



State of Oregon  
Department of  
Environmental  
Quality

# **Water Quality Standards Review and Recommendations: Arsenic**

**Attachment E  
April 21, 2011, EQC Meeting**

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# Executive Summary

The Department of Environmental Quality (DEQ) is proposing to revise Oregon's human health water quality criteria for arsenic as shown in Table 1 below. This issue paper contains discussion of the proposed criteria, the scientific basis and rationale for the proposed revisions and the process DEQ used to review these criteria.

Water body type	Water + Fish Ingestion		Fish Consumption Only	
	Current Criteria	Proposed Criteria	Current Criteria	Proposed Criteria
Freshwater	0.0022	2.1	0.0175	2.1
Saltwater	NA	NA	0.0175	1.0

**Notes:**

- 1) Current criteria are from Table 20 (OAR 340-041-0033).
- 2) All proposed criteria are based on a fish consumption rate of 175 g/d.

**Recommendations:**

1. Establish separate human health "fish consumption" criteria for freshwater and saltwater because marine shellfish (oysters) have much higher bioconcentration rates for arsenic than freshwater finfish.
2. Revise the freshwater criterion for "water + fish ingestion" to 2.1  $\mu\text{g/l}$  as inorganic arsenic.
  - This criterion is calculated using a bioconcentration factor (BCF) of 14 L/kg, which is the geometric mean of the available freshwater fish bioconcentration data.
  - The bioconcentration factor is based on three studies; two for trout and one for bluegill. DEQ used all three studies in selecting the bioconcentration factor because the fish consumption rate also includes a combination of species.
  - These bioconcentration studies were conducted at relatively low water concentrations of arsenic (below 50 $\mu\text{g/l}$ ). DEQ did not use studies conducted at higher concentrations (i.e. greater than 50 up to 1000 $\mu\text{g/l}$ ), since the results are not reflective of arsenic concentrations in Oregon surface waters, which range from less than 0.5 to 16  $\mu\text{g/l}$ .
  - This criterion represents a 1 in 10,000 (i.e.  $1 \times 10^{-4}$ ) risk level and is within the acceptable risk ranges established by the federal Environmental Protection Agency.

3. Revise the freshwater criterion for “fish consumption only” to 2.1 µg/l as inorganic arsenic.
  - DEQ recommends using the same value for “fish consumption only” and “water + fish consumption” as a statewide criterion.
  - DEQ evaluated a “fish consumption only” criterion using a bioconcentration factor of 14 L/kg and a risk level of  $10^{-5}$ , which resulted in a value of 1.9 µg/l. Using this value would result in a criterion to protect eating fish that is more conservative than the criterion to protect both eating fish and drinking water.
  - A “fish consumption only” criterion of 2.1 µg/l is consistent with the “water + fish ingestion criterion” and is protective at a risk rate only slightly greater than  $10^{-5}$ , which is within the acceptable risk range established by EPA.
  - DEQ also recommends establishing site specific criteria based on natural conditions in the future for waterbodies where information demonstrates that the arsenic concentration in the water body due to natural sources is greater than 2.1 µg/l.
  
4. Adopt a “fish consumption only” saltwater criterion of 1µg/l as inorganic arsenic.
  - This criterion represents a 1 to 1.3 in 100,000 (i.e. 1 to  $1.3 \times 10^{-5}$ ) risk level, which is within the acceptable risk ranges established by EPA.
  - The separate arsenic saltwater criterion incorporates the marine shellfish bioconcentration factor data.
  - Use multiple lines of evidence to establish a saltwater criterion. One approach uses the geometric mean of the available freshwater and saltwater bioconcentration factor data to calculate a saltwater criterion, recognizing that the amount of data representing the variety of saltwater species are very limited. Because this approach is based on assumptions regarding the applicability of the freshwater bioconcentration factor data to saltwater finfish species, DEQ also conducted an analysis based on the higher oyster bioconcentration factor combined with a lower inorganic arsenic proportion factor. Finally, DEQ also evaluated these results against natural ocean arsenic concentrations cited in the scientific literature, which are approximately 1.7 to 2 µg/l total arsenic and 1µg/l inorganic arsenic.
  - The ‘water + fish ingestion’ criterion does not apply to saltwater because saltwater is not used for drinking water (domestic water) supply.

## Background

DEQ derived the proposed arsenic criteria using EPA’s calculation method. However, DEQ adapted the calculation for Oregon by using locally appropriate values rather than national default values for specific variables. All the proposed criteria are based on a fish consumption rate of 175 grams per day. The risk level used for each proposed criterion varies as follows: the water + fish ingestion criterion is based on a cancer risk level of  $10^{-4}$ , the freshwater fish consumption only criterion, which is the same value, represents a risk level slightly higher than  $10^{-5}$ , and the saltwater fish consumption only criterion is based on a risk level of  $10^{-5}$ . In addition, the proposed arsenic criteria use bioconcentration factors of 14 and 26 for the freshwater and saltwater criteria, respectively, and a 10 percent inorganic arsenic factor. An alternate calculation for saltwater is also conducted using a bioconcentration factor of 350 and an inorganic proportion of 1 percent. Further explanation of these variables and the criteria calculations is provided in section 5 of this paper.

DEQ proposes adopting locally derived criteria rather than EPA’s nationally recommended criteria values because the natural background levels of arsenic in many Oregon waters are much higher than the national criteria. Naturally-occurring arsenic in Oregon comes from geologic sources and levels are often higher in ground water than in surface waters. DEQ’s proposed criteria for inorganic arsenic are consistent with EPA recommendations. While inorganic arsenic is the form of arsenic that is toxic to humans, it does not bio-accumulate in fish tissue as readily

as total arsenic. While DEQ's proposed water + fish ingestion value is significantly higher than EPA's recommended criteria under the Clean Water Act (CWA), it is lower than the maximum contaminant level established by EPA as protective of finished drinking water under the Safe Drinking Water Act.

DEQ concludes that the proposed criteria represent an appropriate balance of human health protection and recognition that many Oregon waters contain arsenic from natural geologic sources, commonly at levels of 1-3 µg/l, and in some water bodies significantly higher. These natural levels do not represent new or added health risk to the environment. Setting criteria that would trigger widespread identification of Oregon waters as impaired for arsenic, the subsequent development of total maximum daily loads and other Clean Water Act implementation activities would incur large costs for actions that would rarely result in reducing arsenic levels in the water or in fish. Similarly, establishing an arsenic criteria for saltwater below the natural levels of unpolluted coastal and ocean waters would suggest that it is not safe to eat fish from those waters, which is not a conclusion supported by the scientific literature.

DEQ proposes to include an arsenic reduction policy in the state's water quality regulations. This rule establishes a policy to reduce human sources of arsenic that are likely to impact a public drinking water supply and includes requirements for industrial permittees to evaluate data and develop arsenic reduction plans. This provision would apply in instances where the ambient arsenic level is below the numeric criteria in order to minimize the amount of arsenic added to surface waters from human sources.

# Chapter 1. Introduction and Background

The Oregon Department of Environmental Quality (DEQ) reviewed the science behind the human health water quality criteria for some of the naturally occurring earth metals in response to concerns expressed to the Oregon Environmental Quality Commission (EQC) at their October 2008 meeting. Arsenic, iron and manganese are the three metals that DEQ selected to review in more detail. These three earth metals are naturally occurring and are found in Oregon waters at natural background levels greater than the current human health criteria. There are 107 water body segments listed as impaired for these three metals on the 2004/06 303(d) list. In addition, stakeholders point out that the arsenic criteria under the Clean Water Act are much more stringent than the maximum contaminant level for drinking water established under the Safe Drinking Water Act.

At the October 2008 meeting, the EQC directed DEQ to revise Oregon's human health criteria for toxic pollutants based on the recommended increased fish consumption rate of 175 grams per day. DEQ is in the process of conducting that rulemaking. DEQ adopted revisions to the iron and manganese criteria in December 2010 and is now proposing revisions to the arsenic criteria in advance of the full human health criteria rulemaking for several reasons. First, the timeframe for the larger package targets EQC adoption in mid-2011 and the revised criteria associated with that rulemaking will not likely be effective until late 2011 at the earliest, possibly not until mid-2012 or later. Second, the scientific review and early stakeholder review of these revisions are complete and the proposal is ready for adoption. Third, the changes are significant for several NPDES permits that will be renewed over the next year to 18 months. And lastly, 22 stream segments are listed for arsenic. If the proposed revisions are adopted by the EQC in late 2010 or early 2011, they should be effective for use in the 2012 water quality assessment. This will help DEQ to target its resources and those of dischargers to address priority environmental improvements.

# Chapter 2. Arsenic Human Health Criteria Review and Recommendations

## Section 2.1. Concerns about Oregon's Human Health Criteria for Arsenic

DEQ reviewed the science behind the human health water quality criteria for arsenic in response to several concerns, which were expressed to the Oregon Environmental Quality Commission at the October 2008 meeting. First, arsenic is a naturally occurring earth metal found in Oregon waters at natural background levels much greater than the current human health criteria. Second, the human health water quality criteria for arsenic that currently apply in surface waters under the Clean Water Act are much lower than the maximum contaminant level (MCL) developed under the Safe Drinking Water Act for finished drinking water delivered to people's homes.

DEQ's current arsenic criteria are shown in Table 3 and described below. Having arsenic criteria that are well below widespread natural background levels of the pollutant presents several problems for the state and for cities and industries that discharge to waters of the state. First, this situation has resulted in 303(d) listings of water bodies as impaired (currently 22 segments) and DEQ expects many more will be identified as more data are collected, even though the arsenic levels are predominantly due to natural geologic sources. DEQ must then address the listings by developing a total maximum daily load (TMDL) or providing an explanation or plan for situations where the source of arsenic is natural and cannot be controlled. This is not a meaningful use of public resources.

Another result of a water body being listed as "impaired" or having a background pollutant concentration above the water quality criterion is that there is no assimilative capacity or mixing available to cities and industries that discharge to the water body. Therefore, the facility must meet the water quality criterion at the "end-of-pipe," prior to discharging into the river. DEQ expects that under the current arsenic criteria or new criteria based on changing only the fish consumption rate, many municipal wastewater treatment plants and a number of industrial facilities would not be able to meet their resultant permit limits. In some cases, a facility would not be able to discharge the same amount of arsenic they brought into the facility from the river via their intake water. Even if the facility adds no arsenic to its wastewater, if it concentrates the arsenic, which occurs, for example, when the water is used for non-contact cooling, the facility would not be able to achieve the effluent quality necessary to meet the receiving water's arsenic criteria.

While DEQ's standards contain a "natural condition" provision, EPA has stated that this type of provision should not apply to human health criteria. The criteria need to protect the uses, which are fishing (i.e. fish consumption) and domestic water supply. For aquatic life, natural conditions are reasoned to support native aquatic species which have acclimated or adapted to the natural conditions. This same reasoning does not necessarily hold true for humans at the risk levels and life span targeted for human health protection. Therefore, if DEQ proposes to set human health criteria based on natural background levels, DEQ must demonstrate that those levels are protective of human health.

Another concern that has been expressed to DEQ is the fact that the current arsenic criteria and any revised criteria that would be based only on an increased fish consumption rate are significantly

lower than the maximum contaminant level (MCL). The MCL is the regulatory limit set under the Safe Drinking Water Act to protect public drinking water supplies and applies to finished drinking water delivered to people's homes.

For these reasons, DEQ pursued development of revised arsenic criteria with the objective of protecting human health along with the ability to use waters with natural levels of arsenic for domestic water supply where those arsenic levels do not present an unacceptable human health risk. DEQ also recognized the considerable costs associated with meeting requirements based on the current criteria.

## Section 2.2. Arsenic in Oregon

**Background Levels.** Based on the available data, most Oregon waters have natural background levels of arsenic in the range of less than 1 microgram per liter ( $\mu\text{g/l}$ ) up to 3  $\mu\text{g/l}$ . There are limited data available on arsenic concentrations in surface waters, partly because until recently DEQ used 5.0  $\mu\text{g/l}$  as the laboratory method quantitation limit. Therefore, much of the data collected by DEQ or permittees report "non-detectable" levels of arsenic. In 2008, DEQ reduced the quantitation limit for arsenic to 0.5  $\mu\text{g/l}$ .

DEQ data from approximately 1979-1981 indicate that much higher arsenic levels (greater than 5-10  $\mu\text{g/l}$ ) may be present in some south central and southeastern Oregon basins. More recent data also show a range of arsenic levels of less than 1  $\mu\text{g/l}$  to greater than 10  $\mu\text{g/l}$  in upper Klamath basin streams. It is not known whether these levels represent solely natural geologic sources or are elevated due to anthropogenic activity. Some of the samples, which were taken from spring fed creeks and locations upstream of human activity, clearly measured natural levels.

**Natural Sources.** There are natural geologic sources of arsenic in Oregon. The City of Portland has found arsenic levels in the Bull Run reservoir, a primary source of Portland's drinking water that is upstream of human activity in a protected watershed, ranging from less than 1  $\mu\text{g/l}$  (their minimum reporting level) up to 3  $\mu\text{g/l}$ . Data from other Oregon streams show arsenic levels in this range as well, including the Crooked River upstream of Prineville, the Little Deschutes River and some streams in the upper Klamath basin. A spring in the upper Klamath basin had an arsenic concentration of 16  $\mu\text{g/l}$  (Newton Consultants Inc., for City of Klamath Falls, 2008). Samples from the upper Santiam basin were mostly below the 0.5  $\mu\text{g/l}$  detection level.

A 1998 U.S. Geological Survey report on arsenic concentrations in ground water of the Willamette Basin found concentrations ranging from less than 1 to 2,000  $\mu\text{g/l}$ . The report concludes:

1. Regional patterns of arsenic occurrence in the Willamette Basin indicate that the sources of arsenic in ground water are not human related. Arsenic-containing metal oxides, volcanic glass in volcanic rocks of rhyolitic to intermediate composition, and clays are likely sources.
2. High arsenic concentrations (concentrations exceeding the current MCL established by EPA) appear to be associated with particular associations of rock in some areas and with alluvial deposits in others (i.e. the Tualatin basin). (paraphrased)
3. For alluvial ground water of the Tualatin Basin, (1) presence of competing anions and (2) occurrence of reducing conditions may be important controlling factors in arsenic adsorption/desorption reactions. Dissolution of iron oxides, with subsequent release of adsorbed and (or) co-precipitated arsenic, also may play an important role in arsenic mobility in ground water of the Tualatin Basin.

A 1998 arsenic study by the Washington Department of Ecology (Ecology), that included data collection from the Columbia River, reported:

the recent data suggest that total recoverable arsenic concentrations in local rivers and streams are typically in the range of 0.2 - 1.0 µg/L, while concentrations greater than 2 to 5 µg/L may indicate contamination from anthropogenic sources. Arsenic levels in most 303(d) listed waterbodies are not clearly different from waterbodies that have no apparent sources, and some are comparable to rainwater. (Results and Recommendations from Monitoring Arsenic Levels in 303(d) Listed Rivers in Washington, WDOE, 2002)

**Human Sources.** A document titled *Toxicological Profile for Arsenic* (ATSDR, 2007) describes the various means by which people have affected the fate and transport of arsenic in the environment, including the following:

- When ores that contain copper or lead are heated in smelters, “most of the arsenic goes up the stack and enters the air as a fine dust. Smelters may collect this dust and take out the arsenic as a compound called arsenic trioxide (As<sub>2</sub>O<sub>3</sub>).”
- Presently, about 90% of all arsenic produced is used as a preservative for wood to make it resistant to rotting and decay. The preservative is copper chromated arsenate (CCA) and the treated wood is referred to as “pressure-treated.” In 2003, U.S. manufacturers of wood preservatives containing arsenic began a voluntary transition from CCA to other wood preservatives that do not contain arsenic in wood products for certain residential uses, such as play structures, picnic tables, decks, fencing, and boardwalks. This phase out was completed on December 31, 2003; however, wood treated prior to this date could still be used and existing structures made with CCA-treated wood would not be affected. CCA-treated wood products continue to be used in industrial applications. It is not known whether, or to what extent, CCA-treated wood products may contribute to exposure of people to arsenic.
- In the past, inorganic arsenic compounds were predominantly used as pesticides, primarily on cotton fields and in orchards. Inorganic arsenic compounds can no longer be used in agriculture. However, organic arsenic compounds, namely cacodylic acid, disodium methylarsenate (DSMA), and monosodium methylarsenate (MSMA), are still used as pesticides, principally on cotton. Some organic arsenic compounds are used as additives in animal feed.
- Small quantities of elemental arsenic are added to other metals to form metal mixtures or alloys with improved properties. The greatest use of arsenic in alloys is in lead-acid batteries for automobiles.
- Another important use of arsenic compounds is in semiconductors and light-emitting diodes. (ATSDR, 2007)

**Arsenic Impaired Waters.** The streams shown in the table below are currently 303(d) listed for exceeding arsenic criteria. There are 20 water body segments listed for arsenic, out of a total of 249 stream segments on the 2004/06 303d list for a toxic pollutant.

Basin	River	River Miles	Year listed
Multi	Columbia	0-142	1998
Willamette	Willamette	175 – 186	2002
Upper Willamette	A-3 drain	---	2002
Upper Willamette	Amazon Cr.	0-23	2002
Upper Willamette	Willow Cr.	0-3	2002
North Umpqua	N. Umpqua	35-52	2002

North Umpqua	Sutherlin Cr.	0-16	2002
North Umpqua	Unnamed Cr.	---	2002
South Umpqua	Middle Cr.	0-13	2004
South Umpqua	S. Umpqua R.	0-16	2002
Warner Lakes	Twelvemile Cr.	0-17	2002
Warner Lakes	Twentymile Cr.	0-29	2002
Owyhee	Owyhee River	71-200	2004
Jordan	Jordan Cr	0-95	2004
Mid Col-Hood	Lenz Cr	0-1.5	2004
Mid Col-Hood	Neal Cr.	0-6	2004
Molalla-Pudding	Zollner Cr	0-8	2004

### Section 2.3. Potential Health Impacts of Arsenic

Arsenic is a known carcinogen that may cause cancer in skin or internal organs such as the liver, kidneys, lungs and bladder. Other potential health impacts from arsenic include cardiovascular, kidney, central nervous system and hyper pigmentation or keratosis effects (USEPA, 2000). Factors for how to represent these effects in the criteria equations are included in EPA's Integrated Risk Information system (IRIS) database. The EPA recommended arsenic criteria are based on a cancer endpoint and are based on inorganic arsenic.

### Section 2.4. Current Human Health Criteria for Arsenic: State and Federal

The current Oregon and EPA arsenic criteria are shown in the table below.

Table 3. Current Arsenic Criteria		
	Water and fish ingestion ( $\mu\text{g/L}$ )	Fish consumption only ( $\mu\text{g/L}$ )
Currently effective Oregon criteria (Table 20)	0.0022	0.0175
Criteria adopted by Oregon in 2004	0.018*	0.14*
Current EPA criteria	0.018*	0.14*

\* Inorganic arsenic

Oregon's currently effective criteria (OAR 340-041-0033, Table 20) are based on EPA's 1986 recommended criteria and are based on a fish consumption rate of 6.5 g/d. Table 20 does not specify whether the human health criteria are for inorganic arsenic or total arsenic. The toxicity data EPA used to calculate the 1986 recommended criteria were for inorganic arsenic.

EPA's current arsenic criteria for human health and the criteria adopted by the EQC in 2004 are based on a fish consumption rate of 6.5 g/d and a cancer slope factor of 1.75, and are specifically identified as criteria for inorganic arsenic. In 1992, EPA promulgated these arsenic criteria in the National Toxics Rule (USEPA, 1992). Although EPA has since changed the cancer slope factor in its IRIS database to 1.5 (4/10/1998) and changed its recommended fish consumption rate to 17.5 (EPA, 2000), it has not revised the nationally recommended arsenic criteria accordingly. EPA is currently reviewing the cancer slope factor and has released an increased IRIS value for comment.

EPA did not promulgate human health criteria for arsenic in the California Toxics Rule (CTR) in 2000, stating that “a number of issues and uncertainties existed at the time of the CTR proposal concerning the health effects of arsenic.” Neither did EPA include arsenic criteria in its promulgation of criteria for the Great Lakes states in 1995.

Other states have human health arsenic criteria ranging from a low of the current federal criteria to a high of 50 µg/l. Almost half of the states have criteria of 10 or 50 µg/l based on the current or previous Safe Drinking Water Act maximum contaminant level (MCL). About 10 states do not have a “water & organism” arsenic criterion and several do not have a “fish consumption only” criterion. A few states have recalculated their arsenic criteria using EPA equations but altering some of the variables in those equations. The variables states have revised include the bioconcentration factor (BCF), the EPA cancer slope factor (using the current IRIS value of 1.5), the fish consumption rate, and/or the risk level (using 10<sup>-5</sup> rather than 10<sup>-6</sup>). In addition, some states have applied an inorganic proportion to the calculation since the criteria apply to inorganic arsenic.

**How the Federal Arsenic Criteria Were Calculated.** The following two equations and accompanying table describe the variables that were used to calculate EPA’s current national human health criteria for arsenic.

$$\text{Water + fish ingestion Criterion } (\mu\text{g/L}) = 1000 \times \frac{\text{RF} \times \text{BW}}{\text{q1}^*[\text{DW} + (\text{BCF} \times \text{FCR})]}$$

$$\text{Org Only Criterion } (\mu\text{g/L}) = 1000 \times \frac{\text{RF} \times \text{BW}}{\text{q1}^*[\text{BCF} \times \text{FCR}]}$$

Symbol	Description	Value Used for Federal Criteria	Value Used for Oregon Freshwater Criteria	Value Used for Oregon Saltwater Criteria
RF =	risk level factor (dimensionless)	1x10 <sup>-6</sup>	fish only 1.1x10 <sup>-5</sup> water + fish 1x10 <sup>-4</sup>	fish only 1x10 <sup>-5</sup>
BW =	body weight (kg)	70	70	70
q1* =	cancer potency factor (mg/kg/day)-1	1.75	1.5 <sup>a</sup>	1.5 <sup>a</sup>
DW =	Drinking water consumption (L/day)	2	2	2
BCF =	bioconcentration factor (L/kg)	44	14	26
FCR =	fish consumption rate (kg/day)	0.0065	0.175	0.175
IF =	Inorganic proportion factor	No factor	10%	10%

<sup>a</sup>The current cancer potency factor published by EPA in their IRIS data base.

### Section 2.5. DEQ Proposed Revised Arsenic Criteria

DEQ proposes to adopt separate criteria for Oregon freshwater and saltwater due to the apparent differences in bioconcentration and arsenic species and transformations in the marine environment. DEQ proposes to revise the arsenic criteria for freshwater using EPA’s calculation method, substituting values in the criteria equation that have been updated or are more appropriate for Oregon. The proposed criteria are shown in Table 5. DEQ concludes that the proposed criteria protect human health while recognizing that Oregon has widespread natural

background levels of arsenic higher than EPA's recommended criteria. DEQ's Toxics Standards Review Rulemaking Workgroup, a group of stakeholders that provided input to DEQ on this rulemaking, supported revising the arsenic criteria based on Oregon appropriate variables and a higher risk level because of the natural background levels of arsenic found in Oregon waters.

**Table 5. Proposed Arsenic Criteria (as inorganic arsenic)**

	<b>Water + fish ingestion</b>	<b>Fish consumption only: freshwater</b>	<b>Fish consumption only: saltwater</b>
Proposed Criterion	2.1 µg/l	2.1 µg/l	1.0 µg/l
Values used to calculate criteria	FCR=175 BCF=14 IF=10% CSF=1.5 Risk level= $1 \times 10^{-4}$	FCR=175 BCF=14 IF=10% CSF=1.5 Risk level= $1.1 \times 10^{-5}$	FCR=175 BCF=26 IF=10% CSF=1.5 Risk level= $1 \times 10^{-5}$

The Oregon specific variables, shown in Tables 4 and 5 above and discussed in more detail below, include the fish consumption rate (FCR), the bioconcentration factor (BCF), a percent inorganic arsenic factor (IF) and the risk level. In addition, DEQ uses the current IRIS cancer slope factor of 1.5.

### Section 2.5.1 Inorganic Arsenic Criteria for Freshwaters

The proposed inorganic arsenic criteria, shown in Table 5, are based on a fish consumption rate of 175 grams/day, a bioconcentration factor of 14, an inorganic proportion of 10% and a risk level of about one in 100,000 ( $1 \times 10^{-5}$ ). DEQ's rational and supporting information for these variables is discussed here.

**Fish Consumption Rate.** DEQ calculated the proposed criteria using 175 g/d as the fish consumption rate (DEQ, 2008a). The current federal arsenic criteria are based on a consumption rate of 6.5 g/d. Using this higher rate is responsive to EPA's disapproval of Oregon's 2004 human health criteria which was based on their conclusion that criteria based on 17.5 g/d is not sufficient to protect fish consumers in Oregon.

In advance of EPA's action and based on earlier concerns expressed by EPA on this issue, DEQ looked at multiple studies of fish consumption rates with the assistance of experts in toxicology and public health (the Human Health Focus Group), focusing on five studies conducted in Oregon and Washington as well as the national survey used by EPA. The rate of 175 g/d represents the 90<sup>th</sup> to 95<sup>th</sup> percentile of Oregon fish consumers as indicated by these studies (DEQ, 2008b). This value represents the total amount of fish consumed, regardless of species or origin, because it was found that different populations, depending on access and culture, will eat different species of fish. As a result, DEQ, with the support of the Confederated Tribes of the Umatilla Indian Reservation and EPA Region 10, selected 175 g/day as an appropriate value to use for the calculation of human health criteria.

**Risk Level.** When EPA develops recommended human health criteria for carcinogens, it uses a cancer risk level of  $10^{-6}$ , one in one million additional incidents of cancer, which it characterizes as an appropriate level of risk for the general population. However, EPA guidance allows that risk levels of  $10^{-6}$  or  $10^{-5}$  are acceptable for the general population and that highly exposed populations should not exceed  $10^{-4}$ . Within this range, the risk level is a policy decision for States

to make when they establish water quality criteria. To date, DEQ has used the  $10^{-6}$  risk factor for water quality human health criteria and in other environmental protection programs that are based on human health risk, such as the clean-up of contaminated sites. DEQ is not re-evaluating the risk level used for Oregon's human health criteria generally. However, because of the particular fact set associated with arsenic as described throughout this issue paper, DEQ is recommending criteria based on alternate risk levels as shown in Tables 4 and 5 above. The primary reason is because naturally occurring arsenic concentrations in many Oregon waters exceed values based on lower risk levels. The risk associated with natural levels of arsenic has been present since people have been drinking water and eating fish from Oregon streams and lakes. Criteria that are exceeded due to natural conditions on a widespread basis around the state may lead to the expenditure of public and private resources to implement Clean Water Act programs that will not result in reduced water concentrations of arsenic. Communities that obtain their water supply from groundwater are likely to be exposed to higher arsenic levels. Groundwater is not regulated by the Clean Water Act and not subject to water quality criteria.

DEQ concludes that using the higher risk levels for the arsenic criteria is supported and consistent with EPA guidance (EPA, 2000) because Oregon used a fish consumption rate and, subsequently, derived criteria which protect highly exposed populations. The Oregon consumption rate of 175 g/d represents the 95<sup>th</sup> percentile of consumers within the state and protects people who eat fish on a regular basis. This is more inclusive than the rate used in EPA's criteria, which is based on the general per capita population and includes people that do not eat fish, or eat it only occasionally. Moreover, DEQ's consumption rate includes anadromous and marine fish.

As with the freshwater criteria, DEQ used a risk level of  $10^{-5}$  rather than  $10^{-6}$  to calculate the saltwater criterion for two reasons: 1) the criterion is also based on a fish consumption rate of 175 g/d, a high exposure rate, and 2) the presence of naturally occurring arsenic in marine waters (Tanaka, 1995; National Academy of Sciences, 1972; EPA, 2003). See additional discussion of the saltwater criterion and natural arsenic concentrations in seawater in Section 2.5.2 below.

DEQ's proposed criteria balance the objectives of minimizing human health risk and accounting for natural sources of arsenic. Some waterbodies will have natural background levels above the proposed statewide criteria. In these cases, DEQ may pursue site specific water quality standards at a later date.

**Bioconcentration.** Bioconcentration refers to the uptake and retention of a chemical by an aquatic organism from water. A bioconcentration factor (BCF) is the ratio of the concentration of a substance in the tissue of an aquatic organism to its concentration in the ambient water in situations where the organism is exposed through the water only and the ratio does not change substantially over time.

EPA's current BCF of 44 for arsenic is described in *Ambient Water Quality Criteria for Arsenic* (USEPA, 1980). EPA calculated the BCF using data from two species, the eastern oyster (BCF=350) and bluegill (BCF=4). Because it was based on only two species and one of those is the eastern oyster, which has a much greater BCF (350 versus 4), the BCF of 44 most likely overestimates the health risks associated with freshwater finfish consumption (USEPA Region 6, mid-1990s). In addition, the data sets used to establish the BCFs were relatively small (USEPA, 1980).

A more recent analysis by EPA (EPA Headquarters, personal communication, November 2010) incorporated more recent BCF data for rainbow trout with the prior data for bluegill and oysters to provide Oregon several scientifically defensible BCF options, shown in Table 6 below, for use in setting Oregon's criteria. The BCF options are based on geometric means of data from the following four studies, which include five BCF test values reported. EPA used the first two

studies listed to derive the BCF of 44 in the early 1980s; the second two studies are more recent. (see Appendix A for more detail on the results of these studies.

- Ambient Water Quality Criteria for Arsenic (EPA, 1985), which refers to Barrows et al., 1980, Ann Arbor Science Pub., Inc., Ann Arbor MI. pp. 379-392. Whole-body measurement of total arsenic in immature bluegill.
- Ambient Water Quality Criteria for Arsenic (EPA, 1985), which refers to Zaroogian and Hoffman, 1982, Environmental Monitoring and Assessment 1:345-358. BCF value for arsenic eastern oysters.
- McGeachy and Dixon, 1990. Canadian Journal of Fisheries and Aquatic Sciences. 47:2228-2223. Two studies of whole body total arsenic in immature rainbow trout.
- Rankin and Dixon, 1994. Canadian Journal of Fisheries and Aquatic Sciences. 51: 372-380. Whole-body measurement of total arsenic in immature rainbow trout.

Species	BCF	# of Studies	Range of values
All freshwater finfish	14	4	4-27
Coldwater fish (trout)	21	3	17-27
Saltwater (eastern oyster)	350	1	350
All freshwater and saltwater species	26	5	4-350

The above studies were selected because they tested species consumed by humans and were conducted with water concentrations below 50µg/l inorganic arsenic. Studies done at higher concentrations (i.e. 100 to 1000 µg/l) were not included. This segregation is appropriate because natural surface waters in Oregon are in the range of less than 0.5 to 16 µg/l. Studies conducted at higher background concentrations would be more appropriate for evaluating contaminated sites. BCFs from studies conducted at lower arsenic water concentrations tend to be higher and vice versa. Fish can bio-regulate arsenic as they do other metals, which are trace nutrients (DeForest et al, 2007). Organisms are able to take in less and eliminate excess when an abundant supply of the metal is available.

All of the values in Table 6 are the result of measuring total arsenic in the whole body rather than fillet or muscle tissue tests. EPA notes that BCFs for muscle tissue, the portion of the fish typically eaten, should be lower than those for the whole body (Stephan, 1993). Azcue and Dixon (1994; IN USEPA, mid 1990s) conducted a study that exemplifies this. The study measured arsenic in rock bass and found the highest concentrations in bone and scales, followed (in decreasing concentration) by intestines and contents, muscle and liver. A BCF of 0.71 was calculated for muscle tissue whereas the BCF based on whole body concentration was 2.3, three times greater than the muscle tissue BCF. Because the data being used by DEQ to derive a BCF value is based on whole-body testing, DEQ's value may be conservative. It is likely that most of the fish consumption captured by Oregon's rate of 175 g/d is muscle tissue rather than whole body. According to EPA's 2000 Human Health Methodology, data for arsenic in edible tissue of fish and shellfish are preferred over whole body data since that is the portion typically ingested.

One approach to deriving a criterion is to follow EPA's past practice when they derived the BCF of 44. Given the limited data, EPA combined the two data points and developed one recommended criterion to apply to all waters. DEQ pursued an alternative approach and developed separate criteria for fresh and salt waters. EPA stated in a 2003 review of arsenic bioaccumulation in aquatic organisms that estuarine and marine data indicate a possible need for deriving separate BAFs for saltwater systems (EPA 2003, p.7). Given that additional freshwater fish BCF data are now available, with values much lower than the BCF for the oyster, and given the presence of naturally occurring arsenic in Oregon waters, DEQ is recommending the adoption of separate arsenic 'fish consumption only' criteria for freshwater and saltwater.

DEQ proposes using a BCF of 14 for arsenic human health criteria that apply to freshwaters of the state. This BCF is the geometric mean of the data from four finfish studies, which tested rainbow trout (three studies in two publications) and bluegill (one study), as shown in Table 6. A summary of the data from these studies is provided in Appendix A. DEQ's proposed criteria are calculated using a fish consumption rate of 175 g/d, which represents a mixture of fish species. Nearly all of the fish consumed from freshwater, will consist of the muscle tissue of finfish. Therefore, DEQ concludes that a BCF of 14 is a reasonable and protective value to use in calculating arsenic criteria for Oregon's freshwaters. Further discussion of the BCF used for the saltwater criterion is provided in section 2.5.2 below.

**Inorganic Proportion.** Arsenic is present in the environment and in fish tissue in organic and inorganic species. Inorganic arsenic, specifically arsenite (trivalent or As III), is toxic to humans and EPA developed its toxicity data for cancer and other end points based on inorganic arsenic. EPA's recommended human health criteria are for inorganic arsenic, however, the BCF value (44 L/kg) that EPA used in deriving the human health criteria for arsenic are based on total arsenic, not inorganic arsenic. Therefore, some states have also elected to multiply the BCF value by what might be called an "inorganic proportion" factor. For example, the EPA Region 6 Interim Strategy and the State of Colorado use a 30% inorganic variable; Maryland used 4% inorganic in its criteria recalculation.

An EPA (2002) study on fish contaminants in the Columbia River reported the following findings from a TetraTech fish tissue study done in 1996 related to proportion of inorganic arsenic found in fish tissue: (p. 5-78)

- Overall arithmetic average for all composite samples: 6.5%
- Average % inorganic by species ranged from 0.5% in carp to 9.2% in sturgeon
- Anadromous species: about 1.0% on average
- Resident species: about 9% on average

The EPA study also reported that a study of fish tissue in the Willamette River (EVS, 2000) found that an average of 4.2% of the arsenic in carp (whole body) and 3.8 % of the arsenic in bass (fillet) was inorganic arsenic. A risk assessment performed as part of the EPA (2002) study assumed 10% of total arsenic was inorganic for all species.

EPA (2003) states that the consensus in the literature is that approximately 10% of the arsenic found in edible portions of marine fish and shellfish is inorganic arsenic. They also note that because each arsenic species exhibits different toxicities, it may be important to take into account the fraction of total arsenic present in the inorganic and organic forms when estimating the potential risk posed to human health through the consumption of arsenic-contaminated fish and shellfish.

Schoof and Yager (2007) looked at data from 20 studies and found that in freshwater finfish inorganic arsenic was 10% of total arsenic at the 75<sup>th</sup> percentile of the data, with a mean of 7.2%.

DEQ proposes to use a 10% inorganic arsenic fraction to calculate freshwater criteria based on the Columbia River fish contaminant and health risk assessment study (EPA, 2002) and the other information noted above. Recent recommendations on arsenic bioconcentration from EPA also assumed that 10% of the accumulated arsenic was inorganic (arsenic III and V) (personal communications, EPA Headquarters, Nov. 2010). The criteria that result are shown in Table 5 (recommended criteria) above and Table 6 (options considered) below. DEQ observed that the calculation of the water + fish ingestion criterion is not very sensitive to the % inorganic fraction value. Whether DEQ uses a % inorganic fraction of 1, 10 or 30 does not change the water + fish ingestion criterion value. The % inorganic factor does significantly affect the calculated criterion for the fish consumption only criterion.

To incorporate the inorganic factor (IF) into the calculation, DEQ used the revised equations:

$$\text{Water + fish ingestion Criterion } (\mu\text{g/L}) = 1000 \times \frac{\text{RF} \times \text{BW}}{q1 * [\text{DW} + (\text{BCF} \times \text{FCR} \times \text{IF})]}$$

$$\text{Org Only Criterion } (\mu\text{g/L}) = 1000 \times \frac{\text{RF} \times \text{BW}}{q1 * [\text{BCF} \times \text{FCR} \times \text{IF}]}$$

**Toxicity Factors.** DEQ did not review the toxicity data or re-evaluate the cancer slope factor used to derive human health criteria for arsenic. DEQ relies on EPA research to provide toxicity information for its human health criteria. DEQ proposes to use the cancer slope factor in EPA's Integrated Risk Information System (IRIS) data base as of the date of this review, which is  $1.5(\text{mg/kg/day})^{-1}$ . EPA nationally recommended Clean Water Act criteria have not been updated and continue to be based on a cancer slope factor of  $1.75(\text{mg/kg/day})^{-1}$ .

### Section 2.5.2. Inorganic Arsenic Criteria for Saltwater

As with the freshwater criteria, the policy objective for DEQ's proposed arsenic water quality criterion for saltwater is to protect the ability of people to consume fish and shellfish and to reconcile criteria generated by a calculation method, given the limited data, with the presence of naturally occurring levels of arsenic in marine waters. There are uncertainties in the scientific community's current knowledge of the fate and transformation of various species of arsenic in the saltwater environment and in marine and estuarine species. In addition, some forms of arsenic are toxic to humans and others are not. DEQ evaluated the values resulting from two calculation scenarios against the scientific literature describing naturally occurring marine arsenic levels. DEQ concludes that there does not appear to be an unacceptable human health risk associated with eating fish from an unpolluted marine environment, and as a result, it is not desirable to establish an arsenic water quality criterion for saltwater that is below natural marine levels.

Because of the limited data available to calculate criteria for marine waters, DEQ analyzed potential arsenic criteria by considering natural arsenic levels and arsenic cycling in the marine environment and by using EPA's calculation method to estimate potential arsenic risk in 2 ways. In calculating an arsenic criterion for saltwater, the variables used for fish consumption, risk level and toxicity are the same as discussed above for the freshwater criteria. In the two scenarios, DEQ used different bioconcentration and inorganic portion factors for marine waters as summarize here and discussed further below.

#### Summary of analyses for inorganic arsenic criterion for saltwater:

Estimate of natural inorganic arsenic in saltwater	1.0 $\mu\text{g/l}$
Calculated based on BCF 26, inorganic portion 10%	1.0 $\mu\text{g/l}$
Calculated based on BCF 350, inorganic portion 1%	0.8 $\mu\text{g/l}$

Using EPA's calculation method with a BCF of 26 and an inorganic factor of 10% yields an inorganic arsenic criterion of 1.0  $\mu\text{g/l}$ . Using the same calculation method with a BCF of 350 and 1% inorganic factor yields an inorganic arsenic criterion of 0.76 at  $10^{-5}$  risk level. Because the BCF of 350 for the eastern oyster is based on whole body and people generally eat the muscle tissue of marine fish, which accumulated less arsenite, it is a conservative value. A criterion of 1.0  $\mu\text{g/l}$  based on a BCF of 350, an inorganic proportion of 1% and a fish consumption rate of 175 g/d, represents a risk level of  $1.3 \times 10^{-5}$ .

**Natural ocean levels and complexities in the marine environment.** The scientific literature consistently reports natural total arsenic levels of the oceans in the range of 1 to 3  $\mu\text{g/l}$ . (Borak and Hosgood, 2007; EPA, 1976; EPA, 2003; Neff 1997; Tanaka and Santosa, 1995) In a review of arsenic in the marine environment by Neff (1997), the author notes that the concentration of

total arsenic in clean coastal and ocean waters is 1 to 3  $\mu\text{g/l}$  with a mean of about 1.7  $\mu\text{g/l}$ . The dominant form of arsenic in oxygenated marine waters is inorganic arsenic, predominantly arsenate (AsV). The more toxic and potentially carcinogenic arsenite (AsIII) rarely accounts for more than 20% total arsenic in seawater. "In most oxygenated, productive marine ecosystems, arsenite usually represents less than one to no more than about 10-20% of the total arsenic." (Neff, 1997, p. 923)

Tanaka and Santosa provided coastal sea and ocean data for several species of arsenic, including As(V), As(III), total inorganic and total organic. They conclude that in general As(V) is more abundant than As(III), that as much as 50% of the total arsenic in the near shore environment is organic arsenic and that less than 20% of total arsenic is organic in the open ocean. In the northwest Pacific Ocean, average concentrations were 1.1  $\mu\text{g/l}$  inorganic arsenic and 0.1  $\mu\text{g/l}$  organic arsenic. In the southwest Pacific, the concentrations were 1.2  $\mu\text{g/l}$  inorganic and 0.04  $\mu\text{g/l}$  organic arsenic.

Neff (1997) and Tanaka and Santosa (1995) noted that in near shore waters, the concentration of dissolved inorganic arsenic varied seasonally due to biological processes (uptake by phytoplankton). The transformation of arsenic between the metal species also varies based on biological and physical processes.

In a review of seafood arsenic and the implications for human risk, Borak and Hosgood (2007) summarized:

Based on consideration of anticipated dose and anticipated metabolism, it is likely that seafood arsenic does not contribute significantly to arsenic-associated carcinogenicity. The vast majority of arsenic in finfish and crustaceans is in the forms of arsenobetaine, a compound that is essentially inert, non-toxic and excreted without transformation. (p.209)

DEQ has not measured arsenic in Oregon marine waters. From the information available, it would be reasonable to conclude that inorganic arsenic concentrations in Oregon salt waters are likely to be 1.0  $\mu\text{g/l}$  inorganic arsenic and at 1.5 to 2.0  $\mu\text{g/l}$  total arsenic or higher. Therefore, the proposed inorganic arsenic criterion for saltwater of 1.0  $\mu\text{g/l}$  meets the objective that the water quality criterion not be significantly below naturally present inorganic arsenic concentrations.

**Bioconcentration.** DEQ proposed for public comment a fish consumption criterion of 1.0  $\mu\text{g/l}$  inorganic arsenic for saltwater that was calculated using a BCF of 26 and an inorganic proportion factor of 10%. This BCF value incorporates the BCF data for the eastern oyster, the only saltwater species data available, and BCF data for freshwater finfish. DEQ's intent was to represent the fact that people eat a mixture of finfish and shellfish from saltwater. However, there is no BCF data available for saltwater finfish. In the absence of this data, DEQ analyzed two scenarios to represent bioconcentration for all marine finfish and shellfish; one uses a combination of all the BCF data available (BCF = 26) and the second uses only the eastern oyster data (1 study BCF value = 350; see Table 6 above). DEQ concluded that it is reasonable to use the combined freshwater finfish (vertebrate) and marine oyster (invertebrate) data to represent the variety of species consumed from saltwater systems in Oregon. Because the BCF values are for whole body tests, they are a conservative representation of consumption of inorganic arsenic from primarily muscle tissue consumption of saltwater finfish. In addition, because the BCF values are for total arsenic, DEQ also applied an inorganic proportion factor. The inorganic portion used for the saltwater criterion is discussed in the next sub-section.

Part of DEQ's hesitation to rely solely on the oyster data to represent the bioconcentration in marine finfish is that the oyster is an invertebrate. In freshwater, scientific literature indicates an apparent difference in bioconcentration between invertebrates and vertebrates. Mean BCFs for freshwater invertebrates (trophic level 2 species) ranged from 2 to 22 L/kg, while for freshwater

fish, mean BCFs ranged from 0.048 to 14 L/kg (EPA, 2003). Also, EPA (2003) notes that one BAF study done for a marine fish species (mullet) showed a BAF of 3.3 wet weight for total arsenic (Lin et al. 2001). This study may have been done at a high water arsenic concentration.

Data from Neff (1997) also indicates a potential difference in total arsenic between bivalves and finfish, but also shows a high degree of variability. Table 1 from the Neff (1997) paper summarizes total arsenic concentrations ( $\mu\text{g/g}$  dry weight) in the whole body or muscle tissues of marine organisms, including the following:

	No. Samples	Geometric mean	Range
Bivalves	151	10.44	<0.6 - 214
Fish	156	5.59	0.05-449.5

The highest concentrations of arsenic appear to be present in tissues of marine animals that feed primarily on phytoplankton or macroalgae, including planktonic crustaceans, bivalve mollusks, herbivorous snails and some polychaete worms. (Neff, 1997)

Because there may be a difference in bioconcentration between mollusks (invertebrates) and finfish, and because mollusks represent a small portion of total seafood consumption, relying solely on the oyster BCF data is likely to be overly conservative. Schoof and Yager (2007) provide a summary of seafood consumption in the U.S. population (data from the USEPA, 2002 relying on the 1994-96 and 1998 USDA surveys of food intake) showing that estuarine mollusks (oyster, clam and scallop) comprised about 3 percent and all estuarine and marine mollusks together (oyster, clam, scallop, mussels, squid and octopus) represent about 13 percent of total fish and shellfish consumption.

In a review of information on bioaccumulation of arsenic in aquatic organisms, EPA recognizes that the hypothesis that BAFs based on total arsenic may not represent all freshwater ecosystems, *and especially saltwater ecosystems*. Due to variations in the species of arsenic present in the water and tissues of organisms, this remains an issue requiring further consideration. (EPA, 2003, p. 34)

Schoof and Yager (2007) looked at 20 studies that provided data on total and inorganic arsenic in seafood. They found that mean concentrations of inorganic arsenic were approximately 10 to 20 ng/g wet weight for freshwater, anadromous and marine fish, whereas crustaceans and mollusks had mean inorganic arsenic concentrations of 40 to 50 ng/g. This data indicates that crustaceans and mollusks tend to accumulate more inorganic arsenic than anadromous or marine fish.

Marine animals take up very little inorganic arsenic from seawater but can bioaccumulate organic arsenic from their food. (Neff 1997; Borak 2007). While marine invertebrates and fish may contain high concentrations of arsenic, nearly all the arsenic in the tissues of marine animals is organic, particularly arsenobetaine. (Neff 1977, p.923; Borak and Hosgood 2007) Arsenobetaine, the most abundant organoarsenic compound in seafoods, is not toxic or carcinogenic to mammals. Little of the organoarsenic accumulated by humans from seafood is converted to toxic inorganic arsenite. In addition, arsenobetaine and other organoarsenic compounds are excreted rapidly by mammals, and are not toxic to human consumers of fishery products. Therefore, marine arsenic represents a low risk to human consumers of fishery products.

Neff (1997) suggested that the USEPA "...criterion should be revised to reflect the actual concentrations of total and arsenite arsenic in the ocean and in the tissues of marine organisms consumed by humans." While EPA did revise their recommended arsenic criteria for human health to inorganic arsenic, the BCF data is still for total arsenic.

**Inorganic proportion.** There is a growing body of literature indicating that while saltwater organisms may contain more total arsenic than freshwater fish, the dominant form in marine species is organic arsenic as opposed to inorganic arsenic. (EPA 2003; Neff 1997; Schoof and

Yager 2007; Tanaka and Santosa 1995; TetraTech 1996, IN EPA 2002; Williams et.al. 2006). Tissues of marine invertebrates and fish contain high concentrations of arsenic, usually in the range of about 1 to 100  $\mu\text{g/g}$  dry weight, most of it in the form of organoarsenic compounds, particularly arsenobetaine. (Neff, 1997)

Inorganic arsenic, found mainly as arsenate and to a much lesser extent as arsenite, is the predominant form of arsenic in seawater but inorganic compounds comprise only a small proportion of the total arsenic in seafood. An analysis of five types of ocean finfish and shrimp found that inorganic arsenic was less than 0.1% of the total arsenic (Schoof et. al., 1999 in Borak and Hosgood, 2007). Other literature has reported values less than 3% and more recently surveys report values less than 1%. (Borak and Hosgood, 2007)

An EPA (2002) study on fish contaminants in the Columbia River summarized the findings of a TetraTech fish tissue study (1996) that found the following related to proportion of inorganic arsenic found in fish tissue: (p. 5-78)

- Overall arithmetic average for all composite samples: 6.5%
- Average % inorganic by species ranged from 0.5% in carp to 9.2% in sturgeon
- Anadromous species: about 1.0% on average
- Resident species: about 9% on average

DEQ notes that the inorganic portion for the anadromous fish (salmon), which spend most of their life and gain most of their growth in marine waters, was lower than the portion in resident, freshwater species. EPA noted that these findings were consistent with the literature, which shows low percentages of inorganic arsenic levels for most saltwater fish species.

EPA (2003) recognized that because each arsenic species exhibits different toxicities, it may be important to take into account the fraction of total arsenic present in the inorganic and organic forms when estimating the potential risk posed to human health through the consumption of arsenic-contaminated fish and shellfish. The document states, "Clearly only a very small percentage of inorganic arsenic exists in the soft tissues of these organisms [marine bivalve mollusks]...", most often less than 1%; the bulk of the arsenic being arsenobetaine. EPA cites several studies. EPA also states that it is increasingly evident that methylation is critical in controlling biological fate and effects of arsenic.

Neff (1997) states,

*"Inorganic arsenic represents between about 0.5 and 1% of the total arsenic in the edible portions of most marine invertebrates and fish examined to date (Francesconi and Edmonds, 1993). The fraction of total arsenic that is inorganic tends to decrease as the concentration of total arsenic in the tissues increases. Concentrations of inorganic arsenic in the edible portions of marine invertebrates and fish from uncontaminated marine environments generally range from less than 0.001 to about 0.5  $\mu\text{g/g}$  wet weight (Francesconi and Edmonds, 1993). These concentrations are below the MPCs for total arsenic in seafoods set by most countries." (p. 922)*

In marine waters, portions of organic and inorganic arsenic may be influenced by biological activity temperature and other variables. (Neff; Tanaka, 1995). Tanaka shows seasonal variation in the inorganic and organic proportions of total arsenic.

Schoof & Yager, 2007. Authors looked at 20 studies that provided data on total and inorganic arsenic in seafood, noting that a number of recent had become available. Their findings are summarized in the following table.

**Inorganic arsenic as a % of total arsenic in seafood measured as ng/g wet weight**

	<u>Mean</u>	<u>Range</u>
Freshwater	7.2	0.5-26.6
Anadromous fish	1.1	0.03-3.04
Marine fish	1.0	0.001-6.9
Crustaceans	1.3	0.001-7.3
Mollusks	1.8	0.04-6.5

DEQ concludes that the above information supports using an inorganic proportion of 1% to calculate an inorganic criterion for saltwater based on the oyster BCF of 350. The results of this calculation are shown at the beginning of this section.

**Section 2.6. Options Considered for Revising the Arsenic Criteria**

DEQ initially considered three primary alternatives for deriving arsenic criteria as an alternative to EPA's current recommended criteria:

1. Re-calculation of the federal criteria using Oregon appropriate variables,
2. Use of the MCL value for drinking water in some manner, and a
3. Natural background based approach.

Table 7 shows the possible criteria values under these three approaches.

<b>Table 7. Arsenic Criteria Options Considered</b> ( $\mu\text{g/l}$ , inorganic arsenic)		
<b>Approach</b>	<b>Estimated Water + fish ingestion (freshwater only)</b>	<b>Estimated Fish consumption only</b>
OR recalculation: BCF=14, FCR=175, % inorganic=10, CSF=1.5, risk= $1 \times 10^{-6}$	0.021	0.19 for freshwater
OR recalculation: BCF=14, FCR=175, % inorganic=10, CSF=1.5, risk= $1 \times 10^{-4}$	2.1	19 for freshwater
OR recalculation: BCF=26, FCR=175, % inorganic=10, CSF=1.5, risk= $1 \times 10^{-5}$	Not applicable	1.0 for saltwater
OR recalculation: BCF=350, FCR=175, % inorganic=1, CSF=1.5, risk= $1 \times 10^{-5}$	Not applicable	0.8 for saltwater
OR recalculation: BCF=350, FCR=175, % inorganic=1, CSF=1.5, risk= $1.3 \times 10^{-5}$	Not applicable	1.0 for saltwater
Use the water + fish value for both freshwater criteria	2.1	2.1
MCL fraction: $\text{MCL} \times 0.25$	2.5	2.5
Statewide default natural background for freshwater	1-3 total arsenic	1-3 total arsenic
Natural background for saltwater	Not applicable	2 total arsenic 1 inorganic arsenic

Notes: 1. MCL = 10  $\mu\text{g/l}$  total arsenic. 2. HHC will be for inorganic arsenic.  
3. The current IRIS CSF is  $1.5(\text{mg/kg/day})^{-1}$ .

**Option 1: Re-calculated Criteria using Oregon Appropriate Variables.** Option 1 is Oregon's proposed approach, as discussed in the preceding sections and shown in Table 5. DEQ concludes that by using EPA's calculation formulas with locally appropriate values, this option provides a rationale for deriving criteria that is scientifically defensible and can be clearly explained to the public.

DEQ's initial proposal was based on limited data and followed a recalculation method that had been used by other states and described by one EPA Regional office. DEQ received comment that the bioconcentration factor (1) used in that recalculation was not supportable for Oregon. In considering the comment and relooking at the information, DEQ decided that the BCF of 1 was indeed too low for Oregon and not supported by the currently available literature.

DEQ is proposing separate fish consumption only criterion for saltwater, as discussed in the preceding section. Bioconcentration is much greater in marine mollusks than freshwater finfish and therefore a different BCF is appropriate. As with the freshwater criteria, DEQ used a higher risk level ( $10^{-5}$  rather than  $10^{-6}$ ) to calculate the fish consumption only saltwater criterion because 1) the fish consumption rate represents high exposure, and 2) arsenic occurs in marine waters due to natural sources (Tanaka, 1995; National Academy of Sciences, 1972; EPA, 2003).

### **Option 2: Use a Fraction of the Maximum Contaminant Level from the Safe Drinking Water Act to Derive Oregon's Arsenic Criteria.**

The second approach DEQ considered was to use a combination of the maximum contaminant level (MCL) for drinking water and the EPA criteria calculation method to represent exposure through fish tissue. Nearly half of the states have utilized the MCL value of 10 for their arsenic criterion in place of EPA's national criteria recommendations. DEQ believes that using a fraction of the MCL (10) as the water quality criteria is preferable over adoption of the MCL due to the additional exposure to arsenic through consumption of fish tissue.

An MCL is the maximum level of a contaminant allowed in drinking water delivered to the tap (post treatment). MCLs are enforceable standards developed by EPA under the Safe Drinking Water Act. MCLs are set as close to maximum contaminant level goals (MCLGs) as feasible using the best available treatment technology and taking cost into consideration. MCLGs are non-enforceable public health goals that describe the level of a contaminant in drinking water below which there is no known or expected risk to health and allow for a margin of safety. For all carcinogens, MCLGs are set to zero. On January 22, 2001, EPA revised its maximum contaminant level (MCL) for arsenic from 50 to 10  $\mu\text{g}/\text{L}$ , and established a date of January 23, 2006, for all public water supply systems to achieve compliance with the revised MCL.

### **Option 3: Natural Background**

Under this approach based, DEQ would establish a "default" statewide natural background level using the best currently available information on natural background levels of arsenic in the State. The human health criteria for arsenic would then be set at that level. This would prevent widespread identification of waters as "impaired" due to natural sources. This approach could reasonably lead to a water + fish ingestion criterion of 1 to 3  $\mu\text{g}/\text{l}$ . This criterion would be well below the drinking water MCL of 10  $\mu\text{g}/\text{l}$ , and is near the 2.1  $\mu\text{g}/\text{l}$  value calculated to protect fish consumption at a consumption rate of 175 g/d and a risk level of  $1 \times 10^{-4}$ .

A variation on this approach would be to add to the default natural background level, a *de minimis* or insignificant increment for assimilative capacity, making the criterion slightly higher (for example, 1.5 to 2.5). The purpose of setting the criteria slightly above natural background would be to provide some assimilative capacity for mixing in localized areas. This would allow some discharge of arsenic at concentrations that have been increased due to evaporative cooling, for example, which can occur even if there has been no addition of mass. The discharge would be required to meet the criteria at the edge of an assigned mixing zone.

A natural background option for saltwater would lead to a criterion of approximately 2  $\mu\text{g}/\text{l}$  total arsenic or 1  $\mu\text{g}/\text{l}$  inorganic arsenic. Natural background levels for saltwater are discussed in section 2.5.2 above.

While DEQ did not base its criteria on natural background levels, it was part of our policy objective to account for natural arsenic levels as we selected arsenic criteria, particularly for the organism only criteria established to protect the consumption of fish from Oregon waters. We cannot control natural sources of arsenic and the health risks are not sufficiently high to suggest that people should not eat fish from Oregon streams or coastal waters.

**Additional Considerations**

The following additional alternatives could be used for specific water bodies or regions and combined with the three statewide options discussed above. These options are available regardless of what statewide criteria are adopted.

1. Where natural conditions exceed the revised arsenic criteria, DEQ may find it appropriate to develop site specific criteria.
2. Apply the fish consumption only criterion where public domestic water supply is not a designated use and revise beneficial uses in a follow up rulemaking to more narrowly designate water bodies considered suitable for drinking water supply.

# Chapter 3. DEQ's Proposed Arsenic Reduction Policy

DEQ proposes to adopt the following arsenic reduction policy into its water quality standards in addition to the numeric criteria discussed in Chapter 2 above. The goal of this provision is to ensure that Oregon's proposed numeric water + fish ingestion criterion for arsenic, which is intended to account for natural conditions, does not unintentionally allow preventable human health risk due to anthropogenic loading of arsenic from existing or new sources.

DEQ is proposing revised numeric arsenic criteria of 2.1 µg/l for both the fish + water ingestion and fish consumption only criteria for freshwater and a fish ingestion criterion of 1.0 for saltwater. While these proposed numeric criteria protect human health at an acceptable level given the presence of natural sources of arsenic in the state, it is the state's policy to maintain the lowest added human health risk from anthropogenic sources of inorganic arsenic practicable, even when ambient inorganic arsenic concentrations are below the numeric criteria. This policy is targeted to dischargers that add inorganic arsenic to Oregon waters and have the potential, due to their location, to impact a public drinking water supply.

The proposed criteria are based on a fish consumption rate of 175 g/day, which is protective of Oregon fish consumers and risk levels that are considered acceptable and protective. However, the criteria and especially the fish + water criterion are based on a higher risk level than Oregon uses for the rest of its human health criteria ( $10^{-6}$ ). Due to concerns about drinking water exposure, the approach proposed below is targeted to address sources that impact drinking water supplies.

## Section 3.1. Proposed Rule Language:

(4) Arsenic Reduction Policy: The inorganic arsenic criterion for the protection of human health from the combined consumption of organisms and drinking water is 2.1 micrograms per liter. While this criterion is protective of human health and more stringent than the federal maximum contaminant level (MCL) for arsenic in drinking water, which is 10 micrograms per liter, it nonetheless is based on a higher risk level than the Commission has used to establish other human health criteria. This higher risk level recognizes that much of the risk is due to naturally high levels of inorganic arsenic in Oregon's waterbodies. In order to maintain the lowest human health risk from inorganic arsenic in drinking water, the Commission has determined that it is appropriate to adopt the following policy to limit the human contribution to that risk.

(a) The arsenic reduction policy established by this rule section does not become applicable for purposes of ORS chapter 468B or the federal Clean Water Act unless and until the numeric arsenic criteria established by this rule are approved by EPA pursuant to 40 CFR 131.21 (4/27/2000).

(b) It is the policy of the Commission that the addition of inorganic arsenic from new or existing anthropogenic sources to waters of the state within a surface water drinking water protection area be reduced the maximum amount feasible. The requirements of this rule section [OAR 340-041-0033(4)] apply to sources that discharge to surface waters of the state with an ambient inorganic arsenic concentration equal to or lower than the applicable numeric inorganic arsenic criteria for the protection of human health.

(c) The following definitions apply to this section [OAR 340-041-0033(4)]:

(A) “Add inorganic arsenic” means to discharge a net mass of inorganic arsenic from a point source (the mass of inorganic arsenic discharged minus the mass of inorganic arsenic taken into the facility from a surface water source).

(B) A “surface water drinking water protection area,” for the purpose of this section, means an area delineated as such by DEQ under the source water assessment program of the federal Safe Drinking Water Act, 42 U.S.C. § 300j-13. The areas are delineated for the purpose of protecting public or community drinking water supplies that use surface water sources. These delineations can be found at DEQ’s drinking water program website.

(C) “Potential to significantly increase inorganic arsenic concentrations in the public drinking water supply source water” means:

(i) to increase the concentration of inorganic arsenic in the receiving water for a discharge by 10 percent or more after mixing with the harmonic mean flow of the receiving water; or

(ii) as an alternative, if sufficient data are available, the discharge will increase the concentration of inorganic arsenic in the surface water intake water of a public water system by 0.021 micrograms per liter or more based on a mass balance calculation.

(d) Following the effective date of this rule, applications for an individual NPDES permit or permit renewal received from industrial dischargers located in a surface water drinking water protection area and identified by DEQ as likely to add inorganic arsenic to the receiving water must include sufficient data to enable DEQ to determine whether:

(A) The discharge in fact adds inorganic arsenic; and

(B) The discharge has the potential to significantly increase inorganic arsenic concentrations in the public drinking water supply source water.

(e) Where DEQ determines that both conditions in subsection (d) of this section (4) are true, the industrial discharger must develop an inorganic arsenic reduction plan and propose all feasible measures to reduce its inorganic arsenic loading to the receiving water. The proposed plan, including proposed measures, monitoring and reporting requirements, and a schedule for those actions, will be described in the fact sheet and incorporated into the source’s NPDES permit after public comment and DEQ review and approval. In developing the plan, the source must:

(A) Identify how much it can minimize its inorganic arsenic discharge through pollution prevention measures, process changes, wastewater treatment, alternative water supply (for groundwater users) or other possible pollution prevention and/or control measures;

(B) Evaluate the costs, feasibility and environmental impacts of the potential inorganic arsenic reduction and control measures;

(C) Estimate the predicted reduction in inorganic arsenic and the reduced human health risk expected to result from the control measures;

(D) Propose specific inorganic arsenic reduction or control measures, if feasible, and an implementation schedule; and

(E) Propose monitoring and reporting requirements to document progress in plan implementation and the inorganic arsenic load reductions.

(f) In order to implement this section, DEQ will develop the following information and guidance within 120 days of the effective date of this rule and periodically update it as warranted by new information:

(A) A list of industrial sources or source categories, including industrial stormwater and sources covered by general permits, that are likely to add inorganic arsenic to surface waters of the State.

(i) For industrial sources or source categories permitted under a general permit that have been identified by DEQ as likely sources of inorganic arsenic, DEQ will evaluate options for reducing inorganic arsenic during permit renewal or evaluation of Stormwater Pollution Control Plans.

(B) Quantitation limits for monitoring inorganic arsenic concentrations.

(C) Information and guidance to assist sources in estimating, pursuant to paragraph (d) (C) of this section, the reduced human health risk expected to result from inorganic arsenic control measures based on the most current EPA risk assessment.

(g) It is the policy of the Commission that landowners engaged in agricultural or development practices on land where pesticides, fertilizers, or soil amendments containing arsenic are currently being or have previously been applied, implement conservation practices to minimize the erosion and runoff of inorganic arsenic to waters of the State or to a location where such material could readily migrate into waters of the State.

### Section 3.2. Implementation of the Arsenic Reduction Policy

This section describes how DEQ intends to implement the above proposed rule. Nothing in this arsenic reduction policy replaces or supersedes technology-based permit requirements, permit limits based on numeric arsenic criteria or antidegradation requirements. All of these otherwise applicable criteria and policies continue to apply.

DEQ recognizes that we have not specified an analytical method for inorganic arsenic or the quantitation limit (QL) that will be required for permittee monitoring. Because the proposed numeric criteria for arsenic are for the inorganic form, this information will need to be developed regardless of whether or not this reduction policy is adopted.

#### Point Sources – Industrial Sources:

1. Applications for new or renewed individual NPDES permits submitted to DEQ after the effective date of this rule by industrial dischargers that are required to submit arsenic data with their permit application, or are otherwise identified by DEQ as likely to add inorganic arsenic to their wastewater, **and** that discharge to a water body within a drinking water protection area delineated by DEQ for a surface water intake, shall submit with their permit application sufficient data to allow DEQ to make the determinations described in #3 below. This will include source water and effluent inorganic arsenic concentration and flow data and may also include ambient river data.
  - a. A discharger that has sufficient effluent data to demonstrate that its effluent concentration of inorganic arsenic is below DEQ's quantitation limit or below the

ambient river concentration immediately upstream of the discharge may use that information to demonstrate that the discharge does not have the potential to impact the arsenic concentration in a downstream public water supply.

2. DEQ will use the data to determine:
  - a. whether the discharger is adding a quantifiable load of inorganic arsenic to their wastewater (i.e. a quantifiable concentration of inorganic arsenic in the discharge is greater than the inorganic arsenic load taken in from a surface water intake source); **and**
  - b. whether the added load has the potential to increase the concentration of inorganic arsenic in a downstream public drinking water supply. DEQ will determine that a discharge has the potential to increase the concentration of inorganic arsenic in a downstream drinking water supply intake if the source increases the concentration of inorganic arsenic in the river after dilution (near field/point of discharge mixing analysis) by 3% or more, unless the source can demonstrate that their arsenic contribution will not increase the arsenic concentration in the downstream water supply by more than 0.023 µg/l.
3. If the Department finds that the facility is adding inorganic arsenic and that the added load is impacting a public drinking water supply, the permittee shall develop an arsenic reduction plan, which will be incorporated into its NPDES permit subject to DEQ review and public comment. The source shall include the following in their plan:
  - a. Identify how much it can minimize its arsenic discharge through pollution prevention measures, process changes, wastewater treatment, alternative water supply sources or other possible pollution prevention and/or control measures.
  - b. Evaluate the costs, technical and economic feasibility and environmental impacts of the identified arsenic reduction and control measures.

Note 1: It is important to evaluate whether a potential arsenic reduction measure, such as a chemical substitution, represents an equal or worse environmental risk or other environmental impact.

Note 2: DEQ recognizes that evaluating water supply options and the environmental impacts of those is complex and there are many issues to consider other than the arsenic loading. If the source of arsenic is groundwater, there may be few if any feasible options for reduction.
  - c. Estimate the reduced arsenic load and human health risk expected to result from the control measures.
  - d. Propose specific inorganic arsenic reduction or control measures, if feasible, and a schedule for implementing them.
  - e. Specify monitoring and reporting requirements related to implementing the plan and the resulting effluent arsenic load reductions.
4. DEQ will identify factors that the permittee and the agency should consider in weighing the technical and economic feasibility of an inorganic arsenic reduction measure against the reduced human health risk that is expected to result and deciding which measures to implement.
5. If the timing of a permit renewal is such that the facility has not had sufficient time to collect the required data or develop an arsenic reduction plan prior to permit issuance, the permit will include the data collection and/or planning requirements and a reopener clause, which will allow DEQ to incorporate the proposed plan/measures into the permit prior to the next renewal.
6. Arsenic reduction plans and their implementation will be reviewed at each permit renewal to evaluate progress in implementation actions and inorganic arsenic reductions and determine whether and new measures are feasible and/or proposed.
7. There are existing procedures for requesting the re-consideration of a permit that can be used by persons who have grounds to believe that either the data and analysis or the reduction measures included in the permit are inadequate.

### Point sources – Publicly Owned Treatment Works (POTWs)

1. All major POTWs are required to analyze their effluent for arsenic and submit that data to DEQ as part of their permit renewal application.
2. Arsenic III (the primary inorganic form) is included on Oregon's Priority Persistent Pollutant list developed under SB737. DEQ will rely on the water quality criteria and the "SB 737" requirements to address potential arsenic contributions from POTWs. Under "SB 737," the 52 largest POTWs, including all major municipal dischargers, will be required to test for arsenic III in their effluent. If the effluent concentration exceeds the initiation level specified in rule, the facility will be required to develop and implement a pollutant reduction plan for arsenic.

### Point Sources – Other

1. Wood treating facilities – DEQ will incorporate the following into our renewal of industrial stormwater permits for wood treating facilities:
  - Review data on arsenic levels in stormwater runoff
  - Determine the sources of the arsenic on the site
  - Require the facility to identify measures that could be taken to reduce arsenic loading, including chemical substitution, stormwater management and erosion control practices, stormwater treatment, soil testing and remediation, chemical storage and disposal practices, and others.
  - Evaluate the measures, considering: a) potential for reduction of arsenic discharge, b) cost and c) potential environmental impacts (particularly for chemical substitutions), and incorporate appropriate measures into the permit.
2. Municipal stormwater management – DEQ will incorporate the following into our municipal stormwater permitting program:
  - DEQ will review data on inorganic arsenic levels in stormwater runoff and/or UIC wells to determine whether municipal stormwater is a significant source of inorganic arsenic.
  - If it is determined to be a significant source, DEQ will determine whether it is possible to identify the source(s) of the arsenic and whether additional measures or best management practices could be implemented that would reduce the arsenic loading.

### Nonpoint Source Options:

1. Use the agency-wide Toxics Reduction Strategy to evaluate whether any of the following actions would be: a) likely to reduce inorganic arsenic concentrations in surface water drinking water protection areas, or in waters that exceed the water quality criteria for arsenic, and b) cost effective:
  - a limit on the amount of arsenic in fertilizers, pesticides and/or wood treating chemicals, or a ban on products containing arsenic if there are still such products in use;
  - treated wood and/or chemical collection/take back programs,
  - stormwater management in areas with large amounts of treated wood present, and/or
  - enhanced erosion control practices on lands where soil inorganic arsenic levels are elevated.
2. Recommend that adequate control of runoff and erosion from urban development and agricultural lands be implemented for multiple benefits. One benefit would be to prevent arsenic and other toxic pollutants that adhere to soil particles from entering waterways. Some contaminants, such as arsenic, are no longer widely used, but may have built up in soils in certain locations from past use. In addition, such controls would also reduce nutrient (i.e.

phosphorus) and sediment loading from urban and agricultural lands and therefore provide multiple benefits to fish and aquatic life and the quality of Oregon waters.

3. Construction stormwater general permit. Erosion and stormwater control practices should be employed to reduce loading of sediment and chemicals attached to sediments to the stream.

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### Appendix A. Bioconcentration Factor Data

Bioconcentration data and bioconcentration factor (BCF) options used to derive water quality criteria for Oregon.

Water type	Species	BCF
<b>All freshwater and saltwater BCFs</b>		
Freshwater	Bluegill, <i>lepomis macrochirus</i>	4
Freshwater	Trout, <i>oncorhynchus mykiss</i>	10
Freshwater	Trout, <i>oncorhynchus mykiss</i>	17
Freshwater	Trout, <i>oncorhynchus mykiss</i>	27
Saltwater	Eastern oyster, <i>crossostrea virginica</i>	350
	<b>Geometric mean</b>	26
<b>All freshwater fish BCFs</b>		
Freshwater	Bluegill, <i>lepomis macrochirus</i>	4
Freshwater	Trout, <i>oncorhynchus mykiss</i>	10
Freshwater	Trout, <i>oncorhynchus mykiss</i>	17
Freshwater	Trout, <i>oncorhynchus mykiss</i>	27
	<b>Geometric mean</b>	14
<b>Saltwater (Eastern oyster) BCF</b>		
Saltwater	Eastern oyster, <i>crossostrea virginica</i>	350

From EPA, 2010. Personal communication, EPA Headquarters staff, November 2010.

Ambient Water Quality Criteria for Arsenic – EPA 440/5-80-021 and Ambient Water Quality Criteria for Arsenic – 1984, published 1985 refers to Barrows, et al. 1980, Ann Arbor, MI. pp. 379-392. Three different populations of immature bluegill; BCF of 4 reported.

Ambient Water Quality Criteria for Arsenic – EPA 440/5-80-021 and Ambient Water Quality Criteria for Arsenic – 1984, published 1985 refers to Zarogian and Hoffman, 1982, Environmental Monitoring and Assessment 1:345-358. EPA document refers to BCF value for oyster of 350.

<b>Rainbow Trout studies</b>		
	Total arsenic water concentration in µg/l	Total arsenic BCF (wet weight)
McGeachy and Dixon, 1990		
At 5.3°C	10	20
At 15.3°C	10	17
Rankin and Dixon, 1994.		
At 5.3°C	10	27

From EPA, 2010. Personal communication, EPA Headquarters staff, November 2010.

## Appendix B. Supplemental Information on Arsenic

From: *Impact of Land Disturbance on the Fate of Arsenical Pesticides*, Carl E. Renshaw,\* , Benjamin C. Bosticka, Xiahong Fenga, Christine K. Wonga, Elizabeth S. Winstona, Roxanne Karimib, Carol L. Foltb and Celia Y. Chenb. 2005.

### *Fate and transport in the environment*

Inorganic arsenic (As) occurs in two dominant redox states, arsenate (As(V)) and arsenite (As(III)), both highly toxic and carcinogenic (Hopenhayn 2006; Vaughan 2006). The oxidized form, arsenate, behaves chemically similarly to phosphate (P(V)) in the environment, as the two species display similar coordination chemistry and both readily bond with soil solids like iron oxides and clay particles (Stollenwerk 2003). Lab and field studies show that arsenate, like phosphate, sorbs to iron plaques that form on plant roots (Blute, Brabander et al. 2004; Liu, Zhu et al. 2006). Plants generate these plaques by pumping oxygen from the atmosphere to their roots, creating microoxic regimes in otherwise anoxic sediments (Taylor, Crowder et al. 1984).

However, a number of factors interfere with our ability to predict the mobility of As when plants are present. Arsenate, unlike phosphate, easily and commonly shifts redox states in the environment. The reduced form of As, arsenite, tends to be more mobile than arsenate and does not as strongly bond with iron oxides or natural organic matter at low and neutral pH (Stollenwerk 2003; Buschmann, Kappeler et al. 2006). In the root zone, dissolved organic carbon (DOC) exuded by plants will create high oxygen demand that result in anoxic conditions where DOC could then reduce arsenate to arsenite. Additionally, natural organic matter may compete with arsenate for sorption sites on iron oxides (Redman, Macalady et al. 2002). Both As reduction and competitive sorption may lead to greater As mobility. Conversely, both species of inorganic As sorb to natural organic matter, indicating that plants may enhance As retention up to some threshold (Buschmann, Kappeler et al. 2006).

### *Potential nonpoint sources of arsenic*

Our observation of high As and Pb concentrations in the drainages down gradient of the tilled orchard is consistent with a recent regional analysis of stream sediment As and Pb concentrations that found a positive association between stream sediments that contain high As and Pb concentrations and areas inferred to have used arsenical pesticides extensively (Robinson and Ayuso, 2004). Our work extends this regional analysis by demonstrating that: (i) at least below the tilled field the As and Pb were transported to the drainage in two discrete events, with the later mobilization event occurring well after the application of the arsenical pesticides; and (ii) the masses of As and Pb apparently missing from the tilled field and present in the down gradient drainage are consistent with transport due to physical erosion associated with tilling. Most previous work investigating As mobilization due to physical erosion has focused on As contamination due to the erosion of As-rich ores (Black et al., 2004; Oyarzun et al., 2004; Savage et al., 2000). However, tilling-induced mobilization similar to postulated here has recently been documented for other strongly sorbing pesticides (Wu et al., 2004). In contrast, little horizontal redistribution of As has been observed in the untilled As-contaminated soils underlying cattle tick dip sites (Kimber et al., 2002)...

Finally, while this work only considers the effect of tilling on the mobilization of residual arsenical pesticides, our work shows that the Pb and As are bound to small and presumably highly mobile particles. It is therefore likely that other types of land disturbances will also mobilize significant amounts of Pb and As in lands where arsenical pesticides were used, particularly over longer timescales. In southern New Hampshire, for example, former orchard land is currently being rapidly developed and urbanized. Our results suggest that as this land is developed, attention should be given to the possibility of mobilizing previously immobile reservoirs of Pb and As.

**Total Arsenic in Drinking Water Supplies in Oregon (ug/l)**

	All "Surface Water"	Selected Surface Water*	Groundwater under direct influence of SW	Groundwater (see table below)
Minimum	0.5	0.5	0.58	0.1
Maximum	9.0	5.7	14	411
Average	3.0	1.6	4.87	8.8
# samples	45	24	11	1642

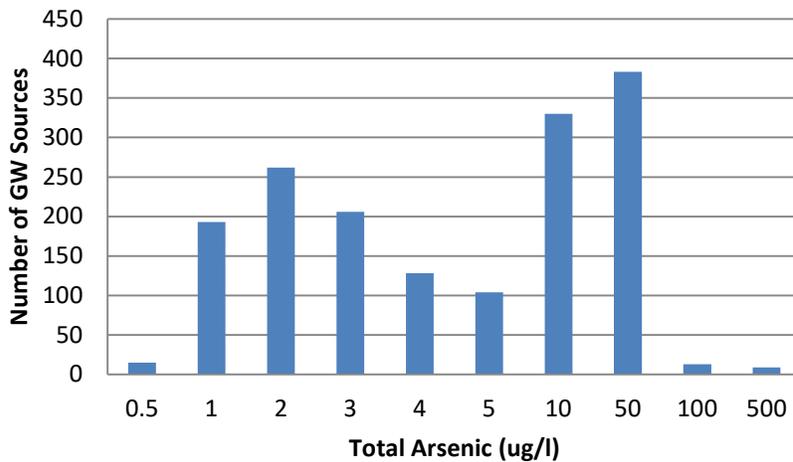
\* Sources that use only surface water and do not include well water as part of their supply.

Note 1: This data is for finish water, which means these are the levels after the raw water has been treated.

Note 2: This data includes only sources with detectable levels of arsenic (0.5 ug/l or more). There are additional sources where arsenic was not detected. Therefore, the data above do not represent the average of arsenic levels in surface water supplies throughout Oregon, but simply represent commonly occurring levels.

**From:** Drinking Water data base, Oregon, May 2009 query

**Arsenic in GW sources in Oregon**



**From:** Drinking Water data base, Oregon, May 2009 query.

Number of GW samples with arsenic values above previous value and up to value shown (i.e. 0.01–0.5; 0.51-1; 1.01-2, etc.).

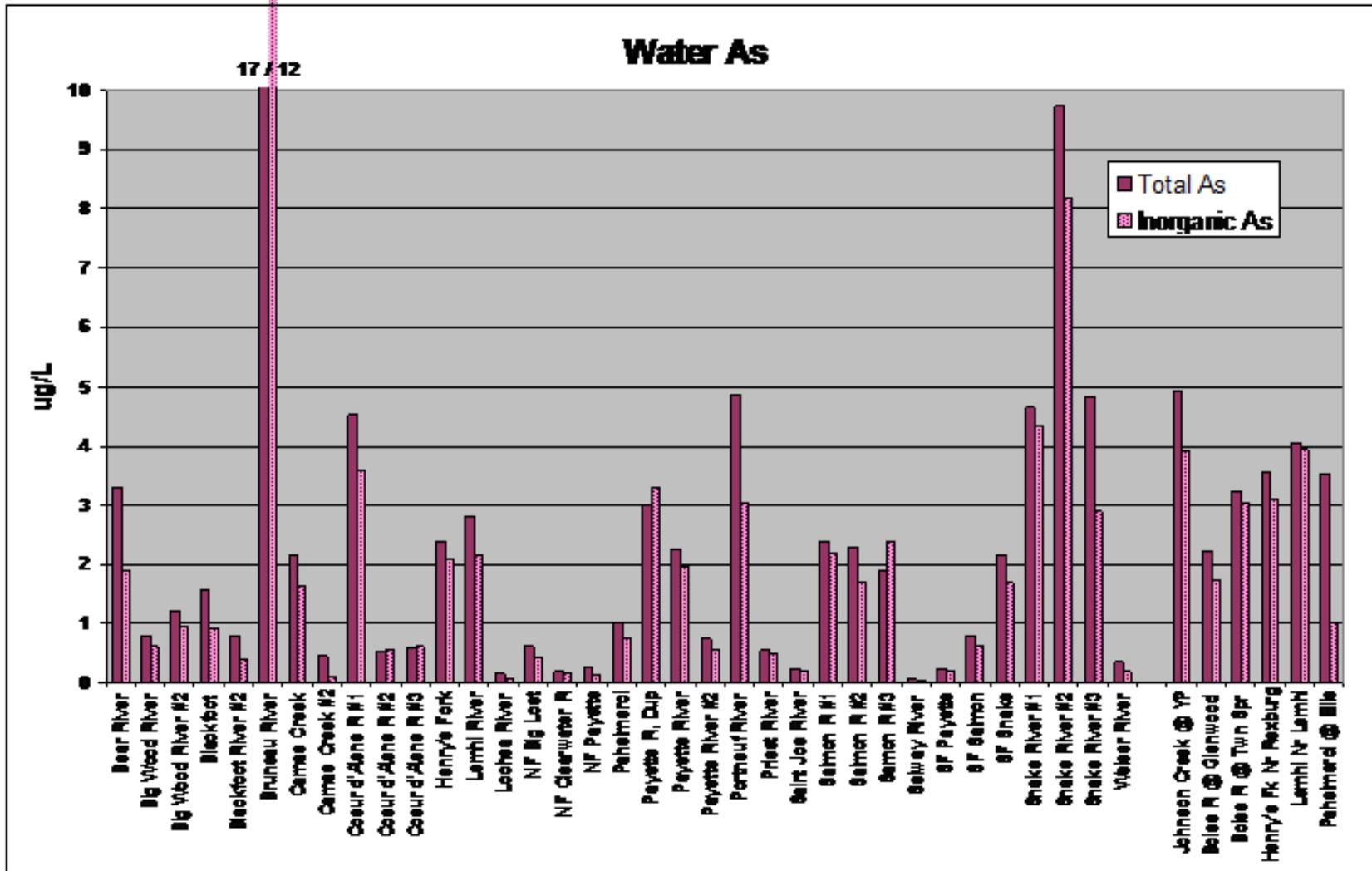


Figure 1. Data on total and inorganic arsenic from Idaho.

2008/09 total arsenic and inorganic arsenic data from 40 sites on major rivers across Idaho ranged from 25% to 100% inorganic arsenic; the mean was 75% inorganic. Idaho DEQ.