) and Collagen Hydrolysate (CH).

Saravana *et al*. [38] conducted subcritical water extraction of seaweed polysaccharides when combined deep eutectic solvent mixed with water at various concentrations. The optima conditions were established to be 150 °C, 19.85 bar, and 70% water content.

In conclusion, the authors applied various extraction techniques using water as an extractant when considering the most appropriate approach for bioactive compounds recovering. Anyway, a lot of specifics should be taken into account in order to achieve good performances and yield. It is clear that the selection of the solvent is crucial for the solvent extraction (Table **2**) and the right solvent leads to higher yield and process efficiency. Water seems to be the greenest solvent, being nontoxic, noncorrosive, non-flammable, environmentally benign, naturally abundant, and available at low cost. In correspondence, Prat *et al*. [81] recommended water in a green solvent selection guide ranking.

Different solvents could be used for extraction - water, methanol, ethanol, absolute acetone, various water-organic solvents mixtures, *etc*. In general, a solvent is a substance that dissolves another substance to form a solute. In this regard, water is capable of dissolving a variety of substances and is therefore assumed as a very good solvent. Since it dissolves more substances than any other liquid, water is known as the 'universal solvent'. In industrial processes, water is incapable of dissolving a large number of substances, necessitating the use of other solvents. These types of solvents are commonly carbon-based and are referred to as "industrial solvents". Solvent formulations that contain mixtures of different chemical agents are also used [82]. For instance, the most common solvents are widely recognized to be of great environmental concern. The reduction of their use is one of the most important aims of green chemistry.

The EU Rules on extraction solvents used in foodstuffs should be take into account primarily the human health requirements but also, be within the limits required for the protection of health, economic and technical needs. In this regard, Directive 2009/32/EC [83] is focused on the extraction solvents used or intended for use in the production of foodstuffs or food ingredients either in the EU or imported into the EU. In compliance with good manufacturing practices for all uses as safe extraction solvents are recognized propane, butane, ethyl acetate, ethanol, carbon dioxide, acetone and nitrous oxide.

3. WASTE WATER A PROBLEMATIC OUTPUT

All food processing in addition to food production is linked to the environmental issue of wastewater [84]. Water is extensively used in the industry. Apart from its important role in product formulations, water is used for cleaning, heating, cooling, steam generation, for the transport of substances or particulates. In 13 EU Member States for manufacturing of food products, the water use is reported to be 4.9 m^3/inhabitant (min. 1.7 m^3/inhabitant in Malta, max. 15.8 m^3/inhabitant in the Netherlands) in 2014 [85]. The primary production of food requires copious amounts of water. More than two-thirds of all freshwater abstraction worldwide (and up to 90% in some countries) goes towards food production [86]. Furthermore, from 1950 to 2000, industrial water consumption has increased from 20 to about 100 km^3/year [87].

Thus, water is a finite resource and there are uncertainties over the future availability of freshwater, and the rising industrial demand for it, many manufacturers have to re-evaluate water and consider it a critical resource [88]. Water management has recently become a major concern for many countries. The quality of water intended for human consumption is regulated by Directive 98/83/EC [89] and the hygiene of foodstuffs by Regulation (EC) No 852/2004 [90]. In the years of increasing water deficiency, it is crucial to apply a sustainable approach and be more responsible for water use regardless of the field of application. Furthermore, the pollution of the environment with various organic solvents as alternative extractants and of the water, in particular, is not the right choice to be of concern.

Adequate water management for achieving sustainable manufacturing is widely discussed [91]. A strong relationship between sustainable water management and economic development is needed and assumes the prime importance to ensure investment in the water sector while taking environmental concerns into account [92].

4. FUTURE INSIGHT AND PERSPECTIVE

Most water resources management problems are either local or regional. Expanding the knowledge on water management is of great importance including the current and future challenges and research directions. In this regard, technology is expected to play a key role in the future of the water sector.

As discussed on the World Economic Forum in Geneva in 2018, the majority of the world's current environmental problems can be traced back to industrialization. As the Fourth Industrial Revolution gathers pace, innovations are becoming faster, more efficient and more widely accessible than before. Technology is becoming increasingly connected, and a convergence of the digital, physical and biological realms is established [93].

The need of environmental thinking and responsible use and application of water either in the laboratory, in industry or at home is thus of critical matter. The efforts of the human should be focused on maintaining a consistent and clean water supply for use in all sectors. The latest technologies applied include 3D surface model analysis and visualization of glaciers, unmanned aerial vehicle video image classification for turfgrass mapping and irrigation planning, ground penetration radar for soil moisture estimation, the tropical rainfall measuring mission and the global precipitation measurement satellite rainfall measurements, storm hyetography analysis, rainfall-runoff and urban flooding simulation, and satellite radar and optical image classification for urban water bodies and flooding inundation. The application of those technologies is expected to greatly relieve the pressures on water resources and allow better mitigation of and adaptation to the disastrous impact of droughts and flooding [93, 94].

CONCLUSION

Water as natively presented in the human body is an essential compound for human life. It is the greenest possible solvent and a lot of processes may be re-thinking because the polar solvent water dissolves various bioactive compounds and is widely employed as solvent by different extraction methods, whether conventional or modern. From this point of view, water could be considered as an eco-friendly crossroad which opens a new way of working safely. The broad spectra of the application of water are increasing and therefore also the wastewater is an issue of great concern for environmental sustainability. As a finite resource on Earth, environmental concerns should be taken into account and need a change of perspective, first of all looking at the water with responsible and thoughtful use.

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CONFLICT OF INTEREST

The authors declare no conflict of interest, financial or otherwise.

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