Environmental Science & Technology

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Projecting Fish Mercury Levels in the Province of Ontario, Canada and the Implications for Fish and Human Health

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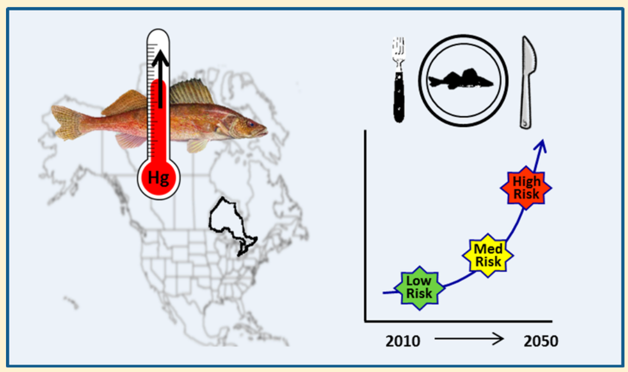
Supporting Information

ABSTRACT: Fish mercury levels appear to be increasing in Ontario, Canada, which covers a wide geographical area and contains about 250 000 lakes including a share of the North American Great Lakes. Here we project 2050 mercury levels in Ontario fish, using the recently measured levels and rates of changes observed during the last 15 years, and present potential implications for fish and human health. Percentage of northern Ontario waterbodies where sublethal effects of mercury on fish can occur may increase by 2050 from 60% to >98% for Walleye (WE), 44% to 59–70% for Northern Pike (NP), and 70% to 76–92% for Lake Trout (LT). Ontario waterbodies with *unrestricted* fish consumption advisories for the *general population* may deteriorate from 24–76% to <1–33% for WE, 40–95% to 1–93% for NP, and 39–89% to 18–86% for LT. Similarly, Ontario waterbodies with *do not eat* advisories for the *sensitive population* may increase from 32–84% to 73–100% for WE, 9–72% to 12–100% for NP, and 19–71% to 24–89% for LT. Risk to health of Ontario fish and humans consuming these fish may increase substantially over the next few decades if the increasing mercury trend continues and updated advisories based on continued monitoring are not issued/followed.



■ INTRODUCTION

Mercury, specifically methylmercury, has been recognized as a global pollutant due to its widespread presence, and bioaccumulative and toxic nature.[1] Mercury is an endocrine disrupter and can damage gonads and alter production of sex hormones in freshwater fish.[2−5] Toxic effects of mercury to humans include damages to the neurological, immune, genetic, enzyme, cardiovascular, respiratory, and gastrointestinal systems.[6−8] Although mercury can be naturally elevated in the environment, anthropogenic activities can increase environmental mercury levels even at remote locations.[9−11] As a result, mercury concentrations in various body parts of a number of animals have increased by more than 5-fold during the industrialization period.[9]

Global atmospheric mercury emissions likely peaked during the 1950s-1970s and then declined due to reductions in North America, Europe, and Russia.[9] Mercury emissions have declined by 90% in Canada between the 1970s and 2011,[12,13] and by 75% in the U.S. between 1990 and 2008.[14−16] In the Province of Ontario (especially northern Ontario), Canada, fish mercury levels declined rapidly during the 1970s and 1980s, which was likely in response to the reductions in atmospheric emissions in North America and worldwide.[17] However, the fish levels have increased somewhat between 1995 and 2012.[17] These increases could be a result of various complicating factors affecting mercury levels in fish, including recently increasing atmospheric mercury emissions from East Asia offsetting continuing reductions in North America, global climate change, and food web alterations due to invasive species.[17]

A number of recent studies have reported either flat or increasing fish mercury levels;[17−24] however, to the best of our knowledge, mercury levels in fish have not been projected on a large scale and potential implications for fish and human health have not been assessed. A number of studies have projected mercury emissions, atmospheric levels, deposition and oceanic levels,[25−28] but such studies are lacking for fish mercury content. A major reason behind this could be challenges in projecting future concentrations by mathematically modeling the widely varying, complex, and, in many cases, interconnected processes that influence fish mercury levels. Utilization of measured rates of recent fish mercury changes for projecting future mercury levels may be a more reliable approach at present.

Using the latest fish mercury measurements (2000–2012) and rates of change for Ontario, Canada observed during the

Received: January 23, 2015
Revised: November 14, 2015
Accepted: November 23, 2015
Published: November 23, 2015

DOI: 10.1021/acs.est.5b03943
Environ. Sci. Technol. 2015, 49, 14494−14502

last 15−17 years (1995−2012),[17] here we project mercury levels in Ontario fish and present potential implications for fish and human health in the context of fish consumption advisories if the increasing mercury trends continue. The rates of changes in fish mercury levels utilized in this study for the projection purpose were derived from an extensive database of >200 000 consistent fish mercury measurements collected by Ontario Ministry of the Environment and Climate Change (OMOECC), Ontario, Canada over the last 40 years (1970s to 2012). Since Ontario contains >250 000 lakes (including Canadian waters of the Great Lakes) and covers a large geographical area (approximately 3 and 4 times larger than Germany and U.K., respectively; spans approximately from 41.5° to 56.5° N and 73° to 95° W), the results presented here may reflect impact, to certain extent, on a large scale.

MATERIALS AND METHODS

Sample and Data Collection. The OMOECC monitors mercury in a variety of sport and forage fish in Ontario, Canada since the 1970s in partnership with Ontario Ministry of Natural Resources Forestry (OMNRF) and various other agencies/institutes. Fish samples are collected using diverse methods such as gill netting, trap netting, electrofishing, and angling. Length, weight and in most cases sex were recorded and a skinless, boneless dorsal fillet was removed for mercury analysis. Skinless boneless dorsal fillet is of prime interest for mercury measurement due to its relevance for human fish consumption. The samples were homogenized and kept frozen at −20 °C until mercury analysis using acid digestion and cold vapor flameless atomic absorption spectroscopy (CV-FAAS) as described by Bhavsar et al.[18]

Selection of Species. For this study, we selected three top predatory fish species namely Walleye (*Sander vitreus*, WE), Northern Pike (*Esox lucius*, NP), and Lake Trout (*Salvelinus namaycush*, LT) due to the following four major reasons. First, biomagnification of mercury in aquatic food webs results in about a million time higher concentrations in top predator fish compared to the surrounding water levels.[29] As such, top predator species are susceptible to higher exposure to mercury, thus making them good biological indicator species for this study. Second, such species are popular among anglers[30] translating into a greater mercury exposure and thereby health risk for humans. Third, recent rates of change in mercury levels are readily available for these three species.[17] Finally, these species are widespread in Ontario, Canada[31] providing a greater spatial coverage.

Data Screening. For an assessment of current mercury levels, measurements collected between 2000 and 2012 were considered to maximize available species/locations and minimize the influence of historical measurements. Since samples could have been collected from different locations in a river/creek over time, such locations were not considered. Further, measurements collected for Canadian waters of the Great Lakes and easily identifiable locations impacted by point source(s) were excluded because they may be experiencing temporal trends that are not representative of large scale changes to inland lakes. The screened data set included 26 036 measurements from a total 938 distinct locations (12 477 WE measurements from 627 locations, 7578 NP measurements from 609 locations, and 5981 LT measurements from 385 locations).

Data Analysis. Since mercury levels in fish considered here increases with fish size,[32] three standard lengths (std-lengths) representing small, medium and large sizes of fish were selected (WE − 40, 50, and 60 cm; NP − 45, 60, and 70 cm; and LT − 45, 60, and 70 cm) based on recent literature[24] and measurements available in the data set to consider the effect of fish length on mercury concentrations. We selected the power function to describe fish length and mercury relationship as it generally performs better for the species considered in this study.[32] In order to calculate mercury concentrations at standardized lengths, first 1581 (618 WE, 590 NP, 373 LT) power series regressions were constructed using the equation $Y = aX^b$ (where Y is fillet mercury concentration in μg/g wet weight, X is fish length in centimeters, and a,b are regression coefficients) for each sampling event (i.e., for every combination of species, location, and year). The power series regressions were conducted by fitting linear regressions on logarithmically transformed values (i.e., $\log Y = \log a + b \log X$).

All location-specific 2000−2012 data were pooled. Only those locations with a minimum of five measurements (Supporting Information Figure S1a,b) and the 10 cm size range (i.e., difference between maximum and minimum fish length; Figure S1c,d) were further considered. In addition, only those locations with the smallest fish smaller than a standard length plus 15 cm (e.g., Figure S1e) and the largest fish larger than a standard length minus 15 cm (e.g., Figure S1g) were considered to avoid large extrapolation of the power series regressions while calculating each standardized length fish mercury concentration. For example, to calculate 50 cm WE mercury for a particular location, the smallest WE measured for that location should be smaller than 65 cm and the largest fish should be larger than 35 cm (Figure S1e−h). Lastly, because mercury concentrations generally increase with fish length (a surrogate for fish age) due to bioaccumulation and biomagnification,[32] only positive relationships between location/species-specific mercury concentrations and fish length were considered. This is because negative relationships could be an artifact of different size classes collected during different sampling events at a location where mercury levels might have changed over time.

After the data screening, 4321 std-length/species/location specific mercury concentrations were calculated (small, medium,

Table 1. Rates of Recent Mercury Change (μg/g Decade ww) in Northern and Southern Ontario Fish under the Fixed Rate of Change and Annual Percent Change (APC) Approaches.[17]

		std-length (cm)	fixed rate change in mercury (μg/g decade ww)	annual percent change (APC) in mercury (%)
Northern Ontario	Walleye	40	0.09	2.19
		50	0.12	1.72
		60	0.16	1.42
	Northern Pike	45	0.01	0.39
		60	0.09	1.96
		70	0.19	2.36
	Lake Trout	45	0.01	1.13
		60	0.02	1.41
		70	0.02	1.29
Southern Ontario	Walleye	40	0.03	−0.14
		50	0.05	0.4
		60	0.07	1.16
	Northern Pike	45	0.02	0.72
		60	0.04	0.87
		70	0.07	0.11
	Lake Trout	45	0.03	1.51
		60	0.05	1.51
		70	0.07	−1.23

DOI: 10.1021/acs.est.5b03943
Environ. Sci. Technol. 2015, 49, 14494−14502

large: WE − 574, 576, 576; NP − 521, 532, 532; LT − 338, 336, 336; respectively). Mercury concentrations for the current (2010) scenario (set as an initial condition, based on the 2000−2012 data; Figure S2a) were used to project future concentrations for 2020, 2030, 2040, and 2050 (Figure S2b), using the rates of mercury changes between 1995 and 2012 that we estimated in a recent study based on the same data set (Table 1).[17] Since northern and southern Ontario locations showed differing fish mercury trends,[17] we present results for the two regions separately.

Two approaches for addressing rates of changes in fish mercury levels were considered. First, fixed rates of mercury changes, which do not depend on initial fish mercury levels, were utilized. Since methylmercury is the predominant form of mercury in fish and forms adducts with S-bearing amino acids that do not behave as methylmercury, it is reasonable to assume that mercury accumulation in fish mainly relies on supply of methylmercury rather than concentration difference between fish body and dietary content in the fish gut.[33] This fixed rate analysis was based on average rates of mercury change in fish for all sampled locations in a region. For LT from southern Ontario, median (instead of average) rates were used as lower number of locations had resulted in unreasonably high average rates.[17]

Table 2. Projected Changes in Percent of Sampled Locations with Fish Mercury Concentrations above the Toxicity Reference Values at Their Maturity Lengths

		2010	2050
Northern Ontario	Walleye	60%	> 98%
	Pike	44%	59−70%
	Lake Trout	70%	76−92%
Southern Ontario	Walleye	43%	44−67%
	Pike	25%	47−52%
	Lake Trout	57%	79−89%

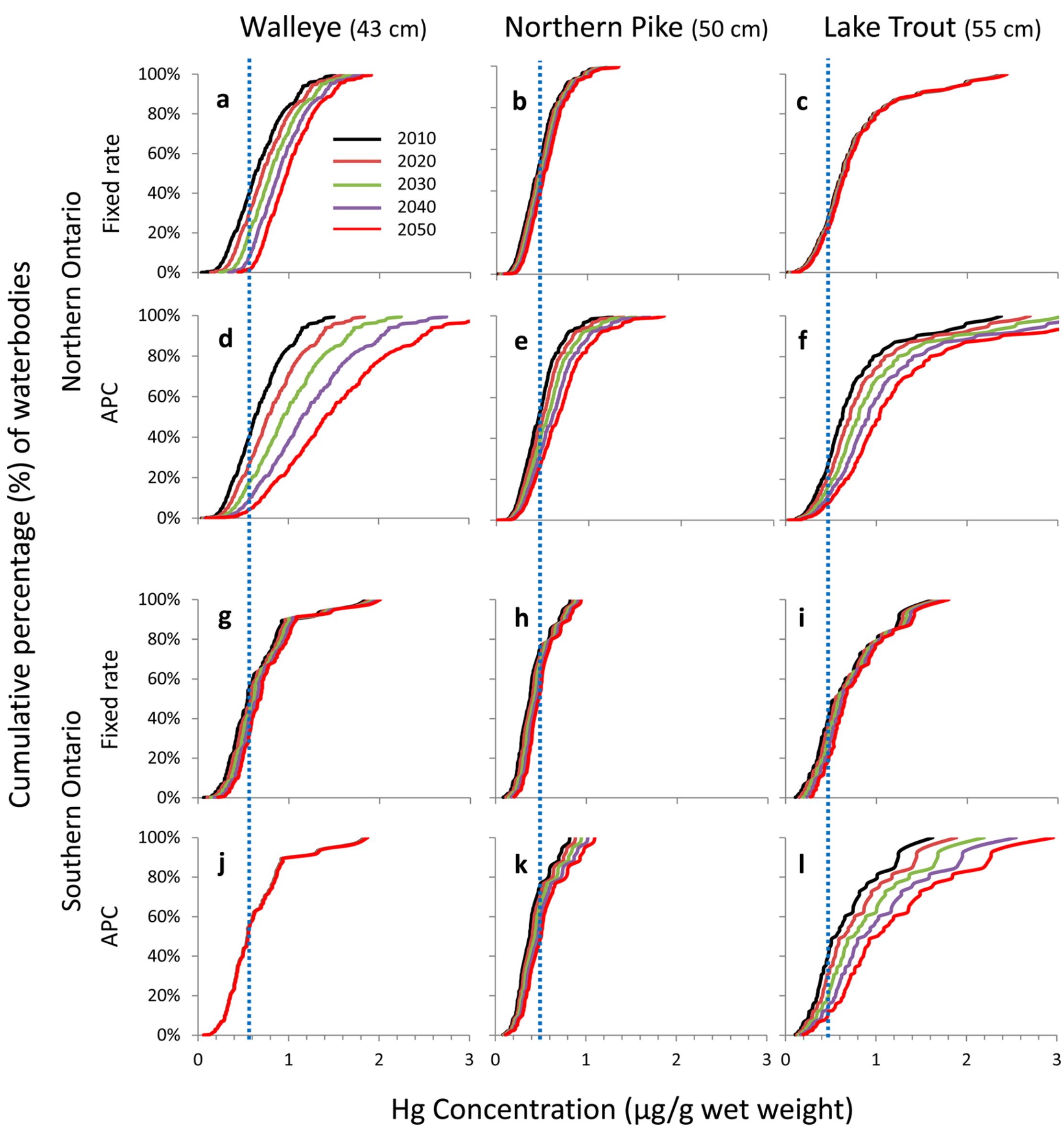


Figure 1. Projected changes with time (2010−2050) in cumulative percentage (%) of northern and southern Ontario waterbodies that are below certain mercury concentrations (µg/g wet weight skin-off fillets) for Walleye (43 cm), Northern Pike (50 cm) and Lake Trout (55 cm) under the fixed rate and annual percent change (APC) approaches. The dotted line represents mercury concentration at which sublethal effects, including changes in reproductive health, have been consistently observed in laboratory and field studies on freshwater fish.[34]

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Environ. Sci. Technol. 2015, 49, 14494−14502

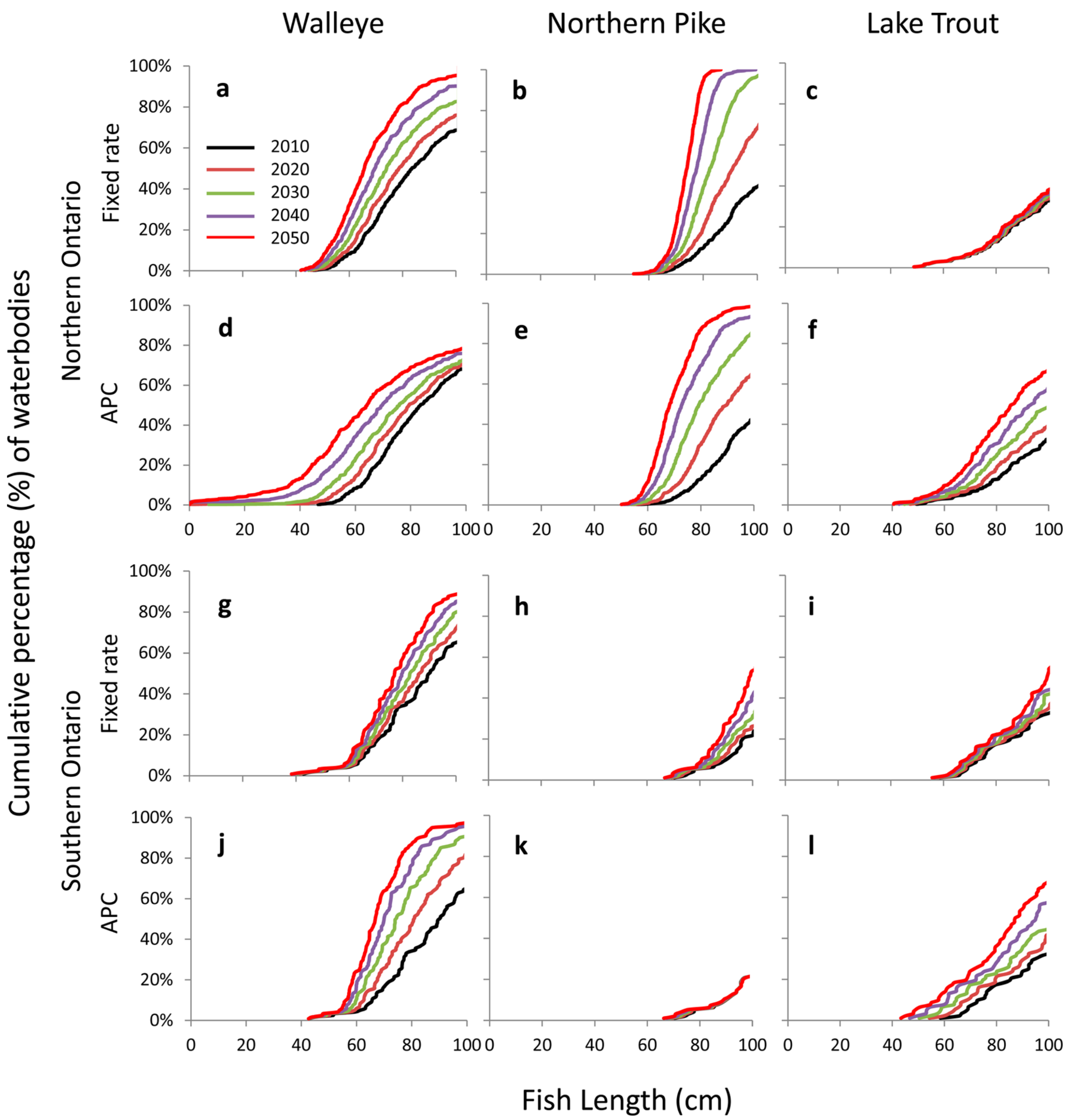

Figure 2. Projected changes with time in cumulative percentage (%) of northern and southern Ontario waterbodies that are exceeding skin-off fillet mercury concentrations of 1.84 μg/g (wet weight) at varying lengths of Walleye, Northern Pike and Lake Trout under the fixed rate and annual percent change (APC) approaches. The mercury concentration of 1.84 μg/g is used as "*do not eat*" advisory benchmark by the OMOECC for the general population.

Second approach was based on annual percent change (APC). The current low or high fish mercury levels reflect the abundance of mercury and its dynamics in the ambient environment, which may not be equally influenced by the factors that are contributing to the recent increases in fish mercury levels in this region. As such, it is plausible that the rates of mercury increase would be dependent on the current fish mercury levels, and hence another set of calculations were conducted using the fish species- and size-specific APC values (Table 1).[17] We believe that the use of the two different approaches provide a comprehensive view of the projected potential impacts.

Risk to Fish. Chemical risk to fish health can be assessed in different ways using toxicity reference value (TRV) for certain type of effects such as reduced growth, disrupted reproduction, and loss of life.[34,35] In this assessment of potential impact on fish, we considered TRV of 0.3 μg/g wholebody for sublethal effects of mercury on freshwater fish including altered reproductive health.[34] We believe this is relevant because an impact on reproduction of fish could translate into a risk at the population level.[34] This wholebody-based TRV was converted to equivalent fillet concentrations of 0.57 μg/g for WE, 0.49 μg/g for NP and 0.47 μg/g for LT.[34,36] Length of female fish at first reproduction (maturity) was selected to standardize observed mercury concentrations for the risk assessment. Selected fish lengths at maturity (mat-lengths) were 43 cm for WE, 50 cm for NP and 55 cm for LT.[34,37] Results are presented in terms of cumulative percentages of waterbodies exceeding fillet equivalent mercury TRV at the mat-lengths. Sandheinrich et al.[34] provided a detailed explanation on appropriateness of the approach for this type of risk assessment.

Risk to Human Consumers of Fish. Fish consumption advisories have been used by various agencies in North America to protect human health.[29,38] These advisories are generally based on a risk assessment approach that considers various parameters, such as tolerable daily intakes and exposure rates.[38,39] As such, a breakdown of the fish consumption advisories can

DOI: 10.1021/acs.est.5b03943
Environ. Sci. Technol. 2015, 49, 14494−14502

Table 3. Projected Changes in Percent of Sampled Locations Resulting in a Particular Type of Fish Consumption Advisory for the General and Sensitive Populations[a]

			8+ meals/month		"do not eat"	
			2010	2050	2010	2050
overall Ontario	Walleye	General	24–76	< 1–33	0–7	< 1–38
		Sensitive	3-21	0–8	32–84	73–100
	Pike	General	40–95	1–93	0–3	0–43
		Sensitive	3–48	0–35	9–72	12–100
	Lake Trout	General	39–89	18–86	0–6	0–21
		Sensitive	8–41	2–29	19–71	24–89
Northern Ontario	Walleye	General	23–75	0–20	0–9	0–41
		Sensitive	2–21	0–2	34–85	88–100
	Pike	General	37–95	0–94	0–3	0–52
		Sensitive	3–46	0–38	10–72	11–100
	Lake Trout	General	35–89	2–87	0–6	0–52
		Sensitive	8–39	3–31	21–72	24–88
Southern Ontario	Walleye	General	26–82	3–84	0–4	0–23
		Sensitive	3–23	0–28	22–82	21–99
	Pike	General	53–96	6–91	0–0	0–3
		Sensitive	4–55	0–31	9–69	13–98
	Lake Trout	General	47–89	15–84	0–7	0–24
		Sensitive	8–47	0–24	13–66	25–95

[a]A more detailed summary is provided in Table S2.

provide indication of severity of risk to human consumers if such advisories are not issued and followed. Fish consumption advisory benchmarks for mercury used by OMOECC (Table S1)[38] were utilized to classify observed and projected mercury levels in the small, medium and large sized fishes into various advisory categories.

Risk Projection. Mercury exposure risk to fish and humans were projected by calculating future concentrations at fish mat-lengths and fish lengths at which mercury concentrations would exceed *do not eat* advisory benchmarks, respectively. This was achieved by first estimating future concentrations for the small, medium and large sized fish using the 2010 levels and rates of mercury change shown in Table 1, and then conducting year-specific power series regressions (Figure S2). This procedure was conducted for each species (WE, NP, LT), location, 2010 and future year (2020, 2030, 2040, 2050), and rate scenario (fixed, APC) totalling 14 280 power series regressions. Only those locations that met the data screening criteria to provide all three std-length concentrations were considered for the regressions. These regressions were then used to project future concentrations at the mat-lengths and to estimate fish lengths at which mercury concentrations would exceed 1.84 and 0.52 μg/g, which were the benchmarks used by OMOECC to issue *do not eat* advisories for the general population and sensitive population of children and women of child-bearing age, respectively. Figure S3a illustrates a location/scenario-specific projection where mercury in WE at the maturity length of 43 cm would exceed fillet equivalent TRV of 0.57 μg/g by 2030. Figure S3b illustrates a location/scenario-specific projection, where WE length exceeding 1.84 μg/g would deteriorate from 69 cm in 2010 to 49 cm in 2050. Increase (or decrease) in fish lengths to reach mercury levels at *do not eat* benchmarks would implicitly indicate decrease (or increase) in risk to human consumers, if they would not follow the fish consumption advisories.

RESULTS

Risk to Fish. For present conditions in northern Ontario, majority of the sampled locations are potentially at risk from mercury toxicity (WE at 60%, NP at 44% and LT at 70%; Table 2, Figure 1a–f). Corresponding values for southern Ontario are lower (WE at 43%, NP at 25% and LT at 57%; Table 2, Figure 1g-l). Percentage of the sampled northern Ontario locations where fish will potentially be at risk of sublethal effects in 2050 is estimated to increase (from 60% to >98% for WE, 44% to 59–70% for NP and 70% to 76–92% for LT; Table 2, Figure 1a-f). For the southern Ontario locations, estimated increases are relatively modest (from 43% to 44–67% for WE and 25% to 47–52% for NP, and greater from 57% to 79–89% for LT; Table 2, Figure 1g–l).

Risk to Human Fish Consumers. Percentage of sampled northern Ontario locations where an 80 cm fish can potentially exceed the *do not eat* mercury advisory benchmark of 1.84 μg/g for the general population in 2050 is estimated to increase substantially (from current 44% to 69–81% for WE, 13% to 87–94% for NP, and 13% to 15–40% for LT; Figure 2a–f). In comparison, the corresponding increases for southern Ontario are estimated to be modest (from current 34% to 57–87% or WE and 6% to 6–9% for NP, and 18% to 22–37% for LT; Figure 2g–l). Increases in percentage of sampled locations exceeding the *do not eat* mercury advisory benchmark of 0.52 μg/g for the

DOI: 10.1021/acs.est.5b03943
Environ. Sci. Technol. 2015, 49, 14494–14502

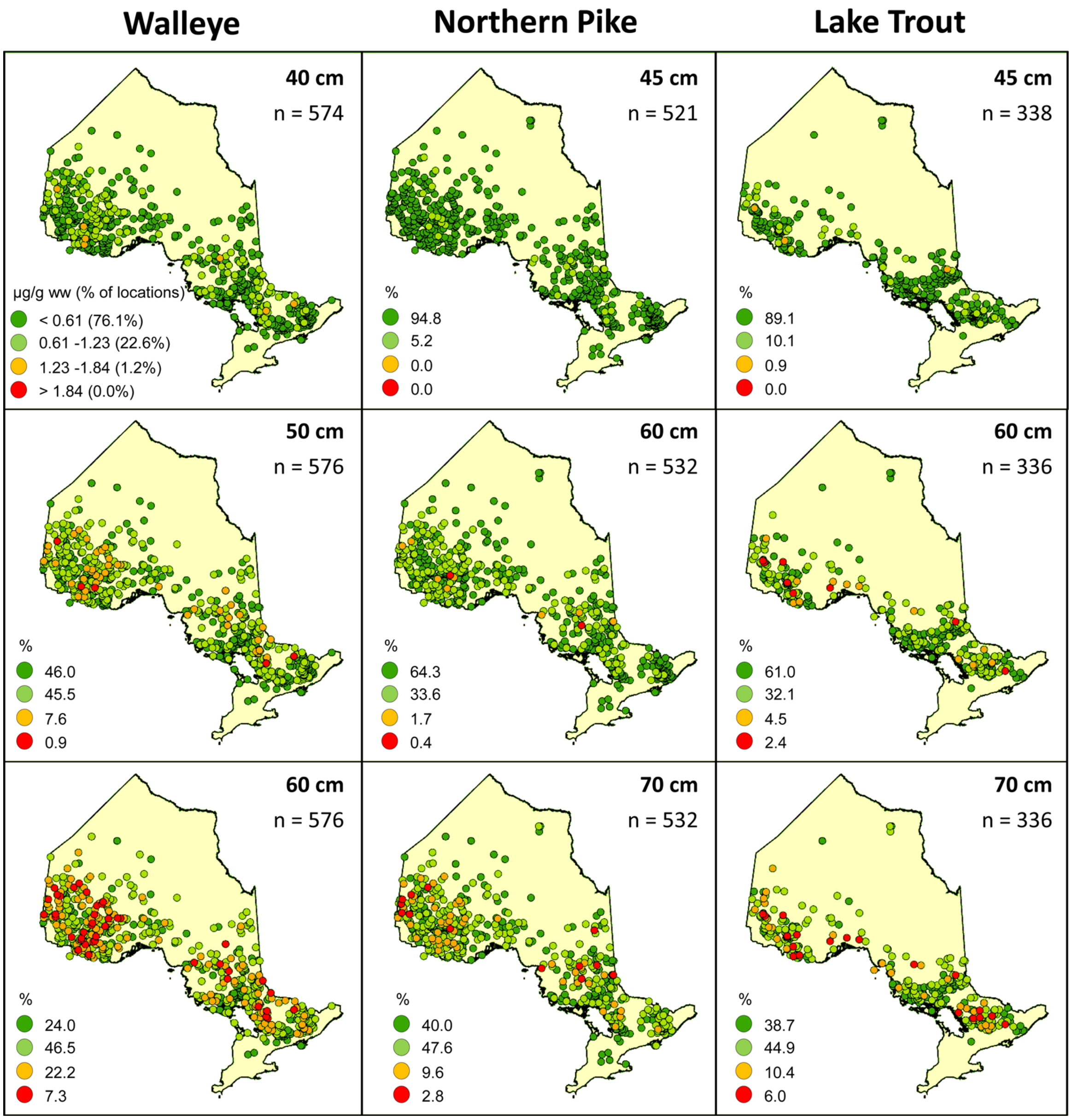

Figure 3. Mercury concentrations (μg/g wet weight) in skin-off fillets of small, medium and large sized Ontario Walleye, Northern Pike, and Lake Trout collected between 2000 and 2012. The concentrations have been grouped into the various categories used by the OMOECC for the purpose of fish consumption advisories geared toward the general population.[38] *n* represents number of locations.

sensitive population by 2050 are estimated to be sharper, especially for WE and NP in northern Ontario (Figure S4; northern Ontario: from 35% to 85–96% of WE (40 cm), 52% to 92–98% of NP (60 cm), and 27% to 34–61% of LT (50 cm); southern Ontario: from 22% to 21–44% for WE, 33% to 57–75% for NP, and 22% to 41–60% for LT).

A summary of the projected changes in percent of the sampled locations resulting in a particular type of fish consumption advisory is presented in Table 3 and Table S2. At present, 76%, 46%, and 24% of the sampled small, medium and, large sized WE Ontario locations have mercury concentrations in the *unrestricted* consumption advisory category for the general population (<0.61 μg/g), respectively (Figure 3). By 2050, proportion of such locations are estimated to deteriorate to 28–33%, 6–16%, and <1–6%, respectively (Figure 4 and Figure S5). For NP and LT, proportion of the sampled locations with such fish mercury concentrations (<0.61 μg/g) are better than WE and deteriorations are also estimated to be less (for NP from 95% to 90–93% (small), 64% to 16–17% (medium), and 40% to 1–10% (large); for LT from 89% to 63–86% (small), 61% to 25–47% (medium), and 39% to 18–24% (large); Figures 3, 4 and Figure S5).

Percentage of WE locations exceeding the *do not eat* consumption advisory benchmark of 1.84 μg/g for the general population is estimated to modestly increase by 2050 (from current 0% to <1–9% for small, < 1% to 6–19% for medium, 7% to 27–38% for large sized WE; Figures 3, 4 and Figure S5). The corresponding percentages for NP are expected to remain at 0% for small, and increase from 0% to 1–13% for medium, and 3% to 19–43% for large size (Figures 3, 4 and Figure S5). For LT, increases are estimated to remain at 0% for small, and increase from 2% to 2–11% for medium, and 6% to 8–21% for large size (Figures 3, 4 and Figure S5).

For the sensitive population, proportion of the sampled locations with fish mercury concentrations exceeding the *do not eat*

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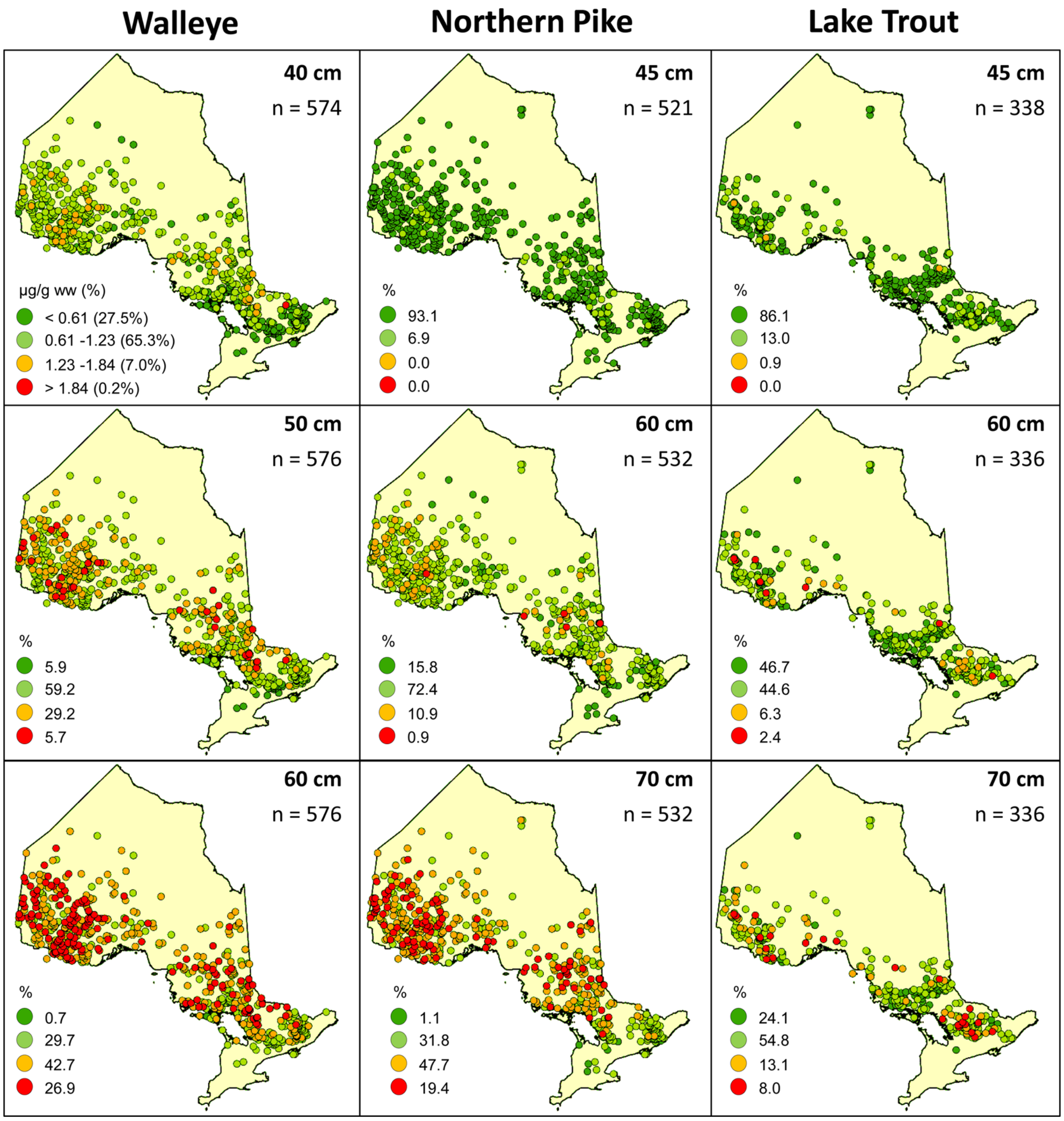

Figure 4. Projected 2050 mercury concentrations (μg/g wet weight) in skin-off fillets of small, medium, and large sized Ontario Walleye, Northern Pike, and Lake Trout under the fixed rate approach. The concentrations have been grouped into the various categories used by the OMOECC for the purpose of fish consumption advisories geared toward the general population.[38] *n* represents number of locations. Similar projections for the APC approach and sensitive population are presented in Figures S5 and S6.

advisory benchmark of 0.52 μg/g by 2050 may increase from 32−84% to 73−100% for WE, 9−72% to 12−100% for NP, and 19−71% to 24−89% for LT (Figure S6a−c). These results suggest that the health risk for the sensitive subpopulation of children and newborns (via maternal transfer) from consuming wild Ontario WE and NP may be high in 30−40 years if the increasing mercury trend continues and updated fish consumption advisories based on continued future monitoring data are not issued and followed. Detailed projections of breakdown of mercury concentrations classified into various advisory categories for the general and sensitive populations for northern and southern Ontario under various scenarios are provided in Figures S7−S12.

■ DISCUSSION

Elevated mercury levels, enhanced by anthropogenic activities mainly since industrialization during the 1800s and early 1900s, have been a concern worldwide.[10] A number of actions taken to curtail mercury emissions produced tangible results in the last half of the 20th century in various parts of the world, especially in North America.[10] However, the magnitude of the declines in North America may not be the same in future. Global emissions have increased during the last few decades largely due to increases from Asian countries, particularly China and India.[10] Although declines in environmental mercury levels have been observed in many cases,[1] various studies have also reported either flat or increasing trends for a variety of environmental media, including fish, in many parts of the world.[10,17,18,20,21,24,26,27,40,41] As discussed in detail by Gandhi et al.,[17] the recent mercury increases observed in Ontario fish are possibly a result of a variety of factors such as continued natural emissions, increased global mercury emissions during the last few decades, increasing trans-boundary flows of mercury (>95% of mercury deposition in

DOI: 10.1021/acs.est.5b03943
Environ. Sci. Technol. 2015, 49, 14494−14502

Canada), and warming weather under climate change. These and other factors may sustain the increasing fish mercury trend in future. The present study highlights potentially serious implications for the health of fish and fish-consuming humans if increases in the fish mercury levels continue.

There are differences among fish species and size classes in the rates of mercury change likely as a result of disparities in their diet matrix and preferred habitats (i.e., cold water LT, cool water WE, and littoral NP). Such differences may result in differential risk for fish populations and human consumers. For example, the maturity lengths of 43 cm for WE and 50 cm for NP are closer to the small sizes considered in this study (40 cm for WE and 45 cm for NP). The rates of mercury change for small sizes of WE and NP are closer to the lower ends of 0.09−0.16 and 0.01−0.19 μg/g/decade, respectively (Table 1). Lower mercury increases at the maturity length and lower current levels (Figure 3) are expected to result in lower impact of increased mercury in the NP than WE population (Figure 1). However, combined effect of currently lower mercury levels and greater increases in large NP than large WE (Figure 3, Table 1) may result in similarly increased risk over time for humans consuming these fish of large sizes (Figures 3, 4 and Figure S5). In contrast, under the APC approach, increases are greater in smaller WE and vice versa for NP (Table 1). If this scenario is true, there will be a greater mercury toxicity risk to the WE populations than inferred from the fixed rate scenario (Figure 1).

International efforts are being invested to reduce risk from mercury. United Nations Environment Programme (UNEP) is actively working on the mercury issue since 2003.[42] These efforts, including four years of negotiations with nations, have recently resulted in the Minamata Convention on Mercury (2013), a global legal treaty to prevent emissions.[43] However, these efforts will generally affect global mercury levels via emission reductions and thereby atmospheric levels and depositions. A modeling study has recently shown that even in the best-case scenario mercury deposition in 2050 may be similar to the present day.[25] Even if direct atmospheric inputs to a freshwater system declines and, in response, rapid improvements in fish mercury levels within a time frame of less than a decade can be expected, a full recovery may take much longer (at the scale of decades to centuries) due to slower watershed responses and storage of historic mercury in sediments.[10]

In addition to global atmospheric occurrence, a number of other elements, such as nearby anthropogenic activities (e.g., mining, coal fire power plant, reservoir impoundment, logging), chemistry related changes (e.g., sulfate input, dissolved oxygen level, water temperature), and other external factors (e.g., input from watershed, climate change, invasive species, re-emission) directly/indirectly affect mercury in fish[10] and may have contributed to recent increases of mercury in Ontario fish.[17] Since all these factors contribute to fish mercury in a complex way with unclear relative importance, it would be challenging to control the dominating processes and reverse the increasing trend in a short time frame.

The Province of Ontario, Canada, created the Green Energy Act in 2009, and in 2014 became the first jurisdiction in North America to fully eliminate coal as a source of electricity generation,[44] which has aided in reducing atmospheric mercury emissions. We can hope that this type of continued effort in addition to substantial (75−90%) reductions in North American emissions from 1970 to 2011, as a result of past actions, will restore a decreasing trend for fish mercury levels in foreseeable future. However, pressures such as hydropower expansion, new resource development in the Far North of Ontario, climate change, invasive species, and global emissions,[28,45−48] among possibly other factors, may lead to increases in mercury levels in the ambient environment of Ontario, especially in the northern region. We can also hope that international efforts on reducing mercury risk, increasing knowledge on behavior of mercury in environment, and actions on, for example, climate change and spread of invasive species will reduce fish mercury levels; however, it would be prudent to prepare for potential implications if increasing trends continue. It is recommended that monitoring activities are enhanced to confirm the status of mercury trends and to generate data for management actions, such as issuing comprehensive fish consumption advisories to protect human health.

In summary, this study projected fish mercury levels in the Province of Ontario, Canada and the implications for fish populations and health of human consumers of wild fish. The results showed a possibility of substantial increases in the risks by 2050. International efforts in solving the mercury pollution problems by preventing new emissions are to be commended; however, it may be too optimistic to expect significant fish mercury improvements in a time frame shorter than a decade, if not longer.[9] It would be beneficial to enhance monitoring, and take appropriate and timely management actions to protect human health from mercury exposure.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b03943.

Additional 2 tables and 10 figures (PDF)

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Notes

The authors declare no competing financial interest.

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DOI: 10.1021/acs.est.5b03943
Environ. Sci. Technol. 2015, 49, 14494−14502

Supporting Information

Projecting fish mercury levels in the Province of Ontario, Canada and the implications for fish and human health

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Number of pages: 24

Number of tables: 2

Number of figures: 10

Table S1: Fish consumption advisory benchmarks for mercury (µg/g wet weight) for the general and sensitive populations used by the OMOE for the advisories published in the 2013-2014 Guide to Eating Ontario Sport Fish (OMOE 2013).

Meals/month	General population	Sensitive population
8	0–0.61	0–0.26
4	0.61–1.23	0.26–0.52
2	1.23–1.84	
0	>1.84	>0.52

Table S2a: Projected changes in percent of sampled locations with a particular type of fish consumption advisory based on the fixed rate approach.

				8+ meals/month					1-4 meals/month					"do not eat"				
		Popn	Fish size	2010	2020	2030	2040	2050	2010	2020	2030	2040	2050	2010	2020	2030	2040	2050
Northern Ontario	Walleye	General	Small	75	65	51	37	17	25	35	49	63	83	0	0	0	0	0
			Medium	44	31	15	4	1	55	68	83	92	93	1	1	2	4	6
			Large	23	10	3	1	0	69	79	81	75	68	8	11	16	24	32
		Sensitive	Small	21	6	1	0	0	45	45	36	17	4	34	49	63	83	96
			Medium	5	2	0	0	0	29	17	6	2	0	66	81	94	98	100
			Large	2	0	0	0	0	13	6	2	0	0	85	94	98	100	100
	Pike	General	Small	95	94	94	94	94	5	6	6	6	6	0	0	0	0	0
			Medium	61	48	35	21	8	38	51	64	78	91	1	1	1	1	1
			Large	37	14	1	0	0	60	80	91	88	77	3	6	8	12	23
		Sensitive	Small	46	44	41	40	38	44	46	48	49	51	10	10	11	11	11
			Medium	9	1	1	0	0	40	35	20	8	1	51	64	79	92	99
			Large	3	1	0	0	0	25	5	1	0	0	72	94	99	100	100
	Lake Trout	General	Small	89	89	88	88	87	11	11	12	12	13	0	0	0	0	0
			Medium	58	56	53	51	49	39	41	44	46	48	3	3	3	3	3
			Large	35	33	31	29	28	59	61	62	64	65	6	6	7	7	7
		Sensitive	Small	39	36	34	32	31	40	42	43	44	45	21	22	23	24	24
			Medium	14	11	10	8	7	33	30	29	29	28	53	59	61	63	65
			Large	8	5	5	5	3	20	21	20	18	19	72	74	75	77	78
Southern Ontario	Walleye	General	Small	82	80	79	74	69	18	20	21	25	30	0	0	0	1	1
			Medium	54	45	40	32	23	44	53	58	65	74	2	2	2	3	3
			Large	26	20	14	7	3	70	76	81	87	90	4	4	5	6	7
		Sensitive	Small	23	17	12	7	2	55	54	52	54	52	22	29	36	39	46
			Medium	6	4	1	0	0	35	30	24	19	11	59	66	75	81	89
			Large	3	0	0	0	0	15	11	6	3	1	82	89	94	97	99
	Pike	General	Small	96	96	96	95	91	4	4	4	5	9	0	0	0	0	0
			Medium	78	74	70	65	52	22	26	30	35	48	0	0	0	0	0
			Large	53	32	25	18	6	47	67	74	81	91	0	1	1	1	3
		Sensitive	Small	55	47	38	26	19	36	43	51	63	68	9	10	11	11	13
			Medium	12	7	3	2	0	56	56	44	34	30	32	37	53	64	70
			Large	4	1	0	0	0	27	22	18	6	2	69	77	82	94	98
	Lake Trout	General	Small	89	88	87	87	84	11	12	13	13	16	0	0	0	0	0
			Medium	69	67	59	52	41	30	32	40	47	58	1	1	1	1	1
			Large	47	37	27	22	15	47	55	64	68	73	6	8	9	10	12
		Sensitive	Small	47	39	35	31	24	40	44	47	48	51	13	17	18	21	25
			Medium	20	14	3	0	0	41	39	40	32	26	39	47	57	68	74
			Large	8	2	0	0	0	26	24	20	14	5	66	74	80	86	95

Table S2b: Projected changes in percent of sampled locations with a particular type of fish consumption advisory based on the annual percent change (APC) approach.

				8+ meals/month					1-4 meals/month					"do not eat"				
		Popn	Fish size	2010	2020	2030	2040	2050	2010	2020	2030	2040	2050	2010	2020	2030	2040	2050
Northern Ontario	Walleye	General	Small	75	60	45	33	20	25	40	54	61	69	0	0	1	6	11
			Medium	44	34	24	15	9	55	64	68	70	68	1	2	8	15	23
			Large	23	16	10	8	6	68	70	68	60	53	9	14	22	32	41
		Sensitive	Small	21	11	6	3	2	45	38	31	20	10	34	51	63	77	88
			Medium	5	3	3	2	2	29	21	13	6	3	66	76	84	92	95
			Large	2	2	2	2	1	13	8	5	4	3	85	90	93	94	96
	Pike	General	Small	95	94	93	92	90	5	6	7	8	10	0	0	0	0	0
			Medium	61	47	34	21	11	39	52	63	71	74	0	1	3	8	15
			Large	37	23	10	4	2	60	68	72	63	46	3	9	18	33	52
		Sensitive	Small	46	43	40	38	36	44	46	48	47	48	10	11	12	15	16
			Medium	9	4	2	1	1	40	32	21	11	5	51	64	77	88	94
			Large	3	1	1	1	1	25	13	5	2	0	72	86	94	97	99
	Lake Trout	General	Small	89	83	76	71	64	11	17	24	28	35	0	0	0	1	1
			Medium	58	48	36	30	25	39	48	60	63	65	3	4	4	7	10
			Large	37	23	10	4	2	60	68	72	63	46	3	9	18	33	52
		Sensitive	Small	39	32	26	24	18	40	43	43	36	34	21	25	31	40	48
			Medium	14	10	8	6	4	33	26	21	18	14	53	64	71	76	82
			Large	8	5	5	4	3	20	19	16	13	9	72	76	79	83	88
Southern Ontario	Walleye	General	Small	82	82	83	83	84	18	18	17	17	16	0	0	0	0	0
			Medium	54	51	46	43	41	44	47	51	54	56	2	2	3	3	3
			Large	26	20	14	9	6	70	74	75	72	71	4	6	11	19	23
		Sensitive	Small	23	23	24	26	28	55	56	55	53	51	22	21	21	21	21
			Medium	6	6	5	4	4	35	34	32	30	28	59	60	63	66	68
			Large	3	1	1	0	0	15	13	8	6	4	82	86	91	94	96
	Pike	General	Small	96	96	94	90	89	4	4	6	10	11	0	0	0	0	0
			Medium	78	72	66	60	44	22	28	34	40	56	0	0	0	0	0
			Large	53	51	48	48	47	47	49	51	51	52	0	0	1	1	1
		Sensitive	Small	55	49	40	38	31	36	41	49	49	53	9	10	11	13	16
			Medium	12	7	7	5	4	56	54	39	31	27	32	39	54	64	69
			Large	4	4	4	4	3	27	27	27	27	28	69	69	69	69	69
	Lake Trout	General	Small	89	87	80	74	60	11	13	20	26	39	0	0	0	0	1
			Medium	69	61	48	34	27	30	37	46	58	60	1	2	6	8	13
			Large	47	34	26	22	16	46	55	58	60	60	7	11	16	18	24
		Sensitive	Small	47	37	34	28	23	40	43	40	32	28	13	20	26	40	49
			Medium	20	14	9	3	1	41	34	26	24	23	39	52	65	73	76
			Large	8	5	2	0	0	26	20	18	16	11	66	75	80	84	89

Figure S1. Illustration of screening of sampling events for (a,b) minimum number of samples for a species in a sampling event, (c,d) minimum size range for a species, and (e-h) calculating 50 cm std-length mercury concentration. To avoid large extrapolation of the power series regressions while calculating each std-length fish concentration, only sampling events with the smallest fish smaller than a std-length plus 15 cm (e) and the largest fish larger than std-length minus 15 cm (g) were considered. For example, as illustrated in this figure, to calculate 50 cm WE mercury for a particular year/location, the smallest WE measured for that sampling event should be smaller than 65 cm (e) and the largest fish should be larger than 35 cm (g).

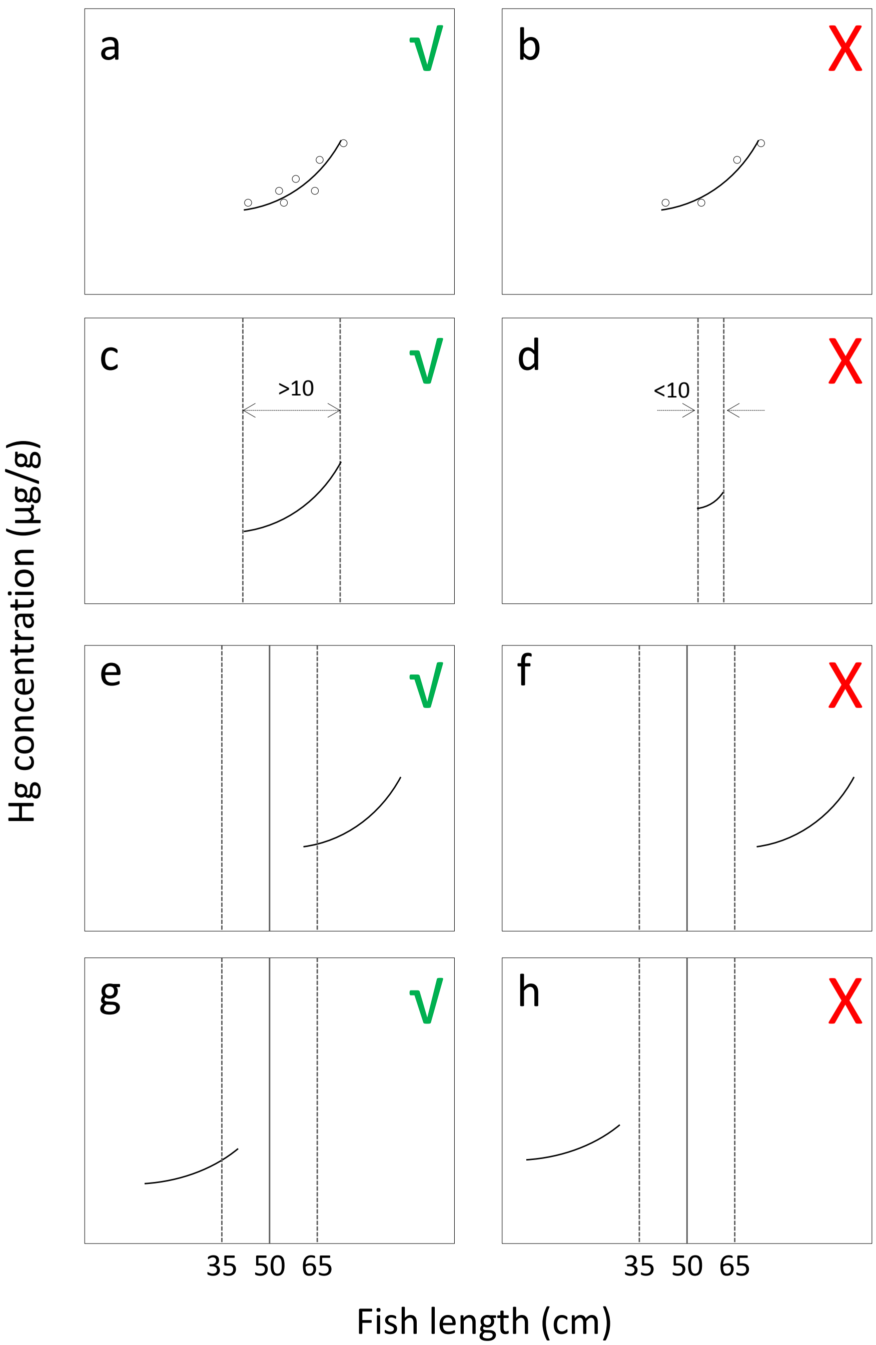

Figure S2. Illustration of estimating mercury levels in WE at the standard lengths (Std-Length) of 40 cm (small), 50 cm (medium) and 60 cm (large) for a particular location for (a) present day (2010) and (b) future years.

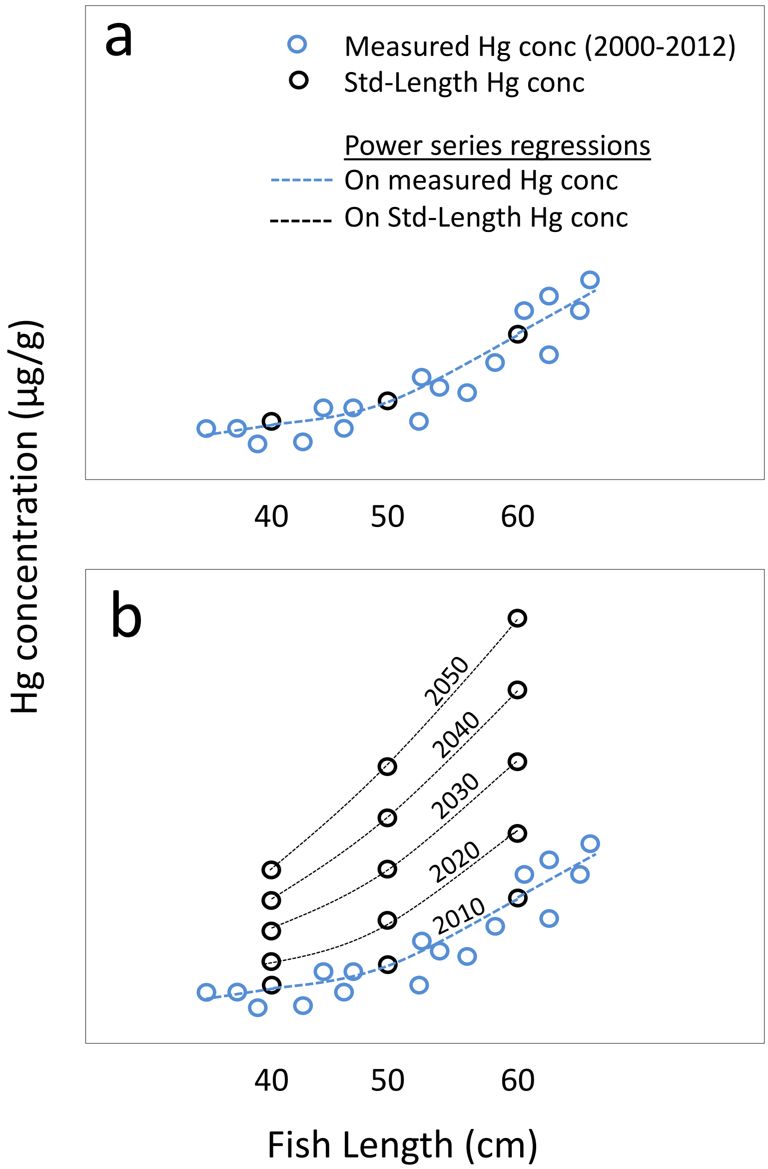

Figure S3. Illustration of estimating (a) mercury levels in WE at the maturity length of 43 cm and (b) WE lengths to reach *do not eat* advisory benchmark of 1.84 µg/g wet weight for the general population used by OMOE over time for a particular location and scenario (i.e., fixed rate or annual percent change APC). Black circles represent mercury concentrations for small (40 cm), medium (50 cm) and large (60 cm) sizes of WE for different years, black dotted lines represent power series regressions for different years, black solid straight lines represent (a) WE length at maturity (43 cm) and (b) the *do not eat* advisory benchmark, red arrows represent (a) mercury concentrations at the maturity length and (b) WE length at the *do not eat* advisory benchmark for different years, and blue dotted line represents an extrapolation.

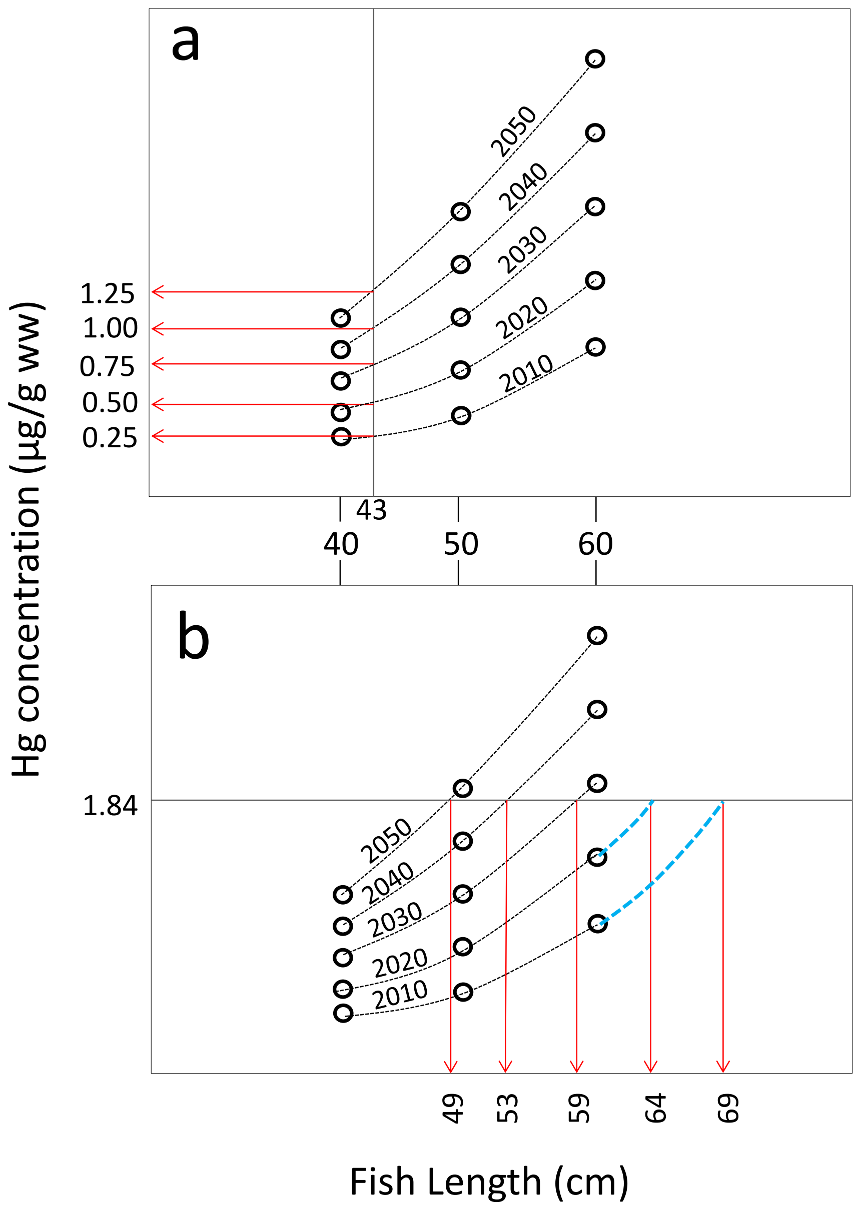

Figure S4. Projected changes with time in cumulative percentage (%) of northern and southern Ontario waterbodies that are exceeding skin-off fillet mercury concentrations of 0.52 µg/g (wet weight) at varying lengths of Walleye, Northern Pike and Lake Trout under the constant rates of change and annual percent change (APC) approaches. The mercury concentration of 0.52 µg/g is used as *'do not eat'* advisory benchmarks by the OMOE for the sensitive population of children and women of child-bearing age.

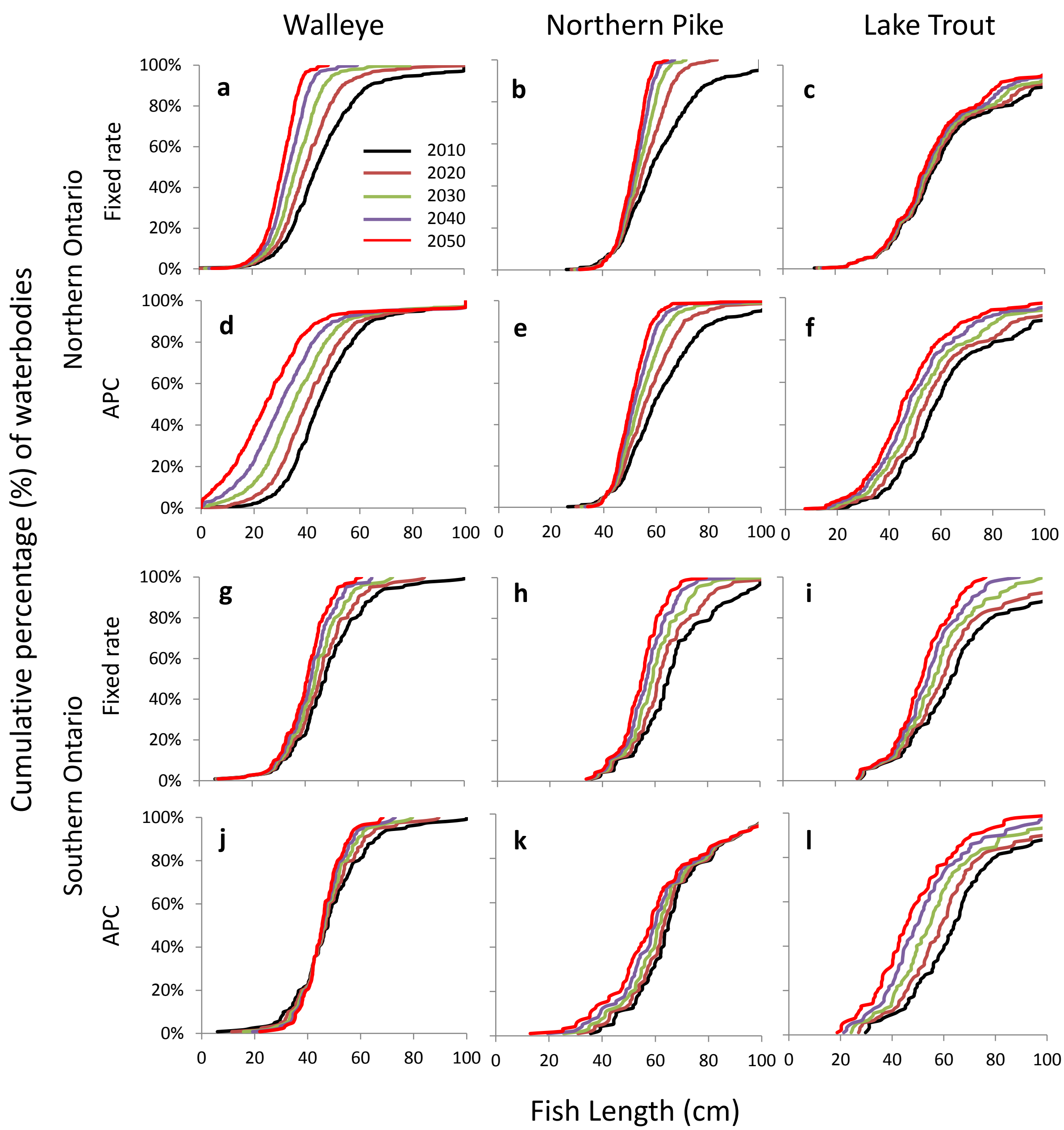

Figure S5. Projected 2050 mercury concentrations (µg/g wet weight) in skin-off fillets of small, medium and large sized Ontario Walleye, Northern Pike and Lake Trout under the annual percent change (**APC**) approach. The concentrations have been grouped into the various categories used by the Ontario Ministry of the Environment for the purpose of fish consumption advisories geared towards the **general population** (OMOE 2013). *n* represents number of locations.

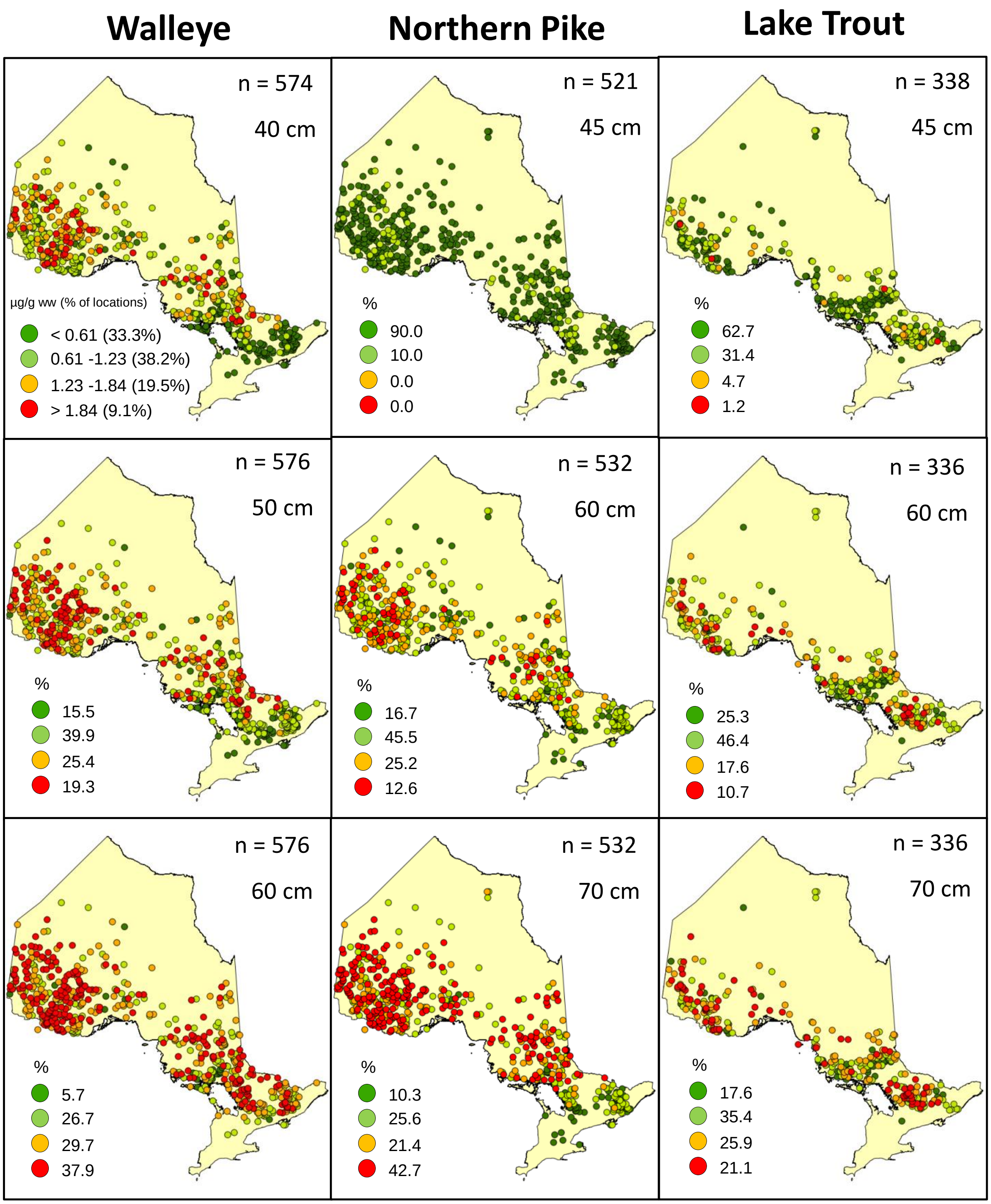

Figure S6a. Mercury concentrations (µg/g wet weight) in skin-off fillets of small, medium and large sized Ontario Walleye, Northern Pike and Lake Trout collected between **2000-2010**. The concentrations have been grouped into the various categories used by the Ontario Ministry of the Environment for the purpose of fish consumption advisories geared towards the **sensitive population** (OMOE 2013). *n* represents number of locations.

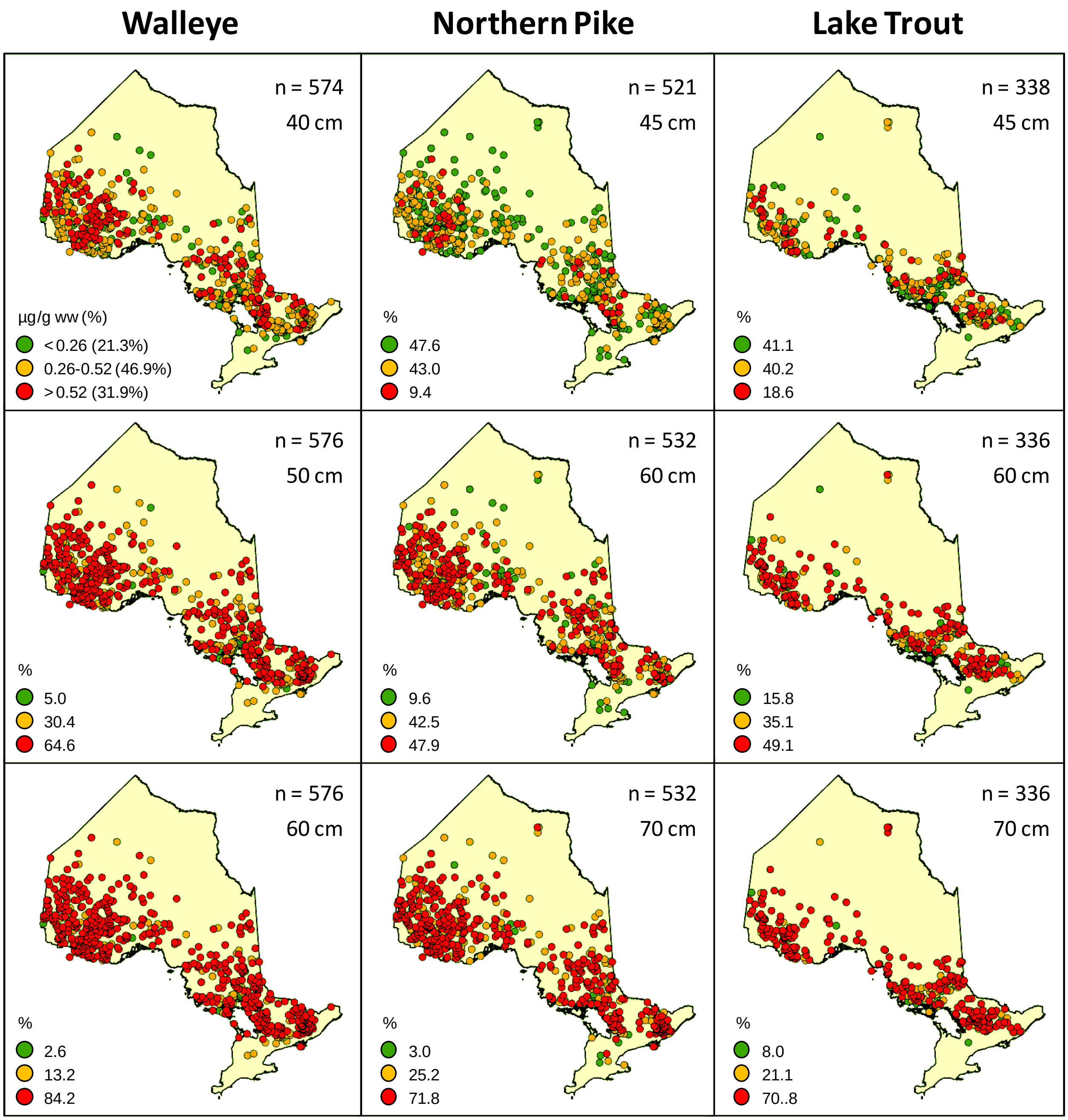

Figure S6b. Projected 2050 mercury concentrations (µg/g wet weight) in skin-off fillets of small, medium and large sized Ontario Walleye, Northern Pike and Lake Trout under the **Fixed Rate** approach. The concentrations have been grouped into the various categories used by the Ontario Ministry of the Environment for the purpose of fish consumption advisories geared towards the **sensitive population** (OMOE 2013). *n* represents number of locations.

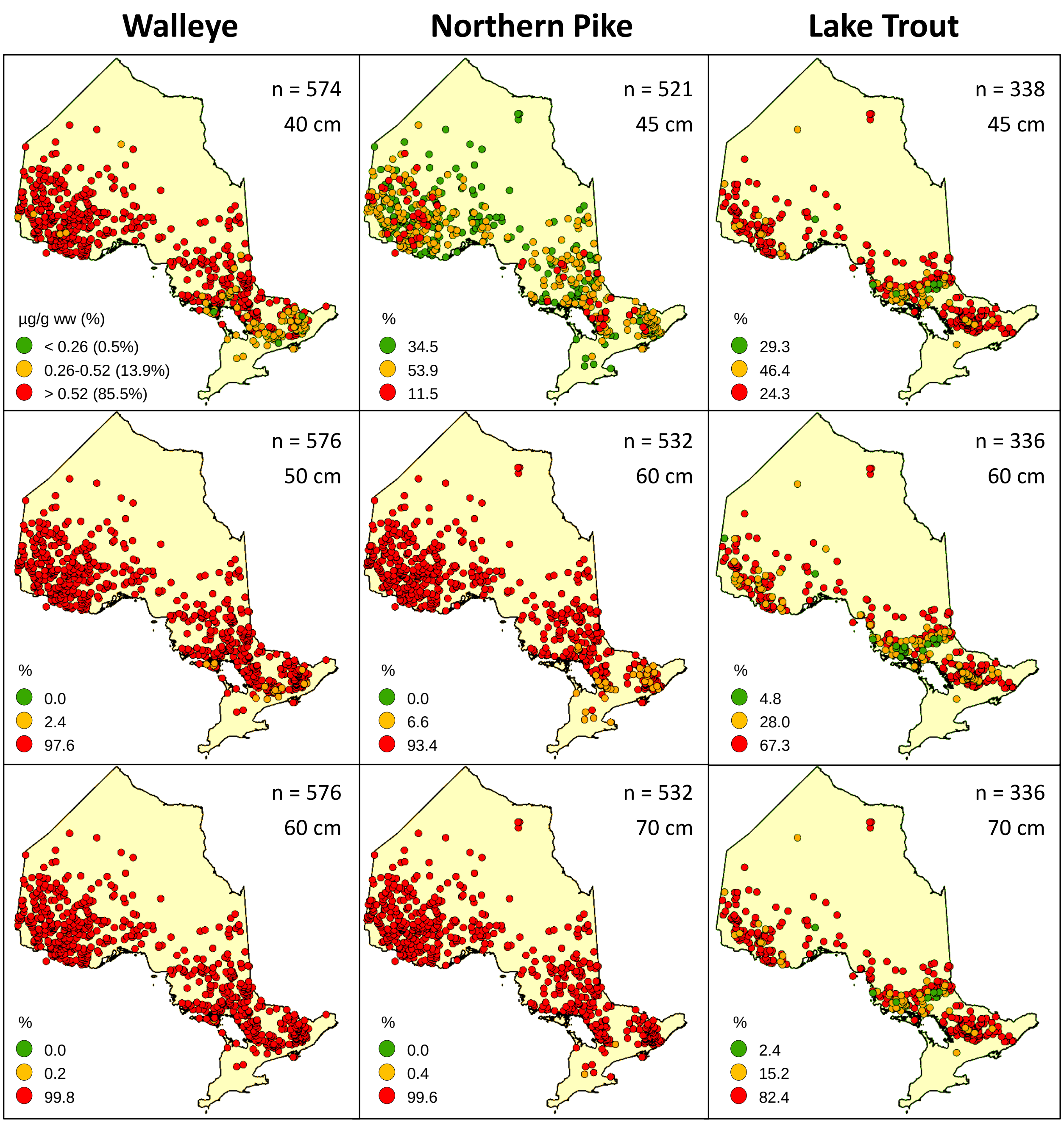

Figure S6c. Projected 2050 mercury concentrations (µg/g wet weight) in skin-off fillets of small, medium and large sized Ontario Walleye, Northern Pike and Lake Trout under the annual percent change (**APC**) approach. The concentrations have been grouped into the various categories used by the Ontario Ministry of the Environment for the purpose of fish consumption advisories geared towards the **sensitive population** (OMOE 2013). *n* represents number of locations.

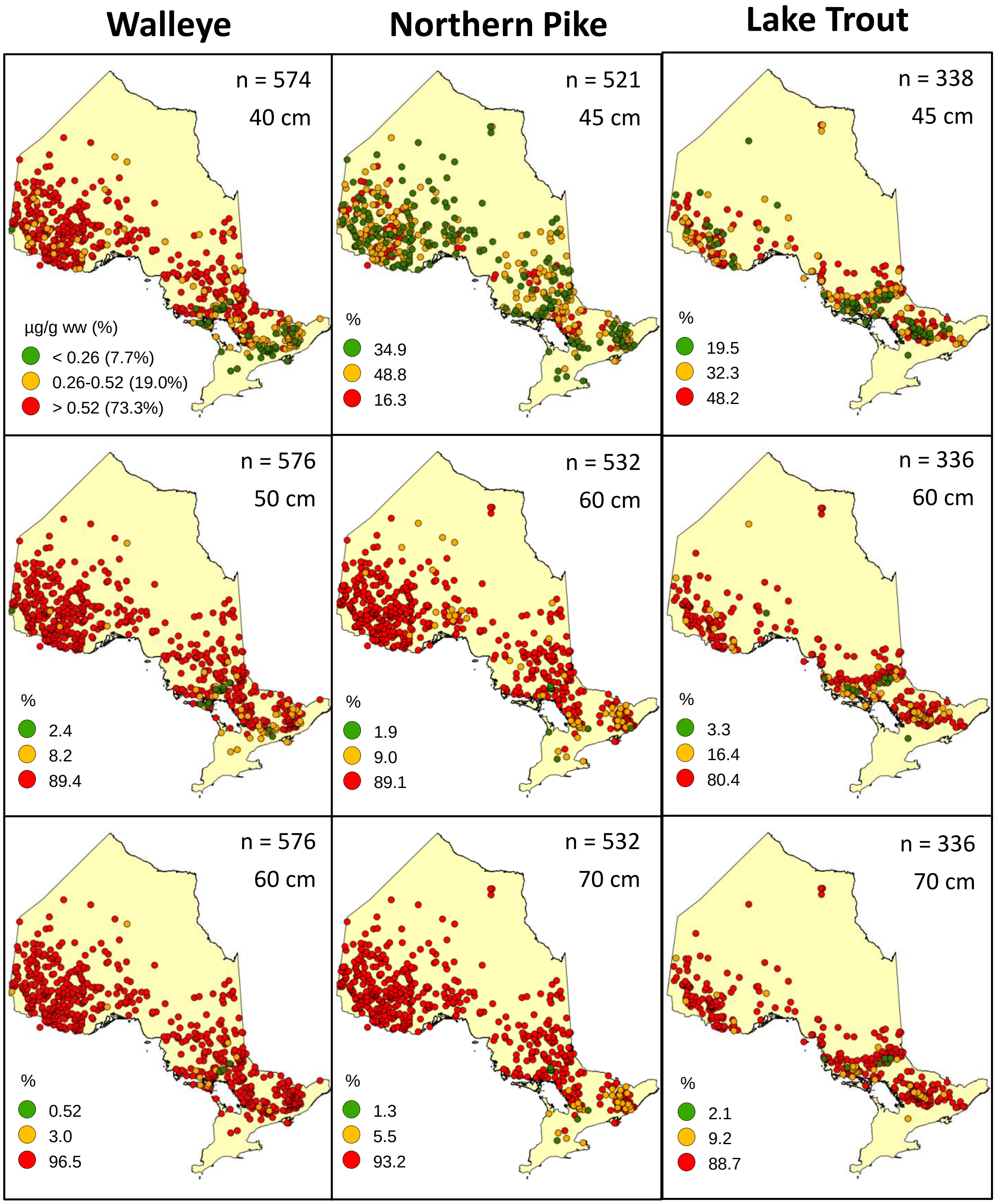

Figure S7a. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **small** sized Walleye, Northern Pike and Lake Trout locations in **northern Ontario** for the general and sensitive populations under the **Average** scenario of the constant rates of change approach.

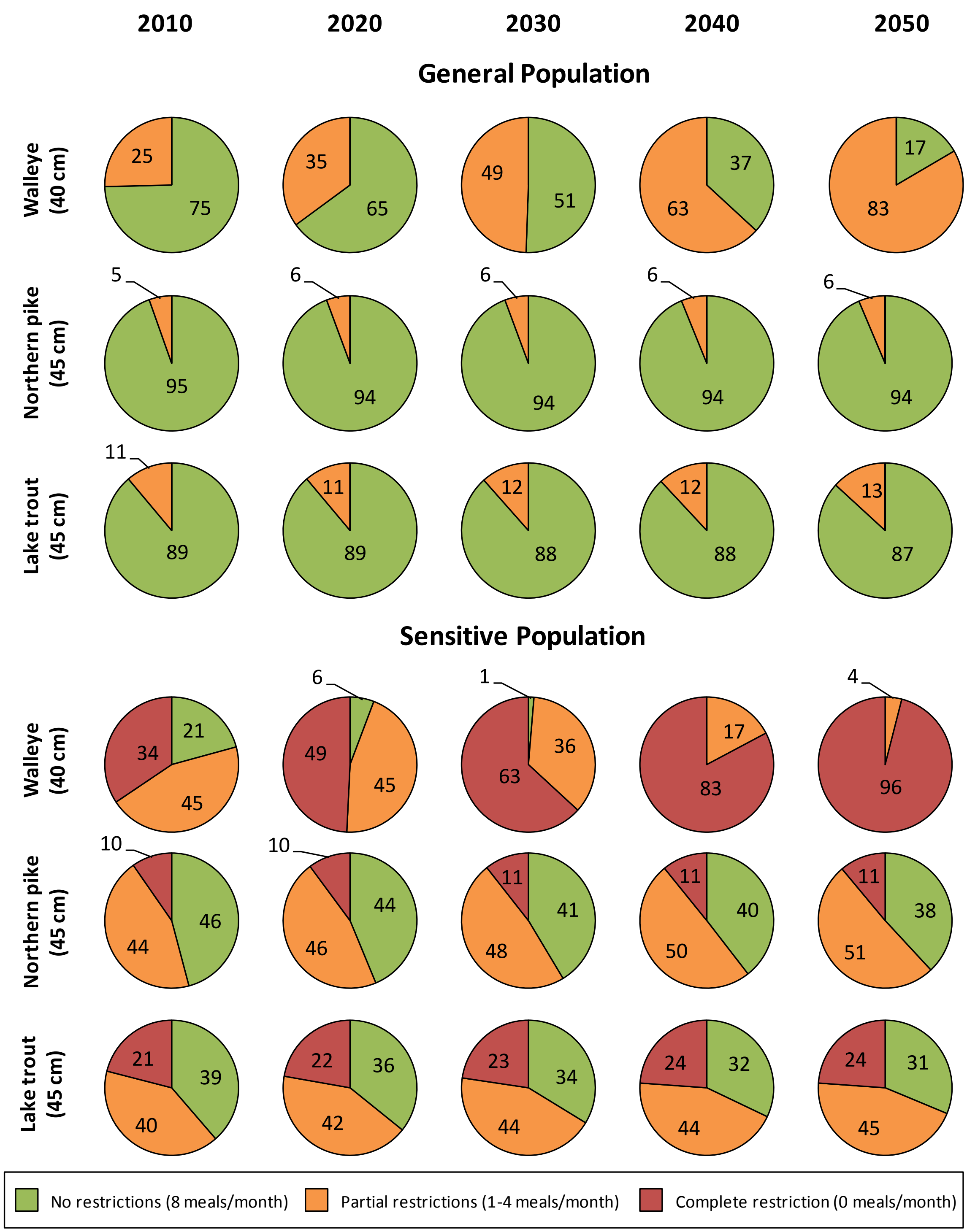

Figure S7b. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **medium** sized Walleye, Northern Pike and Lake Trout locations in **northern Ontario** for the general and sensitive populations under the **Average** scenario of the constant rates of change approach.

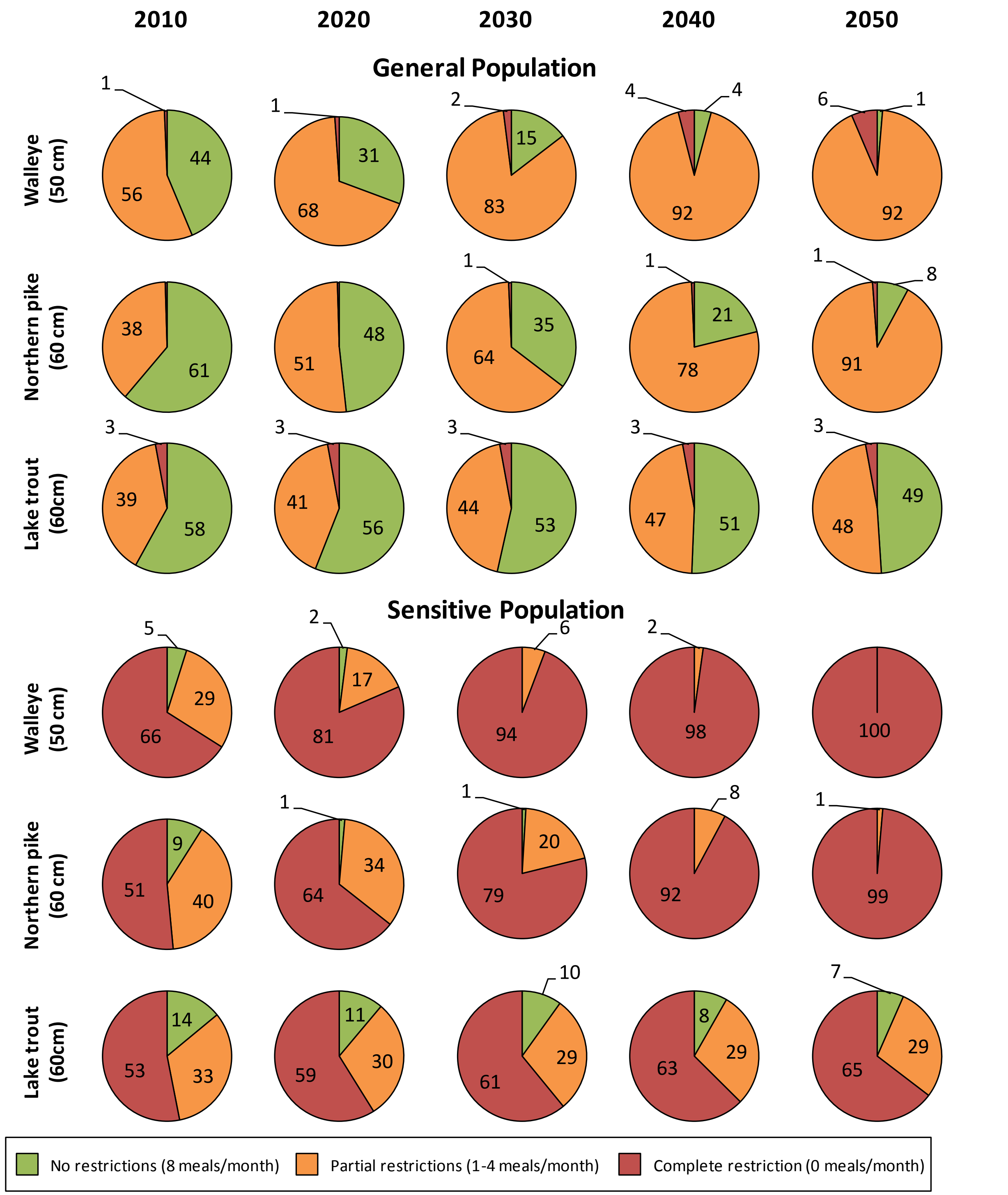

Figure S7c. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **large** sized Walleye, Northern Pike and Lake Trout locations in **northern Ontario** for the general and sensitive populations under the **Average** scenario of the constant rates of change approach.

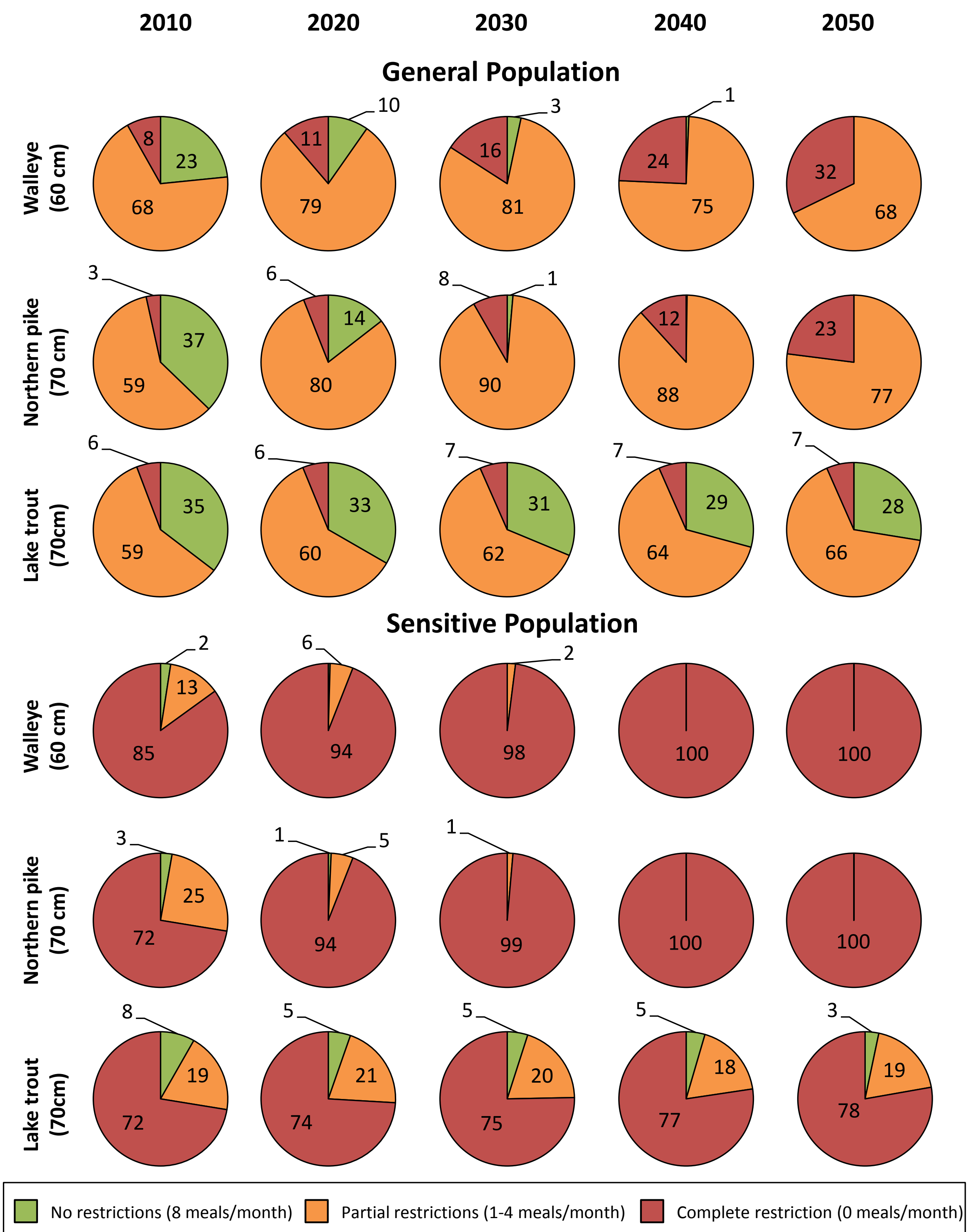

Figure S8a. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **small** sized Walleye, Northern Pike and Lake Trout locations in **northern Ontario** for the general and sensitive populations under the annual percent change **(APC)** approach.

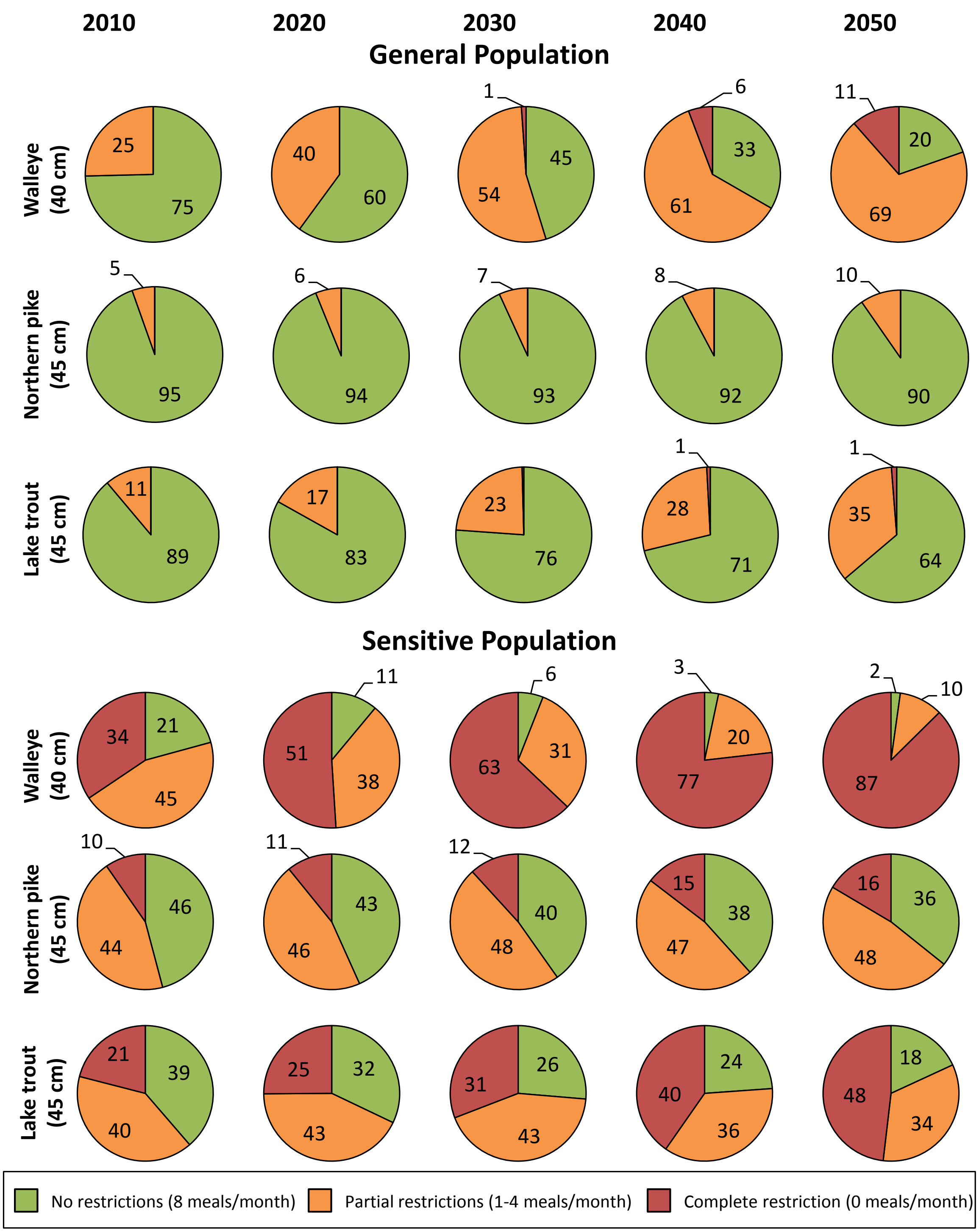

Figure S8b. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **medium** sized Walleye, Northern Pike and Lake Trout locations in **northern Ontario** for the general and sensitive populations under the annual percent change **(APC)** approach.

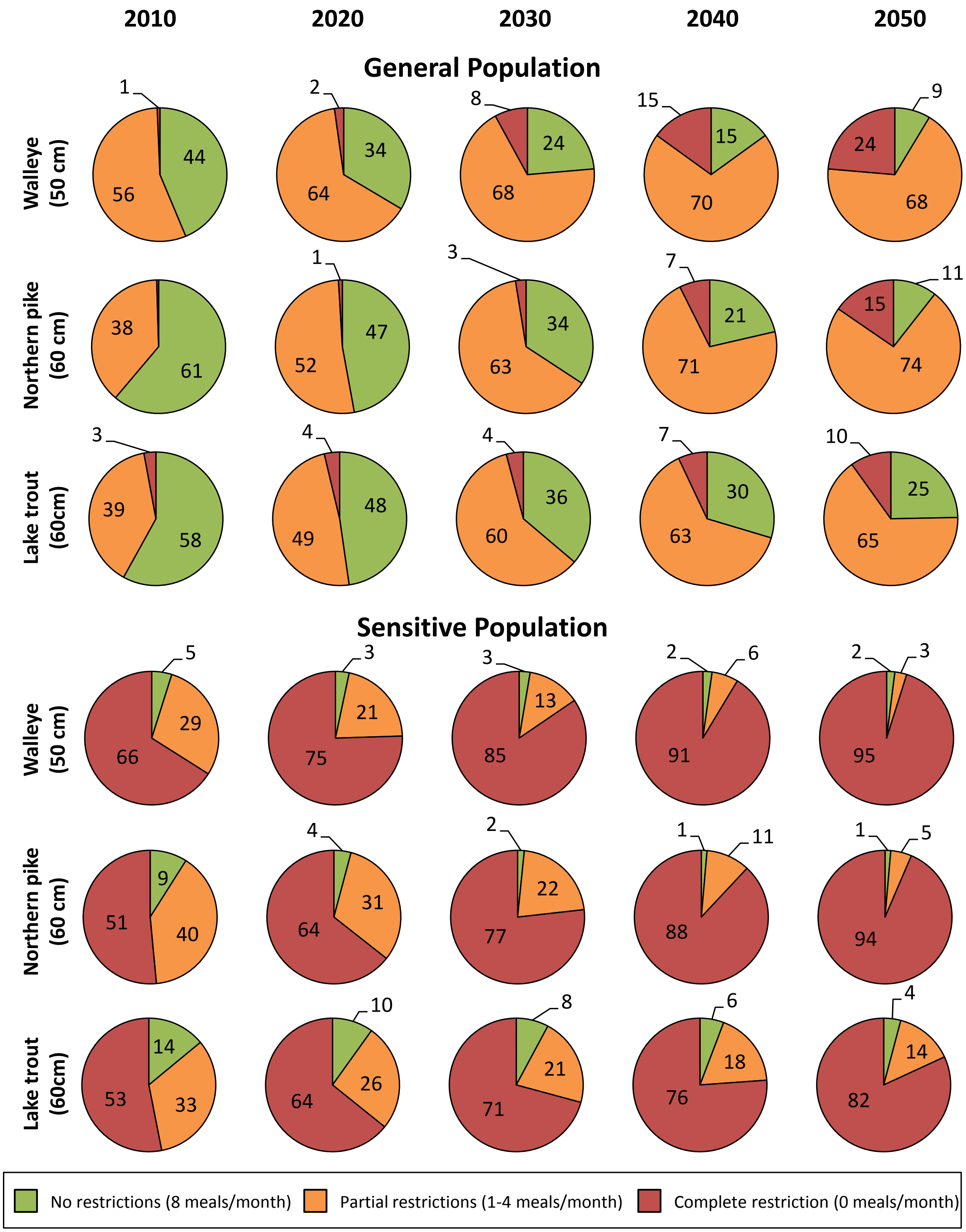

Figure S8c. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **large** sized Walleye, Northern Pike and Lake Trout locations in **northern Ontario** for the general and sensitive populations under the annual percent change **(APC)** approach.

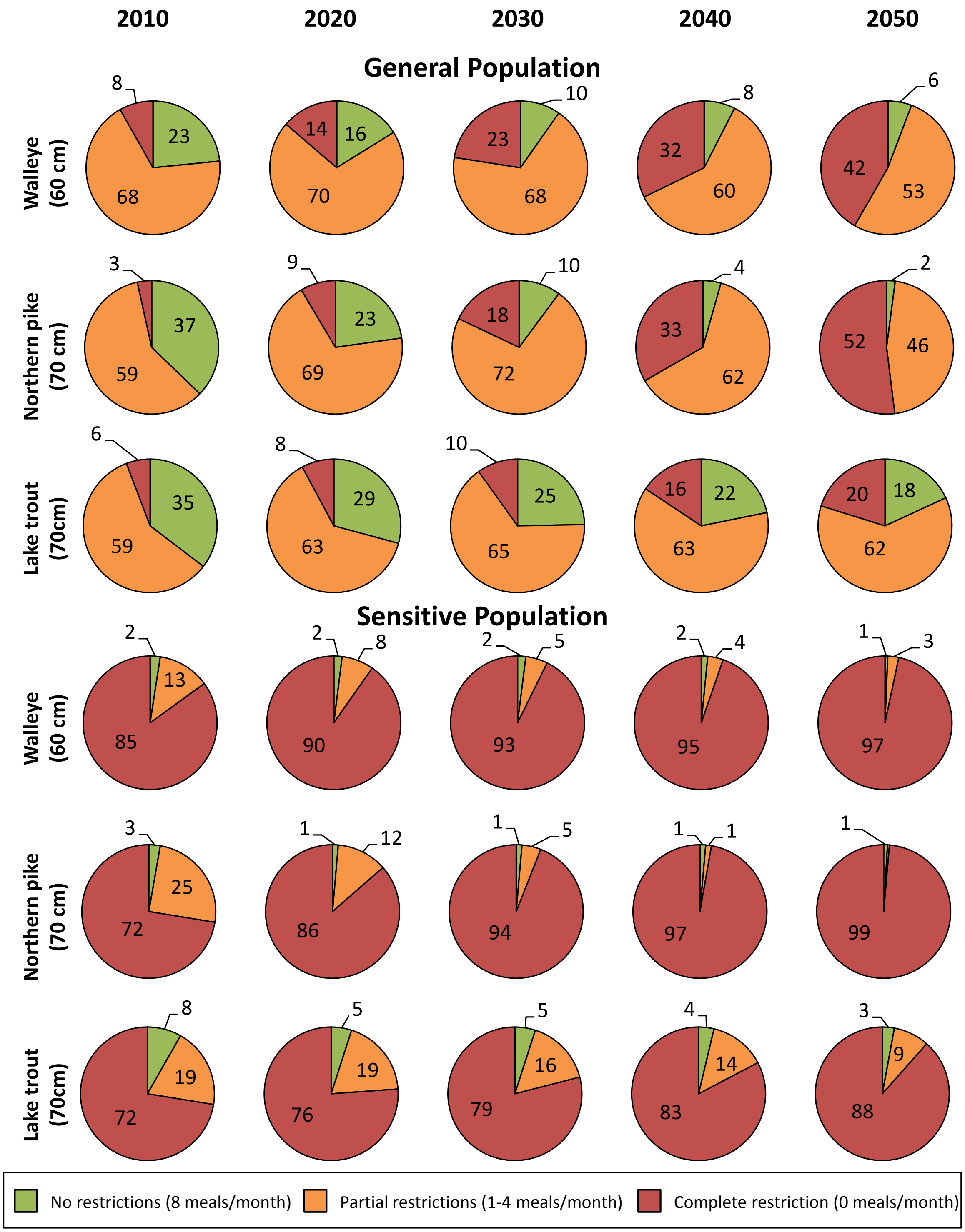

Figure S9a. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **small** sized Walleye, Northern Pike and Lake Trout locations in **southern Ontario** for the general and sensitive populations under the **Average** scenario of the constant rates of change approach.

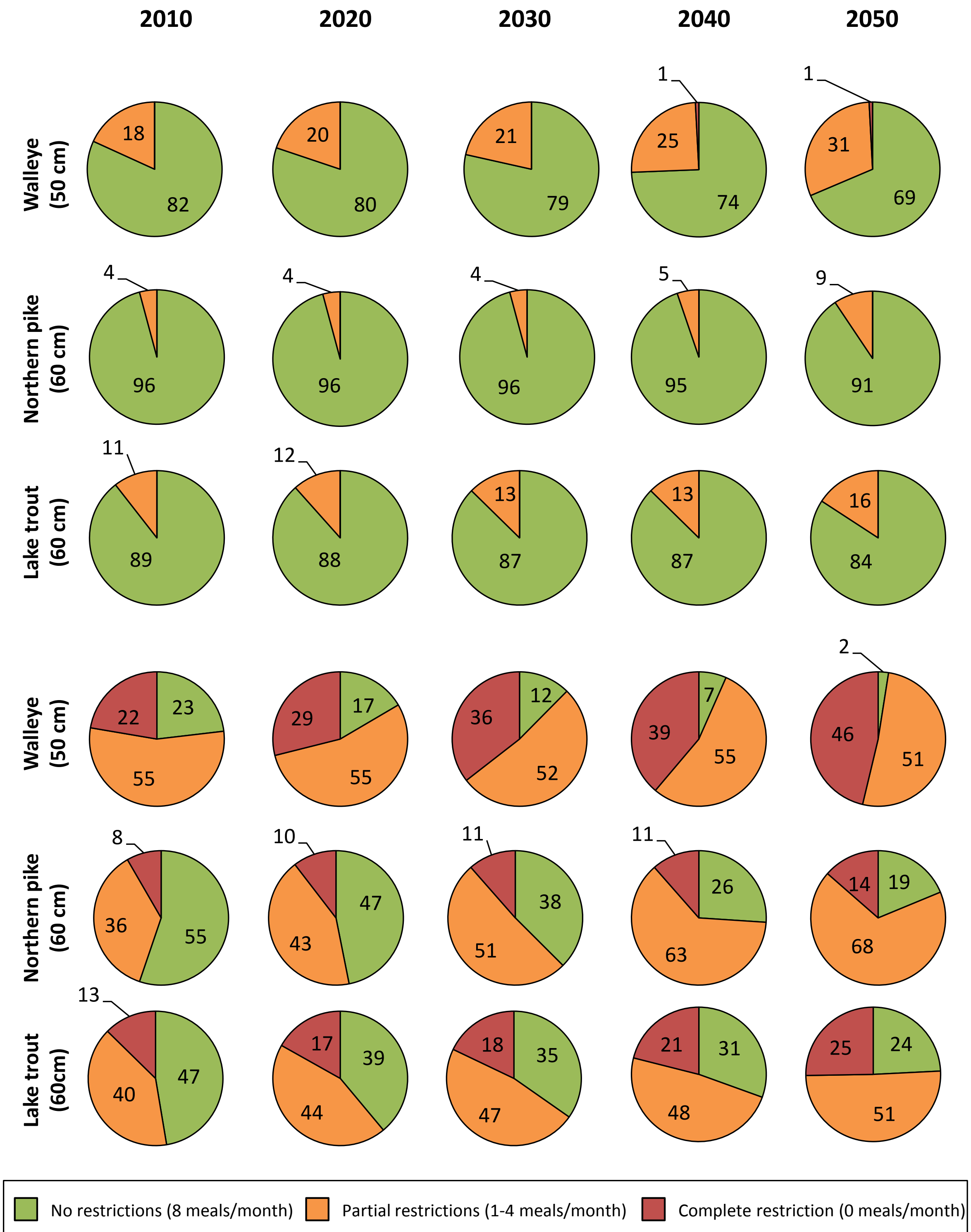

Figure S9b. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **medium** sized Walleye, Northern Pike and Lake Trout locations in **southern Ontario** for the general and sensitive populations under the **Average** scenario of the constant rates of change approach.

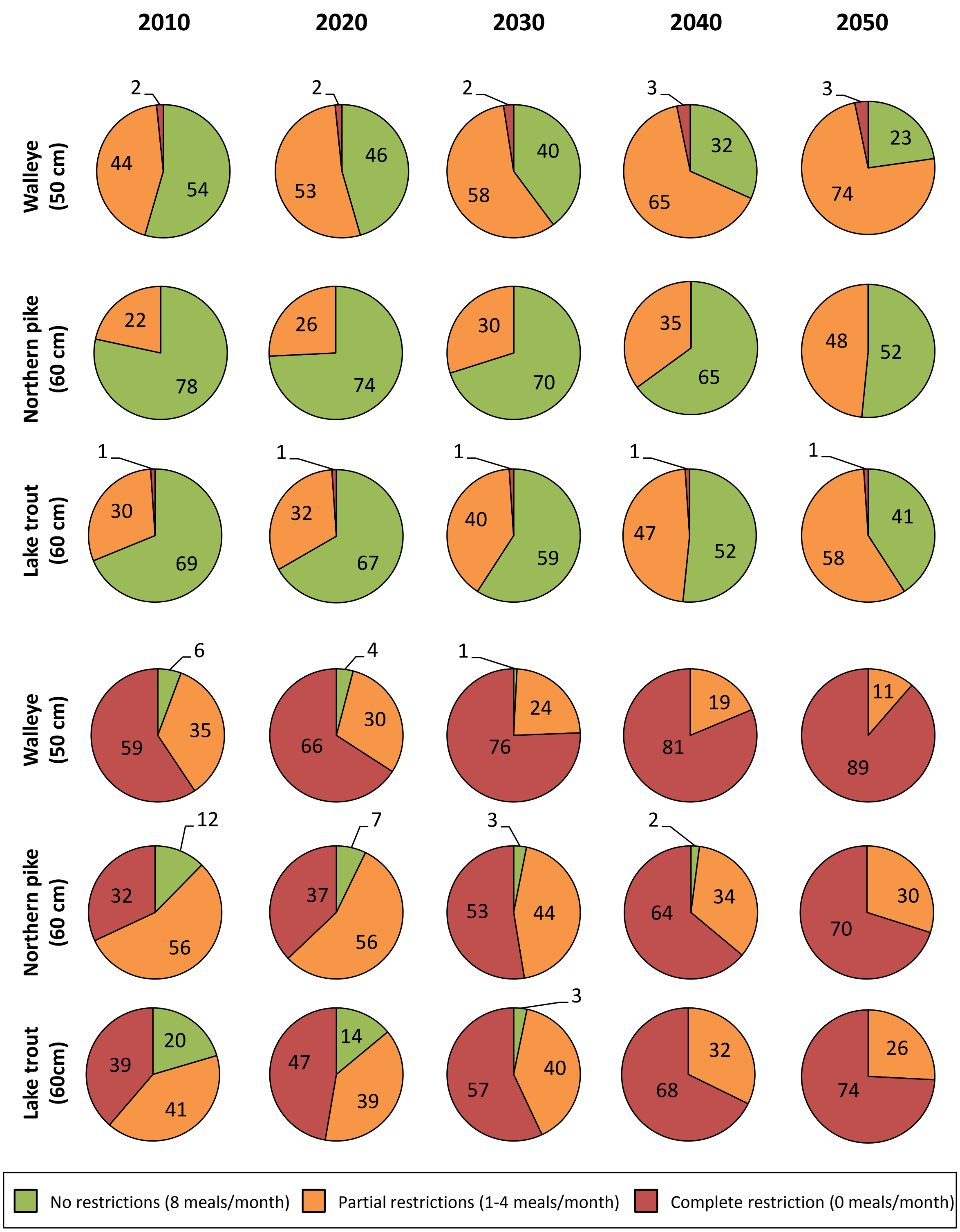

Figure S9c. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **large** sized Walleye, Northern Pike and Lake Trout locations in **southern Ontario** for the general and sensitive populations under the **Average** scenario of the constant rates of change approach.

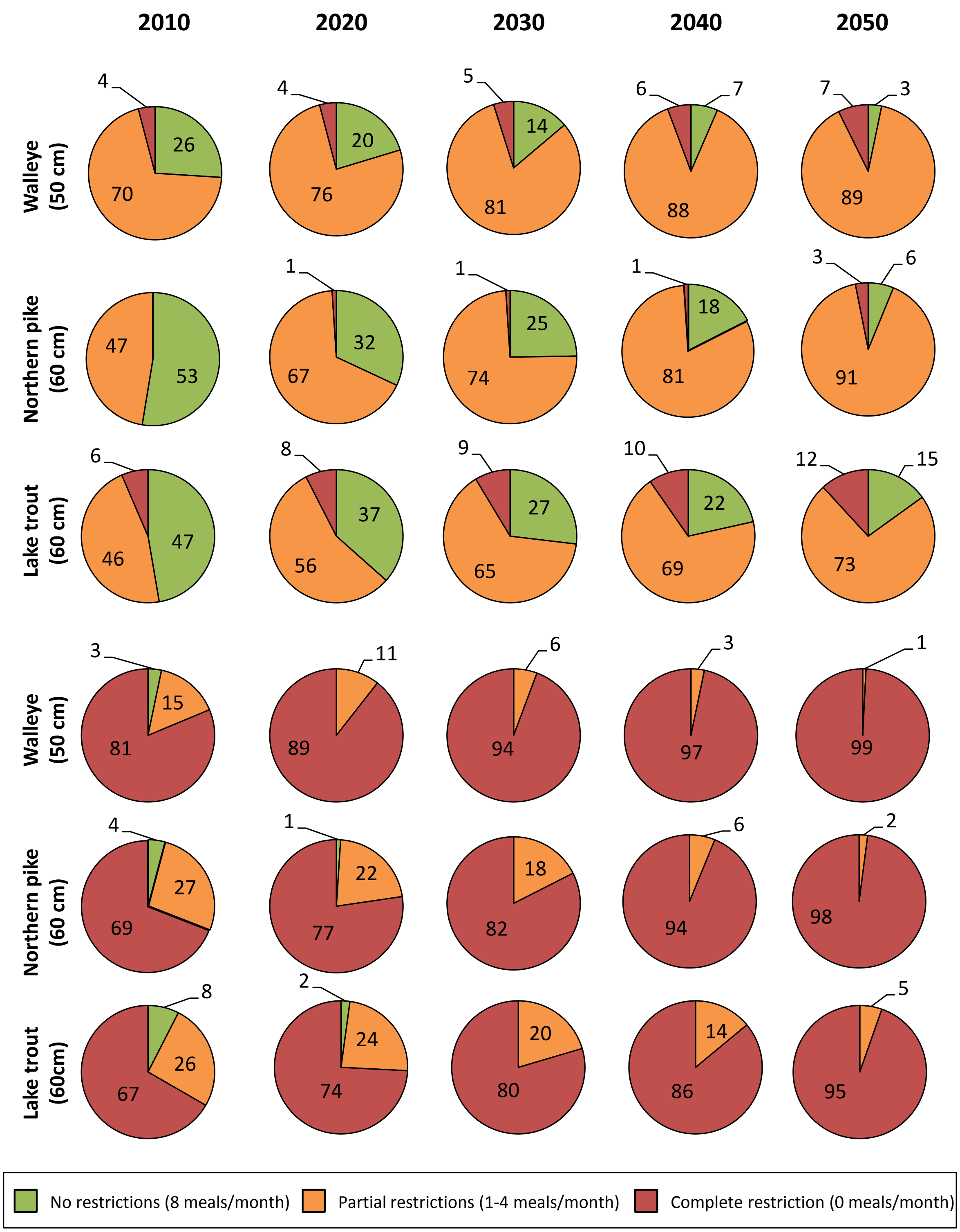

Figure S10a. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **small** sized Walleye, Northern Pike and Lake Trout locations in **southern Ontario** for the general and sensitive populations under the annual percent change **(APC)** approach.

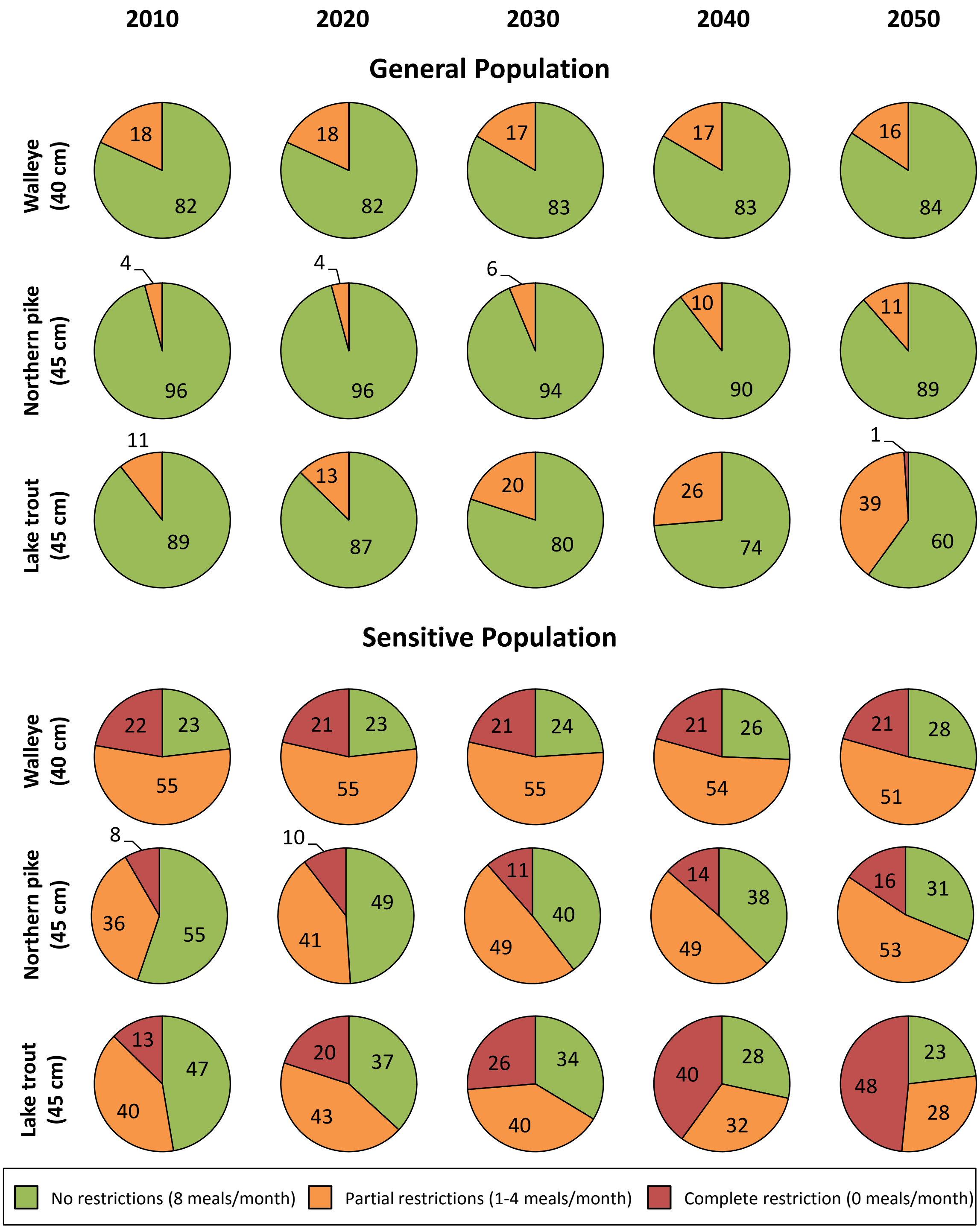

Figure S10b. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **medium** sized Walleye, Northern Pike and Lake Trout locations in **southern Ontario** for the general and sensitive populations under the annual percent change **(APC)** approach.

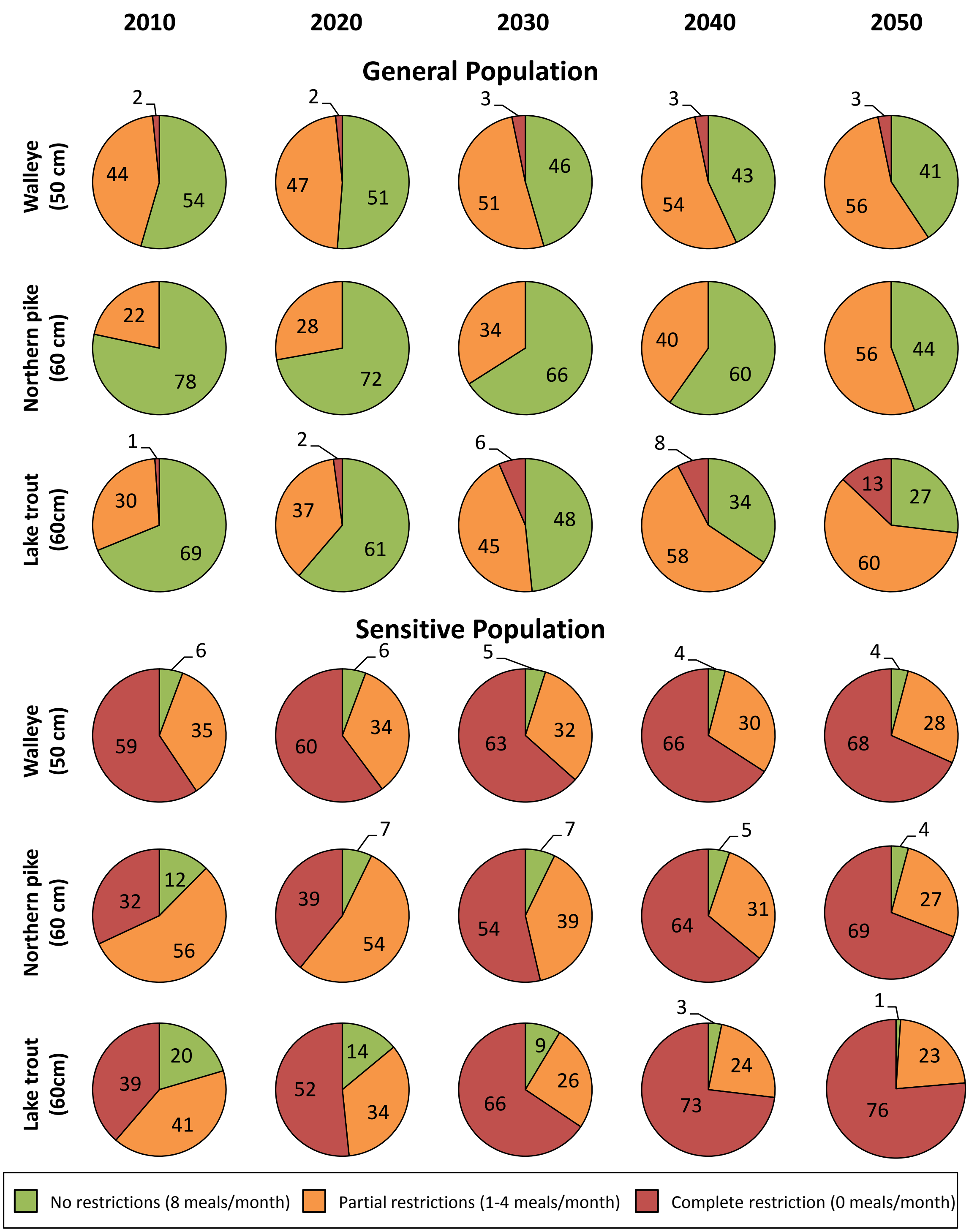

Figure S10c. Pie charts showing changes in breakdown of fish consumption advisories between 2010 and 2050 for **large** sized Walleye, Northern Pike and Lake Trout locations in **southern Ontario** for the general and sensitive populations under the annual percent change **(APC)** approach.