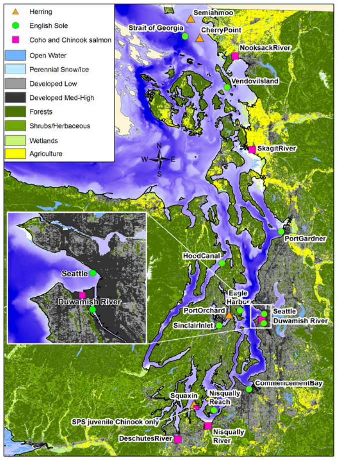
Current conditions, time trends and recovery targets for toxic contaminants in Puget Sound fish: the *Toxics in Fish* Dashboard Indicator

James E. West*, Sandra M. O'Neill‡, Jennifer Lanksbury* Gina M. Ylitalo‡, and Scott Redman^



Stations are classified as "urban" or "non-urban" based on their proximity to developed land.

Recovery Targets for Current Conditions

Compare fish samples from most recent years with most appropriate threshold of harmful effect or reference condition for **fish health**:

- **PCBs** – 2,400 ng/g lipid wt. (Meador, 2002)
- **PBDEs** – 1,600 ng/g lipid wt. Reference concentration (Arkoosh et al., 2010)
- **PAHs** –
  - PAH-induced liver disease in English sole; <5% normal, 5%-10% cause for concern, >10% highly impacted
  - PAH-metabolites in herring bile – threshold currently under development based on harmful effects threshold (Meador et al., 2008)
- **EDCs** - prevalence of vitellogenin (egg protein) in male English sole as an indicator of estrogen-mimicking chemicals. Detection is considered an indication of potential feminization.

OVERVIEW

The *Toxics in Fish* indicator is one of 21 vital signs of ecosystem health that help define expectations for recovered conditions in the Puget Sound by 2020. It condenses key information about exposure of Puget Sound biota to toxic contaminants. Currently *Toxics in Fish* summarizes **three major classes of pollutants**:

1. Persistent bioaccumulative toxics - polychlorinated biphenyls (**PCBs**) and brominated flame retardants (**PBDEs**), measured as tissue residues
2. Hydrocarbons from oil spills and combustion - polycyclic aromatic hydrocarbons (**PAHs**), measured as PAH-related liver disease or metabolites in fish bile
3. Endocrine disrupting compounds (**EDCs**) - measured as the prevalence of vitellogenin (egg protein) in male fish, resulting from exposure to environmental estrogen-mimicking pollutants.

	Current Condition					Time Trends			
	PCBs	BDEs	PAHs	EDCs		PCBs	BDEs	PAHs	EDCs
English Sole	red	yellow	yellow	red	**Elliott Bay**	red	yellow	green	grey
English Sole	red	yellow	red	white	**Duwamish River**	red	yellow	red	grey
English Sole	red	green	yellow	yellow	**Sinclair Inlet**	yellow	green	yellow	grey
English Sole	red	red	green	red	**Comm. Bay**	red	yellow	yellow	grey
English Sole	yellow	green	green	yellow	**Port Gardner**	red	yellow	green	grey
English Sole	yellow	green	green	white	**Eagle Harbor**	yellow	yellow	green	grey
English Sole	yellow	green	green	green	Nisqually	yellow	green	green	grey
English Sole	green	green	yellow	green	Hood Canal	green	green	yellow	grey
English Sole	green	green	green	white	Vendovi Island	green	green	green	grey
English Sole	green	green	green	white	Strait of Georgia	green	green	green	grey
	07, 09	07, 09	2011*	97-01	<< Years Evaluated				
Herring	red	green	yellow	white	**Squaxin**	yellow	green	yellow	white
Herring	red	yellow	yellow	white	**Pt Orchard**	yellow	green	yellow	white
Herring	green	green	yellow	white	Semiahmoo	green	green	green	white
Herring	yellow	yellow	yellow	white	Cherry Point	green	green	yellow	white
	08, 10	06-10	08, 10^		<< Years Evaluated				
Coho	green	green	white	white	Duwamish River	green	green	white	white
Coho	green	green	white	white	Deschutes River	green	grey	white	white
Coho	green	green	white	white	Nooksack River	green	grey	white	white
Coho	green	green	white	white	Nisqually River	green	grey	white	white
Coho	green	green	white	white	Skagit River	grey	grey	white	white
	06, 08	2008			<< Years Evaluated				
Chinook	red	green	white	white	Duwamish River	white	white	white	white
Chinook	red	green	white	white	Deschutes River	white	white	white	white
Chinook	red	green	white	white	Nisqually River	white	white	white	white
Chinook	red	green	white	white	SPS juveniles	white	white	white	white
	03, 04	03, 04			<< Years Evaluated				

red	target is not being achieved
yellow	intermediate or equivocal result
green	target is being achieved
grey	not enough data or inadequate threshold
white	not tracking

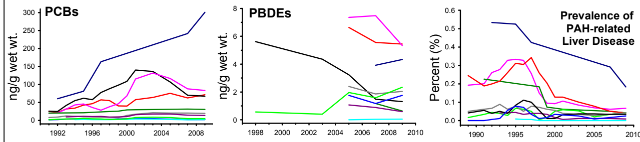
Bold text = urban/industrialized stations
* measured as PAH-related liver disease
^ measured as PAH-metabolites in fish bile

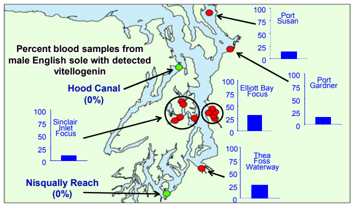
Recovery Targets for Time Trends

Using monitoring data (presented as 3-year moving averages):

- **Achieve a declining trend** in each applicable metric for English sole and herring in urban areas/basins
- **Allow no increasing trend** for these species in non-urban areas
- **Allow no increasing trend** in coho salmon (Chinook not currently tracked)

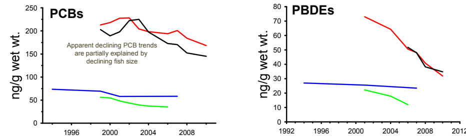
English sole (*Parophrys vetulus*) are bottom-dwelling flatfish that represent the sediment-to-biota contaminant link

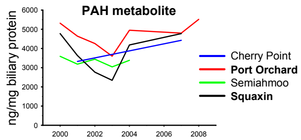




Toxics in Fish tracks PCBs and PBDEs in English sole fillets, as well as PAH-related liver disease. Between 1997-2001 we measured vitellogenin (an egg protein) in male fish, an indicator of exposure to endocrine disrupting compounds.

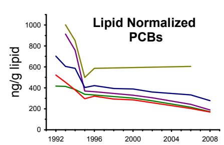
Pacific herring (*Clupea pallasi*) are small, mid-water planktivores that represent the pelagic foodweb

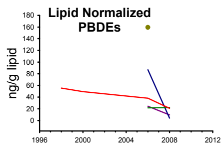




Toxics in Fish tracks PCBs and PBDEs in herring whole bodies as well as PAHs, measured as metabolites in fish bile.

Coho salmon (*Oncorhynchus kisutch*) are short-lived anadromous species that reflect a combination of oceanic and Puget Sound conditions





Throughout their lives salmon may be exposed to contaminants in freshwater, estuarine or marine areas. *Toxics in Fish* tracked PCBs and PBDEs in returning adult coho fillets until 2008.

CONCLUSIONS

English sole – English sole from four urban locations failed to meet recovery targets, or showed uncertain results, for current conditions for most of the PCBs, PBDEs, PAHs and EDCs. English sole from two urban locations (Port Gardner and Eagle Harbor), and for non-urban locations met recovery goals or exhibited intermediate results. English sole from most urban locations showed no declining trend in PCBs and PBDEs (failed target), while most non-urban locations showed no increasing trend (met target). PAHs appear to be declining in English sole from three, and possibly five, urban locations and were low and stable in non-urban locations.

Pacific Herring – PCBs in herring from urbanized basins were above effects thresholds and not changing (failing target). PCBs were below or near thresholds, and not increasing, in fish from non-urbanized basins (meeting target). PBDEs were below effects thresholds for most herring and trends appear to be declining in all basins. The effect of PAHs on herring is uncertain at this point, but conditions are not increasing or decreasing.

Coho and Chinook Salmon – Although currently not being tracked, best data to date indicated that adult coho from all sampled locations were below thresholds for PCBs and PBDEs and stable (meeting targets). Adult Chinook salmon from all locations, and juveniles from one basin, exceeded PCB thresholds, but not PBDE thresholds.

* Washington Department of Fish & Wildlife, ‡Northwest Fisheries Science Center, ^ Puget Sound Partnership

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Overview

Indicators of ecosystem health and related recovery targets have been selected by the Puget Sound Partnership in support of efforts to recover Puget Sound ecosystem health by 2020. This Dashboard of Indicators is meant to define expectations for recovered conditions, or a trajectory towards such conditions, for a wide range of ecosystem Vital Signs. They reflect an effort to simplify science reporting and link it to policy, ultimately resulting in easily communicated policy statements (i.e., targets) that define the desired condition or goals.

The *Toxics in Fish* indicator condenses key information regarding exposure of Puget Sound biota to toxic contaminants, harmful effects from such exposure, and time-trends in exposure-and-effects. This indicator relies on three species representing key food-web pathways: (1) a bottom-dwelling flatfish, English sole (*Parophrys vetulus*), (2) a small, schooling mid-water planktivore, Pacific herring (*Clupea pallasii*), and (3) Pacific salmon species, including coho salmon (*Oncorhynchus kisutch*) and Chinook salmon *O. tshawytscha*). English sole represent the sediment-to-biota contaminant link for contaminants that tend to accumulate in sediments. Pacific herring are prey to virtually every large piscivorous species in Puget Sound, and so generally represent the pathway of contaminants to higher level predators including Pacific salmon, seabirds, and killer whales. Coho and Chinook salmon are short-lived, highly migratory and anadromous species that reflect a combination of oceanic and Puget Sound conditions. They also represent a pathway of contaminants from fish to humans.

Although these three species are typically exposed to a wide range of toxic contaminants, the *Toxics in Fish* indicator focuses on three classes: (1) persistent bioaccumulative toxics such as polychlorinated biphenyls (PCBs) and polybrominated diphenylethers (PBDEs), (2) polycyclic aromatic hydrocarbons (PAHs), and (3) endocrine disrupting compounds (EDCs). Although there is crossover between some individual chemicals among these groups and this list is not comprehensive, organizing results by these contaminant groups helps to simplify communication.

Methods

Using long-term status and trends data collected by Washington Department of Fish and Wildlife's Puget Sound Assessment and Monitoring Program (PSAMP) we evaluated both current conditions and long-term time trends in these contaminants for these species by region (Figure 1).

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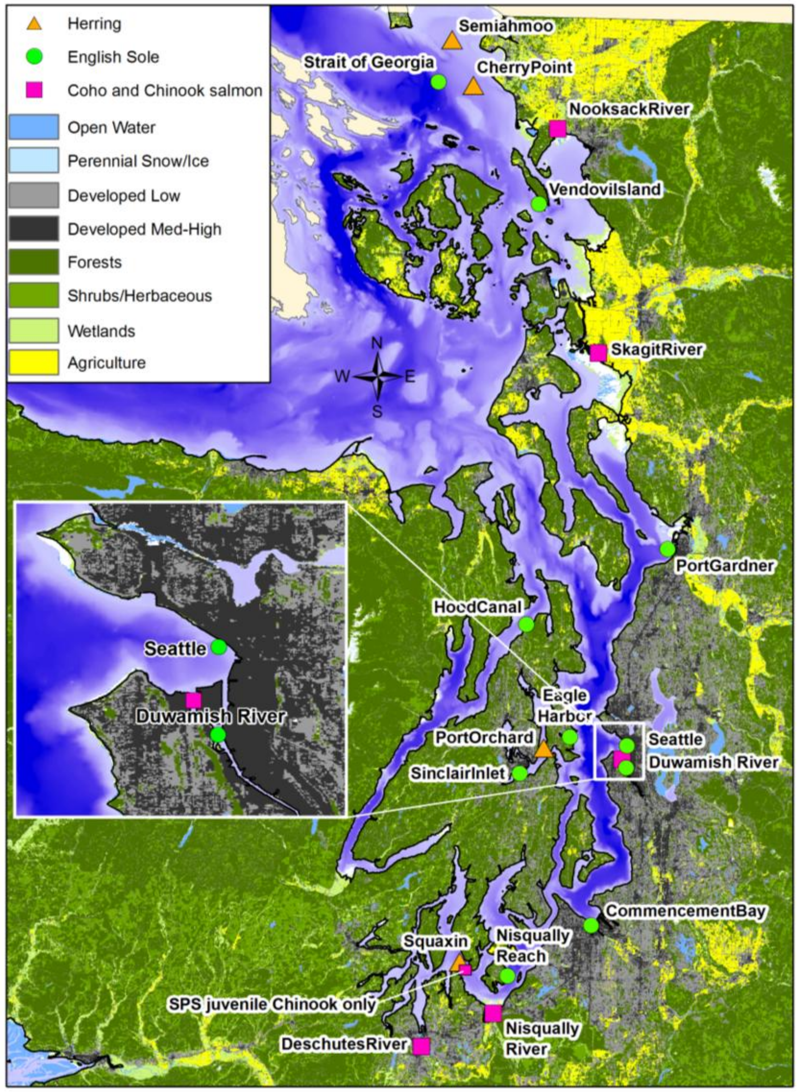

Figure 1. Location of Puget Sound Assessment and Monitoring Program's fish monitoring stations in Puget Sound and adjacent land-use types.

Current Conditions reflect the magnitude or severity of contaminant exposure (typically tissue concentration of a contaminant) or effects (typically some measure of disease) for each species across Puget Sound, using the most recent data. Ideally exposure and effects metrics are then compared to published exposure thresholds (level of contaminants in tissues) that may cause disease or other harmful effects. If appropriate thresholds do not exist, other targets may be selected based on best available science, until more suitable thresholds become available. Currently the *Toxics in Fish* recovery target for current conditions is that 95% of sampled fish exhibit exposure or effects metrics *below* such harmful-effects thresholds.

PCBs: We applied a harmful-effects threshold of 2,400 ng total PCBs/ g total lipids, for whole bodies of fish based on a review of PCB dose-response studies in juvenile salmon and trout (Meador, 2002). We compared whole body concentration of PCBs in herring and salmon against this threshold for current conditions. For English sole we estimated whole body concentration of samples from muscle (fillet) concentration and wet-weight to lipid-weight using conversion coefficients developed for this purpose. The conversion coefficient for whole-to-fillet was based on comparing measured PCB levels in English sole muscle tissue with whole body samples from Elliott Bay, a PCB-contaminated location. The conversion to lipid weight was made using the quotient of PCB concentration for wet-weight /lipid weight for each sample. These concentrations were compared to the Meador (2002) effects threshold. Each species-location or species-population cell was assigned one of three colors: green = target is being achieved (95% of samples below 2,400 ng total PCBs/g total lipids); yellow = equivocal conclusion resulting from uncertainty in lipid weight conversions; red = target is not being achieved (>95 % of samples exceeded the threshold, with little uncertainty).

PBDEs: No suitable harmful-effects thresholds exist for PBDEs in fish. Arkoosh et al., (2010) reported harmful effects (increased disease susceptibility) in juvenile salmon at whole body concentration of

1,400 ng total PBDEs/g total lipids, the lowest exposure concentration in their study. These authors are currently testing lower concentrations to help define the threshold of PBDE exposure above which effects are observed. Until such a suitable harmful effects threshold is developed, we applied the Arkoosh et al., (2010) concentration above as a reference concentration for our herring salmon and English sole, and assigned target colors using the same criteria as PCBs, comparing tissue PBDE concentration with the PBDE reference concentration.

PAHs: Two metrics were used for current conditions of PAHs: (1) measurement of PAH-metabolites in fish bile (herring), and (2) the prevalence of PAH-related liver disease in English sole. No measure of PAH exposure or effects has been monitored routinely for salmon. An effects threshold for PAH metabolites in fish bile is currently being developed, based on a study by Meador et al. (2008).

Liver disease (cancer) has been monitored in English sole from Puget Sound for the past 20 years as both an indicator of PAH exposure and fish health. The PAH-disease causal relationship was identified from extensive field and lab studies (Myers et al. 1990; Collier and Varanasi 1991; Myers et al. 1991; Johnson and Landahl 1993; Johnson and Landahl 1994; Myers et al. 1998). Using this work we assigned targets based on the following: <5% prevalence of liver disease is considered a normal background condition (target met, assigned green value); 5% to 10% indicates cause for concern (intermediate condition, assigned yellow value); > 10% is considered highly impacted (target not met, assigned red value).

EDCs: Endocrine disrupting compounds were measured in English sole using the frequency of occurrence of feminized male fish for each location. Feminization was defined by the presence of vitellogenin, an egg-yolk precursor that is normally only found in females of gonochoristic[4] species like English sole. Although the chemicals causing vitellogenesis in male English sole in Puget Sound are not known, they are presumed to be chemicals that mimic estrogen. Zero prevalence of vitellogenin in male English sole is considered the normal, or background condition (target achieved, assigned green value). We assigned <25%prevalence of vitellogenin a yellow value (cause for concern), and prevalence >25% a red value (target not being achieved). We note that the data for EDCs in Puget Sound were collected during a baseline survey from 1997 through 2001 (Johnson et al. 2008). English sole EDC samples were collected again in 2011 and currently await analysis.

Long-term time trends identify whether exposure or effects metrics are increasing or decreasing for each indicator species, for each pertinent location or population. A distinction in recovery targets is made between urbanized/developed areas and rural/undeveloped areas. The recovery target for fish populations in urbanized basins is to *achieve a declining trend* in exposure or effects metrics. The recovery target for fish in rural basins is *to achieve **no** increasing trend* in exposure or effects metrics. Species/location combinations meeting these targets were assigned green values. Statistically ambiguous results were assigned a yellow value.

Statistical significance of trends for PCB and PBDE concentrations in English sole, herring and coho salmon and for biliary PAH-metabolites in herring were evaluated using a general linear model (GLM) of ln-transformed contaminant data (wet weight) over time (years as the independent variable), with lipids and fish size as covariates. An increasing trend was concluded if the time coefficient exhibited a positive value, with a probability (α) <0.05, and no interaction term with covariates. A decreasing trend was concluded if the time coefficient was negative, and no trend was concluded if the year coefficient

[4] A condition where separate sexes normally exist, as opposed to species which may exhibit hermaphroditism or serially-changing sexes.

exhibited p >0.05. Cases were considered ambiguous when the model fit was poor, or it was difficult to separate covariate effects.

Trends in prevalence of liver disease in English sole were evaluated with a visual inspection of prevalence through time. Future analyses will use logistic regression to evaluate trend-slopes, including adjustment for variability in fish age.

No time trend data are currently available for the EDC metric. Initial status of EDCs in Puget Sound was conducted in 1997 through 2001 (Johnson et al. 2008). English sole EDC samples were collected again in 2011 and currently await analysis.

Results

English sole from four urban locations failed to meet recovery targets (or showed uncertain results) for current conditions for most of the PCBs, PBDEs, PAHs and EDCs (Figure 2). English sole from two urban locations (Port Gardner and Eagle Harbor), and for non-urban locations met recovery goals or exhibited intermediate results. English sole from most urban locations showed no declining trend in PCBs and PBDEs (failed target), while most non-urban locations showed no increasing trend (met target). PAHs appear to be declining in English sole from three (and possibly five) urban locations and were low and stable in non-urban locations.

Pacific Herring. PCBs in herring from urbanized basins were above effects thresholds and not changing[5], and below or near thresholds (and not increasing) in fish from non-urbanized basins (met target). Although a suitable PAH threshold is currently not available[6], PAH-metabolites in herring will likely be comparable to or above such a threshold in all basins. Because of this uncertainty they were assigned a yellow value. PAH conditions are not changing; apparent declines shown in graphs (exhibited on the companion poster) can be explained by declining fish size through time. PBDEs were below effects thresholds for most herring and trends appear to be either declining or remaining stable.

Coho and Chinook Salmon. Although currently not being tracked, the most recent data available indicated that adult coho from all sampled locations were below the threshold/reference value for PCBs and PBDEs and were either declining or stable (meeting targets). No distinction was made regarding the urban/rural character of any salmon samples based on their location of capture. Adult Chinook salmon from all locations (and juveniles from one basin) exceeded PCB thresholds, but not the PBDE effects concentration. Although not summarized here Sloan et al. (2010) observed PBDE concentrations above the reference value at several locations in Puget Sound,

[5] Equivocal results; apparent decline in PCBs can be explained by a decrease in fish length through time, hence yellow value assigned

[6] Evaluation of a PAH-metabolite threshold is currently underway. The harmful effect threshold will probably be in a range from 2,000 to 4,000 ng phenanthrene equivalent/mg biliary protein

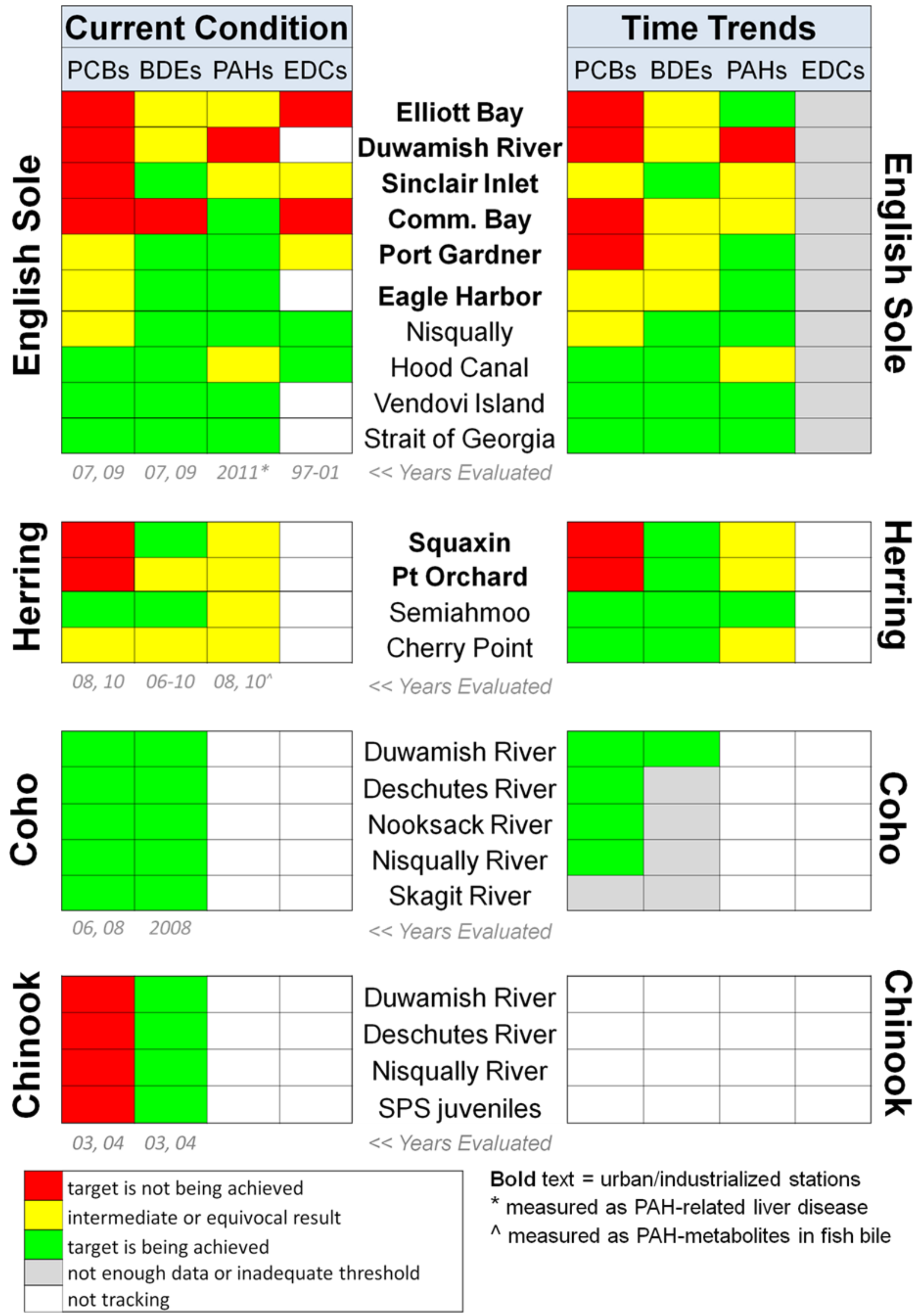

Figure 2. Summary of current conditions and long-term time trends in contaminants for four fish species in various regions of the greater Puget Sound, Washington.

Conclusions and Application

The range of fish species included in the *Toxics in Fish* dashboard indicator reflects the various marine habitats in which these species occur in the Puget Sound, their varied life-histories, and the wide ranges of tissue contaminant levels and degree to which these contaminants harm fish health.

In the bottomfish we observed high exposure to PCBs and endocrine disruptors in populations residing in Puget Sound's most urban embayments, including Elliott Bay and Commencement Bay. PCBs continue to present a problem in Puget Sound's urban bays almost 40 years after they were banned in the US, with concentrations exceeding harmful effects thresholds, and levels that are not declining. This underscores the urban-focal nature of PCB contamination in Puget Sound. The only urban area where English sole exhibited a decline in PCBs was Sinclair Inlet, following significant and widespread reductions in contaminant loads to the system (retrofits to stormwater discharges and combined-sewer-overflows), and dredging and capping of contaminated sediments in the early 2000s (O'Neill et al 2011). This recovery of English sole health suggests that local urban watersheds and sediment PCB reservoirs may continue to act as a source of contamination for local bottomfish, and that local upland source control and remediation of contaminated sediments can result in reduction of PCB exposure in these organisms. Except for equivocal results from one rural location (Nisqually), PCBs have remained low in English sole from non-urban areas over the past 20 years, suggesting that PCB loads are not increasing in sediments in those areas.

The status of endocrine disrupting compounds (EDCs) has not been well studied throughout Puget Sound however limited data summarized in *Toxics in Fish* suggest that EDC problems in English sole may not be limited to urban areas. From the distribution of these results and a review of other studies world-wide, Johnson et al. (2008) implicated stormwater and treated sewage effluent as potential sources of xenoestrogenic[7] EDCs in Puget Sound. These sources may or may not be associated with degree of land-development. Johnson et al. (2008) observed vitellogenin induction in male English sole from at least one rural location. More observations are needed to identify the potential population level effects of EDCs in Puget Sound fish, and whether conditions are getting better or worse.

PAH-related liver disease in English sole remains a problem in the Duwamish River, but has declined in Elliott Bay (wherein sediment PAHs have declined as well) and possibly Commencement Bay. Sediment remediation and source control at a former creosote treatment facility was shown to effectively recover fish health in Eagle Harbor (Myers et al. 2008). It is unknown whether similar, albeit limited efforts to mitigate sediment PAHs in Elliott Bay and Commencement Bay have resulted in declining liver disease, or whether source controls of PAHs have contributed to the decline in English sole liver disease.

Although PBDEs have exhibited an exponential increase in some marine mammals from the Salish Sea in recent years (Ross 2006), these chemicals have shown either a declining trend or no increasing trend in English sole, Pacific herring and coho salmon across Puget Sound. Further, the concentration of PBDEs in these species as well has Chinook salmon have remained relatively low (except for English sole in Commencement Bay). It is possible that an EPA-mediated voluntary cessation in production of common PBDE chemicals in 2004 played a role in preventing increases in Puget Sound.

Unlike bottomfish, PCBs in Pacific herring, a pelagic (open water) species, were high throughout Puget Sound, not just in urban embayments. This has been reported elsewhere (West et al. 2008) and reflects

[7] Estrogen or estrogen-like compounds occurring in the water, or outside the fish's body

the PCB-contaminated nature of Puget Sound's pelagic food web. O'Neill et al. (2006) observed high PCB concentrations in Chinook salmon that remained as residents in Puget Sound, compared to Chinook that had migrated to the Pacific Ocean, implicating residency in Puget Sound as the primary risk factor for PCB exposure. Because of their pelagic nature, it is unlikely that PCB contamination in herring and salmon results directly from contaminated sediments. Their high exposure to PCBs and lack of declining trend suggests an ongoing input to the pelagic food web in the water column, perhaps in plankton, at the lowest levels of the food web (see West et al. 2011).

Although coho and Chinook salmon were selected by the Puget Sound Partnership as key species for the *Toxics in Fish* indicator, salmon have not been monitored for contaminants recently. However, similar to herring, Chinook salmon exhibited high exposure to PCBs and low exposure to PBDEs. Of key importance in evaluating harmful effects thresholds in salmon and other high-lipid species is the degree to which variation in body fat affects PCB concentration in the body, when expressed on a lipid-weight basis[8]. Periods of high fat content result in low PCB concentration in the fish's body, and lower toxicity because PCBs are sequestered in fat. However when fat is burned (such as during reproduction or spawning runs), PCBs can be liberated from lipid storage and circulated to more sensitive tissues (including nervous, respiratory, reproductive and endocrine systems). Hence, PCB toxicity in species such as salmon or herring may fluctuate throughout a fish's life, especially during periods of growth, migration, or spawning). In addition, mobilization of lipid-sequestered PCBs during reproduction can result in maternal transfer of these contaminants to their offspring, via deposition into eggs.

In summary, the fish species monitored in the *Toxics in Fish* Dashboard Indictor allow us to track a wide range of toxic chemicals across Puget Sound's basins, and across important ecosystem compartments. Although not comprehensive, *Toxics in Fish* tells us where harm to fish is occurring in Puget Sound, where conditions are improving or getting worse, and how effective our pollution management strategies may be. *Toxics in Fish* will be updated regularly, and be continually improved as the science that supports it progresses.

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[8] Most harmful effects thresholds are presented on a lipid-weight, rather than wet-weight basis.

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