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Ambient water quality criteria derived using probabilistic risk assessment

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Abstract

National recommendations for numeric human health ambient water quality criteria (AWQC) for toxic substances are derived by the US Environmental Protection Agency (USEPA) using a deterministic approach that combines point estimates for exposure, toxicity, and acceptable risk. In accordance with the Clean Water Act, states, territories, and authorized tribes must either adopt these recommendations or modify and replace them with criteria using an alternative, scientifically defensible method. Recent reports have criticized the deterministic approach, stating that it suffers from compounded conservatism by selecting upper percentiles or maximum values for multiple inputs and that it cannot directly determine what portion of the population a given criterion protects. As an alternative, probabilistic risk assessment (PRA) has been promoted as a more transparent and robust method for deriving AWQC. Probabilistic risk assessment offers several advantages over the deterministic approach. For example, PRA uses entire data distributions rather than upper-percentile point estimates to specify exposures, thereby reducing compounded conservatism. Additionally, because it links acceptable risk targets with specific segments of the exposed population, PRA-based AWQC demonstrably protects multiple subsets of the population. To date, no study has quantitatively compared deterministic and PRA approaches and resulting AWQC using national inputs consistent with USEPA guidance. This study introduces a PRA method for deriving AWQC and presents case studies to compare probabilistically derived AWQC with USEPA's 2015 recommendations. The methods and results of this work will help federal and state regulators, water quality managers, and stakeholders better understand available approaches to deriving AWQC and provide context to assumption- and method-specific differences between criteria. *Integr Environ Assess Manag* 2023;19:501–512. © 2022 The Authors. *Integrated Environmental Assessment and Management* published by Wiley Periodicals LLC on behalf of Society of Environmental Toxicology & Chemistry (SETAC).

KEYWORDS: Ambient water quality criteria; Probabilistic risk assessment; USEPA; Water quality standards

INTRODUCTION

The Clean Water Act (33 U.S.C. §§ 1251 et seq.), and specifically CFR 40 Part 131.4, tasks states, territories, and authorized tribes (hereafter referred to as "states") with the responsibility of reviewing, establishing, and revising water quality standards that are protective of designated uses, including the protection of aquatic life and human health. Accordingly, the US Environmental Protection Agency (USEPA) must subsequently review and approve or disapprove state-adopted water quality standards based on their adherence to guidelines outlined in CFR 40 Part 131.4 that require, among other considerations, that standards be developed using a sound scientific rationale. To aid states in the development and adoption of numeric criteria for the protection of human health from exposure to toxic pollutants through consumption of both aquatic organisms and water, the USEPA provides nationally recommended ambient human health water quality criteria (AWQC) for individual substances. In accordance with the requirements stated in CFR 40 Part 131.11, states may either adopt these numeric criteria in full, adopt modifications for all or specific substances based on site-specific conditions, or establish

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alternative numeric criteria using other scientifically defensible methods. The USEPA's current ambient AWQC (USEPA, 2015b) are derived using the same deterministic methodology (USEPA, 2000a) as the 2002 AWQC (USEPA, 2002) and that has remained largely unchanged since the 1980s (USEPA, 1986; Wiltse & Dellarco, 1996). This method is a risk-based approach that combines estimates of substance toxicity with assumptions about potential exposure and acceptable risk using deterministic equations that calculate the maximum allowable ambient water concentrations, or criteria, for various toxic pollutants.

Despite nationwide requirements and significant implications for the protection of human health, there remains a lack of literature exploring the deterministic method or alternative methods for deriving AWQC. Tatum et al. (2015) noted that the deterministic approach suffers from compounded conservatism, in which multiple conservative assumptions about exposure, acceptable risk, and substance-specific toxicity parameters compound and lead to criteria that are more protective than intended relative to the health protection targets established by state policies and USEPA. They suggested alleviating a portion of this compounded conservatism by either selecting less conservative point estimates for exposure parameters or using an alternative probabilistic risk assessment (PRA) approach (Tatum et al., 2015). Similarly, Idaho's Department of Environmental Quality (IDEQ) released a report (IDEQ, 2014) that explored PRA as an alternative methodology to the deterministic approach to criteria calculation. The IDEQ noted several shortcomings associated with the deterministic approach, including the compounded conservatism described above and the use of point estimates for exposure parameters that may not adequately characterize the diversity of Idaho's residents. The IDEQ also stated that, when using the deterministic approach, it is not possible to determine what percentile (or segment) of the population a given criterion protects at the specified acceptable health protection target. Although Idaho ultimately submitted deterministic criteria to USEPA for approval in 2015 (IDEQ, 2015), IDEQ (2014) concluded that a PRA approach could be used for deriving Idaho's AWQC in lieu of the deterministic method and that, if pursued, would result in criteria that better link specific health protection targets with specific segments of the population.

Florida is the only state that has submitted AWQC for USEPA approval using a PRA approach. The Florida Department of Environmental Protection (FDEP) developed an accompanying technical support document that described the PRA process for deriving AWQC and highlights the same issues associated with the deterministic approach enumerated by IDEQ and listed above (FDEP, 2016). Although FDEP later withdrew their proposed criteria, USEPA did not oppose the use of PRA to derive criteria (USEPA, 2015a).

Probabilistic methods have been used for decades by USEPA for assessing exposure. For example, USEPA (1989) and Smith (1994) describe probabilistic Monte Carlo methods used for risk assessment associated with exposure at Superfund sites, and the 1992 Guidelines for Exposure Assessment also discuss probabilistic appoaches (USEPA, 1992). The USEPA's *Options for development of parametric probability distributions for exposure factors* (USEPA, 2000b), as well as its subsequent Exposure Factors Handbook (USEPA, 2011), includes distributions of exposure inputs. More recently, USEPA released a white paper outlining PRA methods and related case studies (USEPA, 2014b), which draws on previous probabilistic guidance for human health risk assessments produced by the Oregon Department of Environmental Quality (ODEQ, 1998). These data were subsequently used by FDEP to produce PRA-derived ambient AWQC. Despite USEPA's recognition of the benefits of probabilistic methods since the late 1980s and publication of guidance for use of PRA in the Superfund program in 2001 (USEPA, 2001), USEPA has yet to release any guidance supporting derivation of AWQC using a PRA approach.

Since USEPA's release of the 2015 AWQC (USEPA, 2015b), only a few states have fully adopted USEPA's national recommendations. Although this low adoption among states may stem from a lack of state resources, it could also signal a level of apprehension among states regarding USEPA's 2015 AWQC, as well as uncertainty about what derivation methods and assumptions are considered scientifically defensible and, therefore, approvable by USEPA. The only other method that has been evaluated for deriving AWQC, apart from adoption of substance-specific maximum contaminant levels, is PRA, and only Idaho and Florida have written reports that considered the use of PRA for deriving AWQC (FDEP, 2016; IDEQ, 2014). No states have USEPA-approved AWQC derived using PRA.

Criteria that fail to demonstrate the link between a health protection target and the portion of the population protected at that target can provide either lesser or greater protection than intended by regulators and expected by stakeholders. Although providing lesser protection clearly fails to meet required health protection goals, criteria that are more protective than required by regulations may lead to the misallocation of limited public and private resources, including costs for waterbody remediation or compliance with discharge permit requirements. The purpose of this paper is to implement a PRA method to derive AWQC using equations and assumptions consistent with USEPA's current guidelines for deriving AWQC and compare PRA-derived AWQC with USEPA's deterministically derived 2015 AWQC. Although previous studies have examined the inherent conservatism of USEPA's deterministic method for deriving AWQC and contrasted this generally with PRA, no studies have directly compared the resulting criteria using both approaches with nationally relevant inputs. As such, the methods and results of this work fill an important information gap and can be used to better understand the differences between PRA and deterministic approaches to producing scientifically defensible, transparent, and protective AWQC.

APPROACHES TO DERIVING AWQC

Deterministic method

The USEPA's deterministic approach to deriving AWQC is a risk-based framework that combines parameters for acceptable (or target) risk, substance-specific toxicity, and assumptions of exposure (USEPA, 2000a). Using these inputs, AWQC are calculated using the formulae shown below depending on whether the health endpoint of concern is noncancer (Equation 1) or cancer (Equation 2).

$$\text{AWQC Noncancer} = HQ \times \text{RfD} \times RSC \times \frac{BW}{\left[DI + \sum_{i=2}^{4}(FCR_i \times BAF_i)\right]} \times \frac{AT}{EF \times ED}, \quad (1)$$

$$\text{AWQC Cancer} = \frac{ELCR}{CSF} \times \frac{BW}{\left[DI + \sum_{i=2}^{4}(FCR_i \times BAF_i)\right]} \times \frac{AT}{EF \times ED}, \quad (2)$$

where *HQ* is the hazard quotient (unitless), RfD the reference dose (mg/kg-day), *RSC* the relative source contribution (unitless), *ELCR* the excess lifetime cancer risk (unitless), *CSF* the cancer slope factor (per mg/kg-day), *BW* the body weight (kg), *DI* the drinking intake (L/day), FCR_i the trophic level-specific fish consumption rates (kg/day), $i = 1, 2, 3, 4$, BAF_i the trophic level-specific bioaccumulation factors (L/kg), $i = 1, 2, 3, 4$, *AT* the averaging time (days), *EF* the exposure frequency (days/year), and *ED* the exposure duration (years).

If a substance exhibits both noncarcinogenic and carcinogenic effects, then the lower concentration resulting from the two equations becomes the AWQC. Two sets of criteria are included in USEPA's 2002 and 2015 national recommendations. The first are criteria to protect human health for consumption of water and organisms (Water and Organism criteria) and are applicable to waters designated as a water supply and for recreational use. The second are to protect human health for consumption of organisms only (Organism Only criteria) and are applicable to waters designated for recreational use but not as a water supply.

For the noncancer effect equation, the health protection target is the hazard quotient (*HQ*; unitless). The HQ is the ratio of the estimated dose (or exposure) to the allowable dose (i.e., the reference dose or RfD). When deriving AWQC, USEPA sets the HQ equal to 1 such that when the concentration of a substance in ambient water is equal to the AWQC, the dose from consuming water and organisms is equal to the RfD. For the cancer effect equation, the health protection target is the excess lifetime cancer risk (*ELCR*; unitless). The ELCR represents a theoretical incremental cancer risk associated with consuming water and organisms containing a specific substance at a concentration equal to the AWQC. The USEPA's 2015 AWQC use 1×10^{-6} (1-in-a-million) as the ELCR, and all states have currently adopted either 1×10^{-6} or 1×10^{-5} acceptable risk targets. USEPA guidance suggests that the ELCR be set at 1×10^{-6} or 1×10^{-5} for the general population provided that risk to the most highly exposed subpopulations does not exceed 1×10^{-4} (USEPA, 1999, 2020).

Exposure inputs used in USEPA's 2002 and 2015 national AWQC differ (Table 1) and, according to USEPA, reflect the use of more recent data to generate means and percentiles (USEPA, 2011). The exposure components in both Equations (1) and (2) are parameterized using point estimates for body weight (*BW*; kg), untreated drinking water intake (*DI*; L/day), and fish consumption rate (*FCR*; kg/day; Table 1). Bioaccumulation factors (*BAFs*; L/kg) characterize the potential for substances to accumulate in organisms. In USEPA's 2015 criteria updates (USEPA, 2015b), BAFs replaced bioconcentration factors (BCFs) used in USEPA (2002) and account for accumulation of a substance from the water column as well as the food web. The relative source contribution (*RSC*; unitless), which is only included in the noncancer effect Equation (1), varies between 0 and 1 and accounts for nonwater sources of exposure. Although discussed in USEPA's 2000 methodology (USEPA, 2000a), RSCs of less than 1 were not incorporated into most criteria until 2015. Finally, the exposure duration (*ED*; days) and exposure frequency (*EF*; days/year) designate the length and frequency of exposure, respectively. Averaging time (*AT*; days) is simply the duration over which exposures are assumed to occur. When deriving AWQC, USEPA assumes a lifetime (70 years) of daily exposure, equal to 25 550 days. This effectively assumes that an individual is exposed to water and fish from the regulated water body for his or her entire life (e.g., years).

In addition to the above parameters that are explicitly included in the equations used to derive AWQC, several implicit assumptions increase conservatism. For example, the AWQC derivation methodology assumes zero reduction

TABLE 1 Explicit exposure inputs used by the deterministic method to derive USEPA's nationally recommended human health water quality criteria

Year	Body weight (kg)	Untreated drinking water ingestion rate (L/day)	Fish consumption rate (g/day)[a]
2002	70 (mean)	2.0 (86th percentile)	17.5 (90th percentile)
2015	80 (mean)	2.4 (90th percentile)	22.0 (90th percentile)

Abbreviations: AWQC, ambient water quality criteria; BAF, bioaccumulation factor; FCR, fish consumption rate.
[a]USEPA's 2015 national AWQC use a FCR of 22 g/day when the BAF is constant across trophic levels (TLs). However, when TL-specific BAFs are used to derive an AWQC, TL-specific FCRs are also used (i.e., 7.6 g/day for TL2, 8.6 g/day for TL3, and 5.1 g/day for TL4), resulting in a total FCR of 21.3 g/day.

in the concentrations of substances in fish due to cooking. Such reductions can be 30%–50% for some compounds (Sherer & Price, 1993; Zabik et al., 1995). Additionally, the methodology assumes people consume untreated water and are consistently exposed to the maximum allowable concentrations in fish and water over their entire lifetime. The selection of upper-percentile point estimates for drinking water ingestion and fish consumption, and conservative estimates for toxicity combined with the conservative implicit assumptions, may lead to criteria that are more stringent than intended for the selected risk targets or required by regulations because the AWQC are not based on a realistic and representative distribution of underlying population risk (Tatum et al., 2015).

Probabilistic risk assessment

Probabilistic risk assessment methods use the same equations as the deterministic method (Equations 1 and 2) but rely on distributions to represent variability associated with exposure among a population or uncertainty surrounding a parameter's value for some inputs rather than point estimates, followed by an iterative calculation procedure to derive AWQC (Figure 1). The use of distributions does not result in biased protection of certain portions of the population (e.g., under- or overprotection of more susceptible populations such as children or the elderly). Rather, distributions incorporated into the PRA approach more accurately characterize the population for which the criteria are intended to protect and provide the basis for regulatory decisions regarding risk thresholds for various groups within the protected population. In addition, PRA methods require a priori selection of specific percentiles of the population targeted for protection as well as the allowable health protection targets (i.e., acceptable ELCR for cancer effects and acceptable HQ for noncancer effects) that correspond to those population percentiles. Ambient water quality criteria are then determined using an iterative process that generates distributions of risk using specific water concentrations. If the specified health protection targets are met for the targeted percentiles of the population, then the water concentration used during that calculation is identified as the AWQC. However, if the health protection targets are not met (i.e., risk at one or more of the population percentiles exceed the health protection target for the percentiles), a new water concentration is used to generate a new distribution of risk to determine if the health protection targets are met. The process is repeated until a water concentration is identified that meets the health protection target at all percentiles.

This PRA approach to deriving AWQC can be considered a single-objective, constrained optimization problem where the decision variable is the water concentration, and the objective is to maximize its value under the constraints of meeting the specified health protection target(s) at selected percentile(s) of the risk distribution. Numerous optimization algorithms are available (e.g., hypercube sampling, hill-climbing algorithms, genetic algorithms, particle swarm optimization), and any type could be used to solve for the concentration. An example of a PRA implementation procedure using linear regression is shown in Figure 2. Three different water concentrations are used to calculate three different distributions of risk using PRA. The allowable risks associated with one or more percentiles (e.g., 50th, 90th, 99th), which is specified by the user a priori, is identified for each distribution. The three pairs of water concentrations and associated allowable risks are used to generate a linear regression equation that relates water concentration and risk at the specified percentile of the risk distributions. The user-specified health protection target (either the ELCR or HQ) is then used with the regression equation to derive an AWQC by solving for the water concentration associated with that specified health protection target. The process can be repeated using different percentile and health protection target pairs to produce criteria protective of various percentiles of the population (subpopulations).

Each input used in the PRA derivation of AWQC can be either a point estimate or a full distribution, depending on available data and the intention of the user. Developing

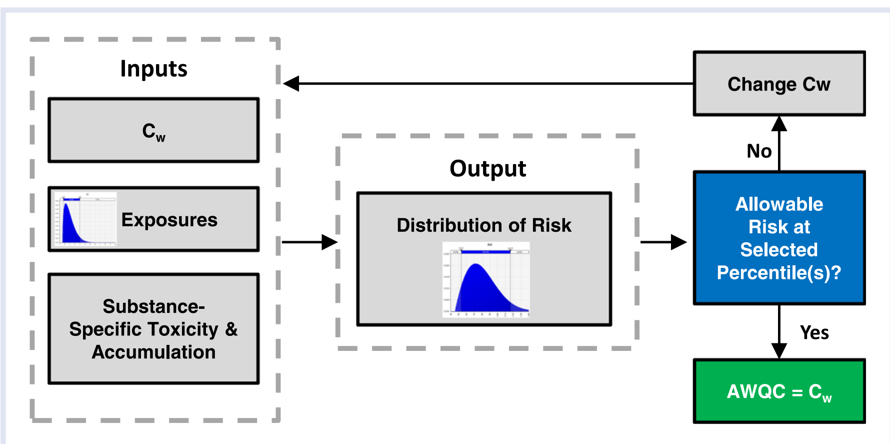

FIGURE 1 Overview of the probabilistic risk assessment (PRA) approach to deriving human health ambient water quality criteria (AWQC) water concentration values

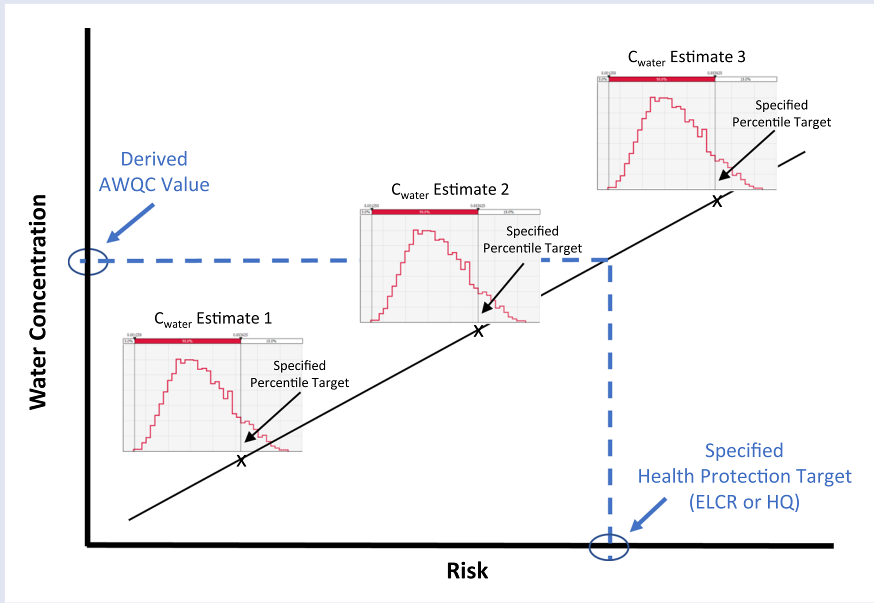

FIGURE 2 Graphical depiction of the probabilistic risk assessment (PRA) process for deriving ambient water quality criteria (AWQC) using a linear regression approach

parameter distributions for use in PRA has, at times, been viewed as a challenge because of a perceived lack of robust sources of data. However, in many cases, robust statistical data are available and can be used to characterize the variability and uncertainty in parameters that determine AWQC. For example, USEPA's *Exposure Factors Handbook* contains distributions of national data for drinking water ingestion rate, body weight, and fish consumption rate (USEPA, 2011). These distributions are the basis for the point estimate inputs in USEPA's 2015 national AWQC, where specific percentiles of the distributions are chosen as point estimate inputs.

Finally, the unique characteristic that distinguishes PRA-based AWQC from deterministic-derived criteria and makes their protectiveness transparent is linking specific segments (percentiles) of the population with specific health protection targets. Establishment of health protection targets by states or USEPA is largely a policy decision. For carcinogenic effects, FDEP considered risks acceptable when the 50th percentile of the population (representative of the general population) had a risk equal to (or less than) 1×10^{-6}, the 90th percentile had a risk equal to (or less than) 1×10^{-5}, and the 99th percentile (highest exposed subpopulation) had a risk equal to (or less than) 1×10^{-4} (FDEP, 2016). The FDEP then selected the most stringent, lowest concentration resulting from the three scenarios as the final AWQC to be protective of all portions of the population. In comparison, ODEQ considers cancer risks acceptable when the 90th percentile has an ELCR equal to or less than 1×10^{-6} and the 95th percentile has an ELCR equal to or less than 1×10^{-5} (ODEQ, 1998). Both of these acceptable risk approaches may be viewed as consistent with USEPA's recommendation "that both 10^{-6} and 10^{-5} may be acceptable for the general population and that highly exposed populations should not exceed a 10^{-4} risk level" (USEPA, 2000a). Yet, the ELCR at the 90th percentile considered allowable by ODEQ is tenfold lower (more stringent) than FDEP, and it is not clear whether USEPA would consider the 90th percentile representative of the general population but rather a highly exposed subset of the general population, referred to as the reasonable maximum exposure (RME; USEPA, 2001).

For noncarcinogenic effects, ODEQ's PRA guidance considers risks acceptable when the HQ is less than 1 for the 50th and 90th percentiles of the population and less than 10 for the 95th percentile of the population (ODEQ, 1998). Similarly, to derive noncarcinogenic AWQC for the noncancer endpoint, FDEP set the HQ to 1 for the 90th percentile (FDEP, 2016). Alternative combinations of health protection targets and population percentiles can also be implemented in PRA depending on policy goals.

METHODS

"Water and Organism" and "Organism Only" AWQC were generated for the 94 substances in USEPA's updated criteria recommendations (USEPA, 2015a, 2015b) using deterministic and PRA approaches. Both the deterministic and PRA approach used inputs for exposure, toxicity, and risk consistent with USEPA's 2015 national AWQC (USEPA, 2015b). The deterministic method derivation assumed 2.4 L/day drinking water intake, 80 kg body weight, and substance-specific RSC, BAF, and toxicity parameters. Following USEPA's methodology, fish consumption rates of 22 g/day were used when BAFs were constant across trophic levels, and trophic-level-specific fish consumption rates were used when BAFs varied across trophic levels for individual substances (i.e., 7.6 g/day for TL2, 8.6 g/day for TL3, and 5.1 g/day for TL4, totaling 21.3 g/day; USEPA, 2015b). The

TABLE 2 National exposure distributions obtained from USEPA's analyses of National Health and Nutrition Examination Survey (NHANES) data

	Distribution type	Mean	10th	25th	50th	75th	90th	95th	99th
Drinking water consumption (L/day)	Lognormal	1.043	–	0.227	0.787	1.577	2.414	2.958	4.405
Body weight (kg)	Weibull	80.8	57.4	66.1	77.9	92.4	107.0	118.0	–
Fish consumption (g/day)	Lognormal	–	–	–	5.0	11.4	22.0	31.8	61.1

PRA approach used distributions of exposure inputs for drinking water, body weight, and fish consumption obtained from the National Health and Nutrition Examination Survey (NHANES) (Table 2). Drinking water consumption data were based on NHANES 2003–2006 per capita estimates of combined direct and indirect ingestion of community water for individuals aged 21 years and over (see Table 3–23 in USEPA, 2011). The 90th percentile of this distribution corresponds to the 2.4 L/day used in USEPA's deterministic method for deriving AWQC. The distribution for body weight was based on USEPA's analysis of NHANES 1999–1996 data for both males and females aged between 30 and under 40 years (see Table 8–3 in USEPA, 2011). This age range was chosen because the mean value matched USEPA's chosen mean value used in the deterministic method. The fish consumption distribution was based on NHANES data from 2003 to 2010 that included consumption rates of fish and shellfish from inland and nearshore marine waters for the US adult population 21 years of age and older. The 90th percentile of this distribution is 22.0 grams per day (see Table 9a in USEPA, 2014a) as used by USEPA to derive the 2015 national AWQC. Substance-specific toxicity parameters (i.e., RfD and CSF) and remaining exposure parameters (i.e., RSC and BAF) were consistent with those used to derive USEPA's 2015 AWQC (USEPA, 2015b; Supporting Information: Table A1). Example substances with both carcinogenic and noncarcinogenic effects and a range of BAFs are shown in Table 3.

For this analysis, both the deterministic and PRA methods used predetermined health protection targets in the carcinogenic and noncarcinogenic effect equations (ELCR and HQ, respectively). To derive criteria protective of carcinogenic effects, the deterministic acceptable ELCR target was set at 1×10^{-6} to match the value used to derive USEPA's 2015 AWQC (USEPA, 2015b). For the PRA method, criteria were derived using acceptable risk guidance from FDEP and ODEQ. Following ODEQ's approach (ODEQ, 1998), criteria were calculated for two pairs of acceptable risk-percentile targets, 1×10^{-6} for the 90th percentile and 1×10^{-5} for the 95th percentile, and the lowest criterion was chosen as protective of both targets. Consistent with FDEP's approach (FDEP, 2016), criteria were calculated for three pairs of acceptable risk-percentile targets, including 1×10^{-6} for the 50th percentile, 1×10^{-5} for the 90th percentile, and 1×10^{-4} for the 99th percentile. The final PRA-derived AWQC was then chosen as the lowest of the three criteria to be protective of all three.

To derive criteria protective of noncarcinogenic effects, the HQ in the deterministic method was set at 1, identical to that used to derive USEPA's 2015 AWQC (USEPA, 2015b). Probabilistic risk-assessment-based AWQC were derived following both ODEQ and FDEP acceptable risk guidance. The ODEQ assigns an HQ of 1 to the 50th and 90th percentiles, and an HQ of 10 at the 95th percentile (ODEQ, 1998). The final criterion protective of noncarcinogenic effects is the lowest criterion from the three percentile-target risk pairs and protective of all percentiles of the population. The FDEP assigns an HQ of 1 at the 90th percentile (FDEP, 2016) and is the basis for criteria protective of noncarcinogenic effects following FDEP acceptable risk guidance.

Excel software with the @Risk plug-in (Palisade, 2016) was used to construct continuous distributions using the percentiles in Table 2 using the @Risk "Fit Distributions to Data" tool. Drinking water consumption was represented as a generalized beta distribution, RiskBetaGeneral ($\alpha 1 = 1.5792$,

TABLE 3 Substance-specific inputs used to derive AWQC for selected carcinogenic and noncarcinogenic substances comparing deterministic versus probabilistic derivation methods

Substance	Cancer slope factor (mg/kg-day)	Reference dose (mg/kg-day)	Relative source contribution	BAF, TL2 (L/kg)	BAF, TL3 (L/kg)	BAF, TL4 (L/kg)
Benzo(*a*)pyrene	7.3	NA	NA	3900	3900	3900
Chlordane	0.35	0.0005	0.2	5300	44 000	60 000
Dieldrin	16	0.00005	0.2	14 000	210 000	410 000
1,1-Dichloroethylene	ND	0.05	0.2	2	2.4	2.6
Pyrene	ND	0.03	0.2	860	860	860
Dimethyl phthalate	ND	10	0.2	4000	4000	4000

Abbreviations: AWQC, ambient water quality criteria; BAF, bioaccumulation factor; NA, not applicable.

$\alpha2 = 9.282$, $\min = -0.43583$, $\max = 9.4839$), where $\alpha1$ and $\alpha2$ are shape parameters for a beta distribution, and the distribution is then scaled from the original [0, 1] range to instead have specified minimum and maximum values. Body weight was represented as a Weibull distribution using RiskWeibull ($\alpha = 1.8767$, $\beta = 39.604$, shift $= 45.388$), where α is the shape parameter, β is the scale parameter, and shift denotes a change in the domain of the corresponding distribution. Fish consumption rate was represented as a lognormal distribution RiskLognorm ($\mu = 0.01125$, $\sigma = 0.013467$, shift $= -0.0022134$), where μ is the mean, σ is the standard deviation, and shift denotes a change in the domain of the corresponding distribution. The fish consumption rate was partitioned into trophic-level-specific fish consumption rates (see footnote in Table 1). Criteria were then estimated for both deterministic and probabilistic methods for all 94 substances updated by USEPA (USEPA, 2015b). Criteria protective of carcinogenic and noncarcinogenic effects were calculated separately as described above, and the lower of the two was chosen to be the final AWQC. Several risk-percentile scenarios were implemented using PRA and compared with the deterministic approach to select substances exhibiting carcinogenic and noncarcinogenic effects. Then, criteria estimates were examined generally for all 94 substances updated by USEPA (USEPA, 2015b). Comparisons were expressed as a ratio of the PRA-based criterion to the deterministic-based criterion. Thus, a ratio of 2 indicates the PRA-based criterion for a substance was two times larger than the deterministic criterion. A ratio of 0.9 indicates the PRA-based criterion for a substance is 0.9 times (or 10%) lower than the deterministic criterion. Kernel density plots, which are nonparametric histograms that provide a continuous estimate of the relative frequency of values in a dataset, were used to visualize the relative magnitude and frequency of the criteria ratios between the deterministic and probabilistic AWQC across all substances.

RESULTS

Results are divided into general sections. The first presents criteria protective of carcinogenic and noncarcinogenic effects for select substances with the goal of illustrating how criteria for each substance are affected by changes in risk management paradigms (e.g., changes in acceptable risk levels and the percentiles of the population to which acceptable risk levels are applied; Tables 4 and 5). The second section compares deterministic and PRA criteria for all 94 substances whose AWQC were updated by USEPA in 2015 (Figures 3 and 4).

Criteria protective of carcinogenic effects

When the same acceptable ELCR (e.g., 10^{-6}) was applied to successively higher percentiles of the population, PRA-based criteria decreased (Table 4). Probabilistic risk assessment-based criteria also decreased when more stringent ELCRs were applied to the same subpopulation (e.g., changing the acceptable ELCR from 1×10^{-5} to 1×10^{-6} for the 90th percentile; Table 4). Using ODEQ acceptable risk guidance, criteria derived using an acceptable ELCR of 1×10^{-6} applied to the 90th percentile were lower than criteria derived by applying an acceptable ELCR of 1×10^{-5} to the 95th percentile for all compounds. As a result, all final PRA-based AWQC were equal to the criteria derived assuming a 1×10^{-6} acceptable ELCR applied to the 90th percentile. Following FDEP guidance (i.e., choosing the lowest criteria among the 1×10^{-6} ELCR/90th percentile, 1×10^{-5} ELCR/95th percentile, and 1×10^{-4} ELCR/99th percentile scenarios), the final PRA criteria were equal to the criteria derived assuming a 1×10^{-6} acceptable ELCR applied to the general population (the 50th percentile; Table 4).

Criteria protective of noncarcinogenic effects

The PRA-based criteria were consistently lower when the same HQ was applied to more highly exposed subpopulations (e.g., HQ = 1 applied at the 50th vs. 90th percentiles; Table 5). However, the increase in HQ from 1 to 10 for the 95th percentile, following ODEQ guidance, resulted in higher criteria although the 95th percentile represents a more highly exposed subset of the population than the 90th percentile. For both ODEQ and FDEP approaches, the lowest PRA-derived criteria resulted from an HQ target

TABLE 4 Water and Organism and Organism Only criteria driven by carcinogenic effects for select substances derived using deterministic and probabilistic methods

Criteria type	Criteria assumptions and substance	Deterministic		Probabilistic					Final criterion	
	Acceptable ELCR	10^{-6}	10^{-5}	10^{-6}		10^{-5}		10^{-4}		
	Target percentile	N/A		50th	90th	90th	95th	99th	ODEQ	FDEP
Water and Organism	Benzo(a)pyrene (ng/L)	0.12	1.24	0.53	0.12	1.2	0.85	4.22	0.12	0.53
	Chlordane (ng/L)	0.31	3.14	1.35	0.3	2.94	2.07	10.3	0.3	1.35
	Dieldrin (pg/L)	1.25	12.48	5.38	1.18	11.73	8.27	41.11	1.18	5.38
Organism Only	Benzo(a)pyrene (ng/L)	0.13	1.28	0.56	0.12	1.21	0.86	4.26	0.12	0.56
	Chlordane (ng/L)	0.32	3.15	1.35	0.3	2.94	2.08	10.31	0.3	1.35
	Dieldrin (pg/L)	1.25	12.49	5.39	1.18	11.73	8.27	41.11	1.18	5.39

Abbreviations: FDEP, Florida Department of Environmental Protection; ODEQ, Oregon Department of Environmental Quality.

TABLE 5 Water and Organism and Organism Only criteria driven by noncarcinogenic effects for select substances comparing deterministic versus probabilistic derivation methods

	Criteria assumptions and substance	Deterministic	Probabilistic				
	Target hazard quotient	1	1		10	Final criterion	
Criteria type	Target percentile	N/A	50th	90th	95th	ODEQ	FDEP
Water and Organism	Benzo(*a*)pyrene (ng/L)	0.33	0.98	0.31	2.46	0.31	0.31
	Chlordane (ng/L)	0.02	0.09	0.02	0.16	0.02	0.02
	Dieldrin (pg/L)	1.77	7.59	1.71	12.09	1.71	1.71
Organism Only	Benzo(*a*)pyrene (ng/L)	16.29	69.08	15.1	105.99	15.1	15.1
	Chlordane (ng/L)	0.03	0.11	0.02	0.16	0.02	0.02
	Dieldrin (pg/L)	1.82	7.95	1.74	12.19	1.74	1.74

Abbreviations: FDEP, Florida Department of Environmental Protection; ODEQ, Oregon Department of Environmental Quality.

of 1 applied to the 90th percentile and, therefore, were chosen as the final noncancer endpoint criteria. The final PRA-derived criteria were similar to, although generally slightly lower than, USEPA's deterministic criteria protective of noncarcinogenic effects.

Comparison of AWQC for 94 substances

Differences in PRA-derived AWQC following ODEQ and FDEP guidance and deterministically derived AWQC for all 94 substances updated by USEPA in 2015 (USEPA, 2015b) depended on toxicological endpoint and criteria type (i.e., Water and Organism [Figure 3] vs. Organism Only [Figure 4]). Negative *x*-axis values indicate that the respective PRA method results in criteria with a lower concentration than the deterministic method. Positive *x*-axis values indicate that the respective PRA method results in criteria with a higher concentration than the deterministic method. The *y*-axis shows the probability density for the kernel density estimation and can be interpreted as a relative frequency of occurrence within each plot. For Water and Organism criteria, PRA criteria derived using ODEQ acceptable risk guidance and the deterministic criteria were generally similar, with criteria ratios ranging from 0.9 to 1.1, although, for the majority of compounds, PRA-derived criteria tend to be slightly lower (Figure 3i: A–C). For all compounds, Water and Organism PRA criteria derived using FDEP acceptable risk guidance were higher than deterministic criteria for the cancer endpoint (criteria ratios = 2.5–4.5), and similar to the deterministic criteria for the noncancer endpoint (criteria ratios = 0.9–1.1). There were no differences in noncarcinogenic endpoints between PRA-derived criteria following ODEQ and FDEP acceptable risk guidance because both methods used an HQ = 1 at the 90th percentile (Figure 3i,ii: B). Because final AWQC are the lower of the carcinogenic and noncarcinogenic endpoint criteria, differences in AWQC reflect combined distributions that include substance AWQC driven by either noncarcinogenic or carcinogenic effects (Figure 3i,ii: C). Final AWQC differences between PRA-derived criteria were close to ±10% following ODEQ guidance but ranged from 10% lower to 4.5-fold higher than deterministic AWQC following FDEP guidance.

Organism Only criteria derived using ODEQ acceptable risk guidance were lower than deterministic criteria for carcinogenic and noncarcinogenic endpoints and final AWQC, although criteria ratios did not fall below 0.9 (Figure 4i: A–C). In contrast, PRA-derived Organism Only criteria following FDEP acceptable risk guidance were approximately 4- to 4.5-fold higher than deterministic criteria for carcinogenic effects (Figure 4ii: A) and less than 10% lower for noncarcinogenic effects (Figure 4ii: B). The range of criteria ratios between the PRA and deterministically derived criteria protective of carcinogenic effects following FDEP guidance was substantially smaller for Organism Only than for Water and Organism criteria (Figures 3ii vs. 4ii: A) due to the inclusion of drinking water consumption exposure in the latter criteria. The final Organism Only AWQC were similar between the PRA-derived criteria following ODEQ acceptable risk guidance and those derived using the deterministic method, with all PRA criteria within approximately 10% of deterministic criteria (Figure 4i: C). Virtually all Final Organism Only PRA-derived AWQC following FDEP guidance fell into two ratio ranges (Figure 4ii: C): the first within approximately 10% of USEPA's deterministic AWQC and the second approximately 4.5-fold higher than USEPA's deterministic criteria. The two sets of ranges corresponded to whether the endpoint determining the Final Organism Only AWQC were noncarcinogenic and carcinogenic effects, respectively.

DISCUSSION AND CONCLUSION

Water resource managers and state agencies are charged with establishing water quality standards that reflect the latest scientific knowledge and are protective of human health. As the first study to quantitatively compare probabilistic versus deterministic methods for deriving human health AWQC using USEPA's latest national recommendations, our results demonstrate that, although PRA methods are often assumed to result in less stringent criteria, the criteria-derivation approach used does not influence resulting criteria values.

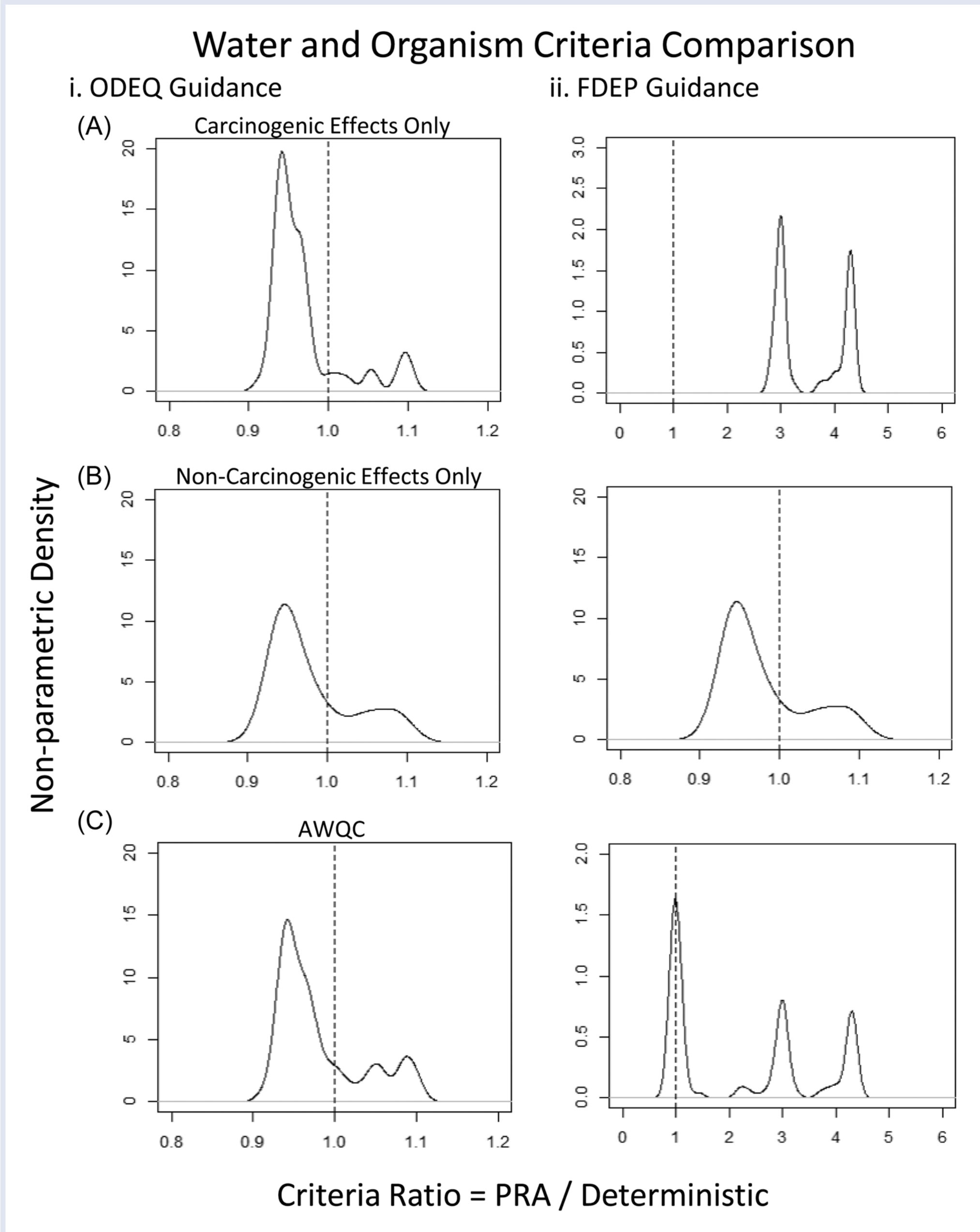

FIGURE 3 Nonparametric kernel density plots showing criteria ratios Water and Organism criteria between probabilistic risk assessment (PRA) criteria calculations following (i) Oregon Department of Environmental Quality (ODEQ) and (ii) Florida Department of Environmental Protection (FDEP) guidance and deterministic criteria for (A) carcinogenic effects, (B) noncarcinogenic effects, and (C) ambient water quality criteria (AWQC), which is the lesser of the carcinogenic or noncarcinogenic effects criteria for each substance. The scale of the *x*-axis varies by fivefold between some plots

Rather, the stringency of criteria depends on the underlying inputs and assumptions for exposure, toxicity, and, especially, target risk and the percentile of the population to which it applies. Nonetheless, the PRA approach outlined in our study offers several advantages over the deterministic method for deriving AWQC. Like the deterministic method, PRA can use national-, state- or region-specific inputs. Yet, although PRA accommodates entire distributions of exposure inputs to derive AWQC, the deterministic method is limited to point estimates from these distributions and ignores the remaining data. The use of distributions more realistically characterizes the variety of exposures experienced throughout a population and helps to reduce compounded conservatism associated with selecting multiple upper-percentile point estimates for individual exposure inputs (Tatum et al., 2015). The use of PRA in AWQC derivation also has the advantage of generating criteria that are explicitly protective of multiple segments of the population characterized by unique levels of exposure. The USEPA's AWQC guidelines have stated that, whereas excess lifetime cancer risks of 1×10^{-6} or 1×10^{-5} are suitable as health protection targets for the general population, the most highly exposed subpopulations must be protected at a 1×10^{-4} allowable risk level (USEPA,

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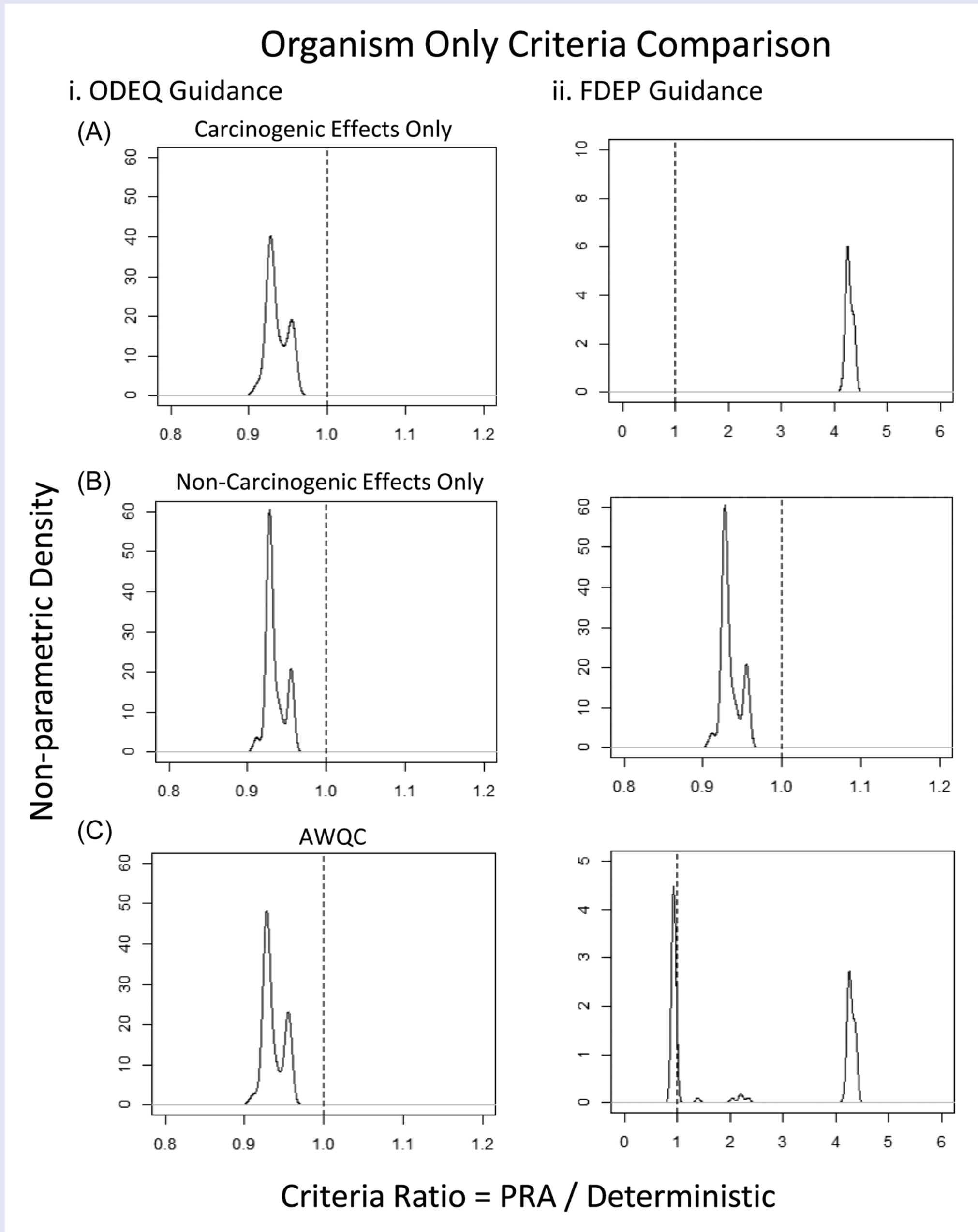

FIGURE 4 Nonparametric kernel density plots showing criteria ratios Organism Only criteria between probabilistic risk assessment (PRA) criteria calculations following (i) Oregon Department of Environmental Quality (ODEQ) and (ii) Florida Department of Environmental Protection (FDEP) guidance and deterministic criteria for (A) carcinogenic effects, (B) noncarcinogenic effects, and (C) ambient water quality criteria (AWQC), which is the lesser of the carcinogenic or noncarcinogenic effects criteria for each substance. The scale of the *x*-axis varies by fivefold between some of the plots

1999, 2020). Probabilistic risk assessment can derive criteria that demonstrate these conditions are met; the deterministic method cannot. Instead, the deterministic method defines an exposed population through a combination of point estimates for individual characteristics (i.e., body weight, fish consumption, drinking water intake) and does not (and cannot) explicitly determine the percentage of the population to which all these assumptions apply. As such, PRA represents a more transparent methodology that clearly connects intended health protection targets to specific segments of the exposed population.

Despite these advantages, several challenges remain to the successful implementation and widespread adoption of PRA among states. In particular, the perceived complexity of the PRA method may be an impediment to its use. The development of user-friendly tools, such as the Excel software and the proprietary plug-in @Risk used in this study, may ease this burden. However, overcoming costs, both in terms of fee-based software and time dedicated from staff at state and federal agencies to implement the PRA method and develop state-specific probabilistically derived ambient AWQC, remains a challenge. In the past, perceived lack of data to support robust input distributions may have also led to reticence among regulatory agencies to adopt a PRA approach. Now, such data are readily available from national (USEPA, 2011) and regional (FDEP, 2016; IDEQ, 2014; ODEQ, 1998; WDOE, 2013; WVDEP, 2008) data sources. Perhaps the most important challenge to the successful implementation of PRA for deriving AWQC is the lack of guidance from USEPA regarding appropriate probabilistic inputs and demonstrated willingness to approve PRA-based AWQC. For example, our implementation of PRA to derive AWQC followed acceptable risk guidance from Florida (FDEP, 2016) and Oregon (ODEQ, 1998) linked health protection targets to percentiles of the population for which they apply. Although PRA guidance relating to Superfund applications refer to the 50th percentile as central tendency exposure and 90th to 99.9th percentiles as reasonable maximum exposures (RME; USEPA, 2001), there is no USEPA PRA guidance related to AWQC that specifically defines the "general population" to be protected at 10^{-6} and 10^{-5} and "most highly exposed subpopulations" to be protected at 10^{-4}. Although this might give leeway to states implementing PRA approaches, it remains uncertain whether USEPA will approve probabilistically derived AWQC submitted by states. As a result, states may be reluctant to dedicate limited resources to a criteria-derivation approach that, although more appropriate to meet their objectives, might subsequently need to be redone if rejected by USEPA.

Although we used distributions to more fully characterize some exposure parameters including body weight, drinking water intake, and fish consumption in our probabilistic approach, distributions could also be used for the other exposure parameters shown in the equations used by USEPA to derive AWQC (i.e., exposure frequency, exposure duration, RSC, and BAF). Additionally, the equations could be refined and expanded to include implicit exposure parameters such as cooking loss and the assumption that the concentration of regulated substances is equal to the maximum allowed by AWQC over the entire exposure period (lifetime). Ideally, though not currently allowed by USEPA, reference doses and cancer slope factors should also be represented by distributions to capture uncertainty and improve overall characterization of the level of protection associated with AWQC. However, even absent such refinements, these study results should prompt federal regulators to consider producing guidance on the use of PRA for deriving AWQC to help both federal and state regulators understand available approaches to deriving AWQC, and perhaps expand the use of PRA to establish scientifically defensible, transparent, data-inclusive, and more explicitly protective AWQC.

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CONFLICT OF INTEREST

The authors declare no conflicts of interest.

AUTHOR CONTRIBUTIONS

Brad Barnhart: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Validation; Visualization; Writing—original draft; Writing—review & editing. **Camille Flinders**: Formal analysis; Investigation; Methodology; Validation; Visualization; Writing—original draft; Writing—review & editing. **Giffe Johnson**: Formal analysis; Investigation; Methodology; Validation; Writing—original draft; Writing—review & editing. **Paul Wiegand**: Conceptualization; Formal analysis; Investigation; Methodology; Validation; Writing—original draft; Writing—review & editing. **Paul Anderson**: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Software; Validation; Writing—review & editing. **Emily Morrison**: Formal analysis; Investigation; Methodology; Software; Validation; Writing—review & editing. **Gina Houck**: Formal analysis; Investigation; Methodology; Software; Validation; Writing—review & editing.

DATA AVAILABILITY STATEMENT

All data, associated metadata, and calculation tools generated by this article are available, either as Supporting Information or by requesting access from corresponding author Brad Barnhart (bbarnhart@ncasi.org).

SUPPORTING INFORMATION

Table A1. Listing of inputs used to derive ambient water quality criteria (AWQC) for 94 substances listed in USEPA's 2015 update. Note that benzene has two listings ("Low" and "High") because USEPA 2015 guidance specifies a range of criteria values.

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