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Mercury in Marine Mussels from the St. Lawrence Estuary and Gulf (Canada): A Mussel Watch Survey Revisited after 40 Years

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Abstract: Various species of marine mussels have been used, in the last 50 years, as sentinel organisms for monitoring metal contamination along marine coasts. There are two main reasons for this: these mollusks concentrate metals in their soft tissue and they are geographically widespread. In practice, trace metal concentrations in mussel soft tissue reveal (after some correction for biotic effects) the contamination level of their surrounding environment. We present the results of a mercury (Hg) survey in *Mytilus* spp. collected in the summers of 2016, 2018, and 2019 at 51 stations distributed along the coasts of the Estuary and Gulf of St. Lawrence. Mercury concentrations ranged from 0.063 to 0.507 µg g^{-1} (dry weight, dw), with a grand mean of 0.173 ± 0.076 µg g^{-1} dw (±1 standard deviation), and a median of 0.156 µg g^{-1} dw for the 504 individuals analyzed. Mercury contents per individual mussel were significantly ($p < 0.01$) related to shell length and dry tissue weight, with the smaller individuals having the highest Hg concentrations. To take into account these biotic effects, we normalized Hg concentrations of the mussel soft tissue for constant shell length (L) and soft tissue weight (TW) based on the log-log relationships between Hg content and L or TW. The normalized Hg contents of mussels varied from 10.9 to 66.6 ng per virtual individual of 35 mm length and 0.17 g dry weight. A similar normalization procedure applied to 1977–1979 data, yielded a very similar range: 12 to 64 ng. This observation suggests that the Hg bioavailable to marine mussels in the study area did not change over a span of 40 years. Regional Hg distribution patterns indicate a gradual decrease of Hg content in mussels downstream from freshwater discharges to the St. Lawrence Estuary and the Baie des Chaleurs, suggesting that rivers constitute a significant Hg source in these estuarine systems. Atmospheric Hg deposition and concentration in marine waters of the Atlantic Ocean are known to have decreased in the last decades. However, in coastal environments, the response to these changes does not seem to be rapid, probably because of the long residence time of Hg in soils before being exported to coastal areas.

Keywords: mercury; mussel; mussel watch; *Mytilus*; St. Lawrence

1. Introduction

Marine mussels (*Mytilus* spp.) have been successfully used over the past fifty years as sentinel organisms for monitoring metal contamination along marine coasts [1–9]. The reason for this is that this mollusk genus is geographically widespread in sub-boreal and temperate environments and it concentrates metals in its soft tissue in proportion with the concentration in surrounding waters [10–12]. Thus, trace metal concentrations in the soft tissue of the blue mussel reveal the contamination level of the waters of its environment. Applied to the monitoring of temporal and geographical trends of chemical contamination, this approach has been named "Mussel Watch", and has been adopted as one of several coastal environmental quality-monitoring strategies by United Nations programs [3,13]. However, biological factors related to mollusk growth rate also control metal uptake and excretion and must be taken into account in order to optimize the use of mussels as sentinel organisms [4,10,14–16]. Several Mussel Watch programs have been carried out to monitor trace metal contamination along the eastern coasts of Canada and US [8,17–19], and especially mercury (Hg) contamination in the Estuary and Gulf of St. Lawrence [1,20,21]. These programs have not been maintained over more than a few years and, consequently, temporal trends are not as well documented. Nevertheless, they constitute baselines against which future assessments can be compared [22].

Here, we present the results of a Hg survey carried out in the summers of 2016, 2018, and 2019 in the St. Lawrence Estuary and Gulf, forty years after the first Mussel Watch was completed in this area. We used a normalization model to minimize the effects of biological factors on the Hg content of the mussel soft tissues. Our observations suggest that the amount of Hg bioavailable to marine mussels in the study area is similar to what it was 40 years ago. Sub-regional Hg distribution patterns indicate a gradual decrease of Hg content in mussels downstream from the main freshwater tributary (St. Lawrence River) to the St. Lawrence Estuary.

2. Materials and Methods

2.1. Sampling and Pre-Treatment

Adult specimens of *Mytilus edulis* ranging from 23 to 84 mm in length were sampled at 51 sites, at mid-tide level along the shores of the Estuary and Gulf of St. Lawrence (Figure 1). Geographical coordinates are given in Appendix A Table A1. Ten to twelve specimens (except for Station 36 with only 4 individuals) were collected at each site between 2–10 August 2016 along the South Shore of the Lower St. Lawrence Estuary (LSLE) and around the Gaspé Peninsula, between 27 August and 8 September 2018 along the North shore of the LSLE and Moyenne Côte Nord, and between 22–30 August 2019 along the Baie des Chaleurs, Northumberland Strait and Cape Breton (Figure 1). A few individuals with a shell deformity or green soft tissue were excluded to avoid the species *Coccomyxa*-infested *Mytilus trossulus*, for which background information about their trace element bioaccumulation properties is scarce [23–25]. In the field, mussel shell lengths were measured with a Vernier caliper, weighted, and their soft tissues freed from the shell. Soft tissues were kept at +4 °C in 2 mL of an ethanolic solution (90%, *v*/*v*) during the sampling periods, then kept at −18 °C until freeze-dried.

2.2. Chemical Analyses

The freeze-dried soft tissue of each mussel was individually analyzed using an automated atomic absorption spectrometer designed for Hg determinations (AMA-254, Altec, Czech Republic). The analytical procedure is described as EPA Method 7473 [26]. The analytical accuracy was checked every 10 determinations, using Certified Reference Materials (CRM): BCR-278R from the Institute for Reference Materials and Measurements, or IAEA-336 from the International Atomic Energy Agency, or DORM-4 from the National Research Council of Canada. BCR-278R is a mussel tissue, whereas IAEA-336 and DORM-4 are lichen and fish muscle, respectively. Analytical results were always within the target range of the certified values. Reproducibility, calculated as the variation coefficient (i.e., confidence interval/mean) of 6 replicate analyses of CRMs, varied between 2% and 5%. The detection limit, defined as 3 times the standard deviation of the 6 blank replicates, was 0.002 μg g^{-1} (dry weight, dw). The amount of Hg dissolved in the ethanolic solution was measured in 20 samples taken at random; it never exceeded 2% of the total Hg burden in the soft tissues.

2.3. Normalization Procedure and Statistics

Mercury mussel content depends upon mussel shell length and soft tissue mass. The former because Hg accumulated throughout the lifespan of the mussel, the latter mainly because of its seasonal weight gains and losses [4,10]. To minimize the influence of mussel size on their Hg content, the normalization procedure described by Cossa and Rondeau [1] was applied. Normalization consists of correcting Hg raw data based on a multilinear regression function relating logHg content and shell length (logL) and soft tissue dry weight (logTW) (see Section 4.3). This normalization minimizes the influence of size on Hg content of the animals, thereby enabling the interpretation of mussel Hg content in terms of Hg bioavailability in their surrounding environment. According to Cossa and Rondeau [1] a two-fold Hg content distinction, environmentally-sound in terms of Hg bioavailability, can thereby be achieved.

The statistical analyses were performed with XLSTAT software from Addinsoft (https://www.xlstat.com/). As their variance tends to increase with their mean, the Hg concentration values were $\log_{10}$-transformed before any statistical treatment.

3. Hydrological and Ecological Settings of the Area of Study

The studied area comprises two main regions, the LSLE and Gulf of St. Lawrence (Figure 1). The LSLE stretches from the mouth of the Saguenay Fjord (Station 26) to Pointe des Monts (Station 35); the south and north shores of the LSLE strongly differ in their hydrographic characteristics. The south shore is characterized by an estuarine circulation as the brackish waters, flowing out the St. Lawrence Estuary, run along the south shore to form the Gaspé Current, whereas the north shore of the LSLE is affected by upwellings that bring deep waters of the Gulf of St. Lawrence to the surface. The Gulf is an epicontinental sea open to North Atlantic water inputs by two straits: the Cabot Strait between Cape Breton and Newfoundland, and the Strait of Belle Isle between Labrador and Newfoundland. The Northeastern shore of the Gulf, called “Moyenne Côte Nord” (Figure 1), is under the hydrographical influences of upwellings (especially near the Anticosti Gyre), freshwater inputs from large rivers draining the Canadian Shield, and Labrador surface waters inflowing the Gulf westward through the Strait of Belle Isle. The estuarine waters of the LSLE that spread along the Gaspé Peninsula are diluted by more salty waters from the Anticosti Gyre (Figure 1). The shores of the Baie des Chaleurs, the Northumberland Strait, and the west coast of Cape Breton are characterized by relatively shallow waters and warmer water temperatures than in the northern part of the Gulf. In addition, the surface waters of the Baie des Chaleurs are impacted by river inputs, mainly from the Restigouche River, located at the western end of the Bay. A geographical partition of the Estuary and Gulf of St. Lawrence has been proposed based on ecological criteria [27]. These authors concluded that the most biologically significant hydrological features of the system are the LSLE, the Gaspé Current that hugs the coast of the Gaspé Peninsula, and the Northwestern Gulf, i.e., the Anticosti Gyre whose waters intercept the Moyenne Côte Nord west of Anticosti Island. These findings are supported by fluorescence distributions in the surface waters of the St. Lawrence Estuary and Gulf [28]. Based on the above-listed hydrological and ecological criteria, we decided to distinguish the following six regions: (1) North shore of the LSLE, (2) Moyenne Côte Nord, (3) South shore of the LSLE, (4) Gaspé Peninsula, and (5) Baie des Chaleurs, and (6) Northumberland-Cape Breton continuum.

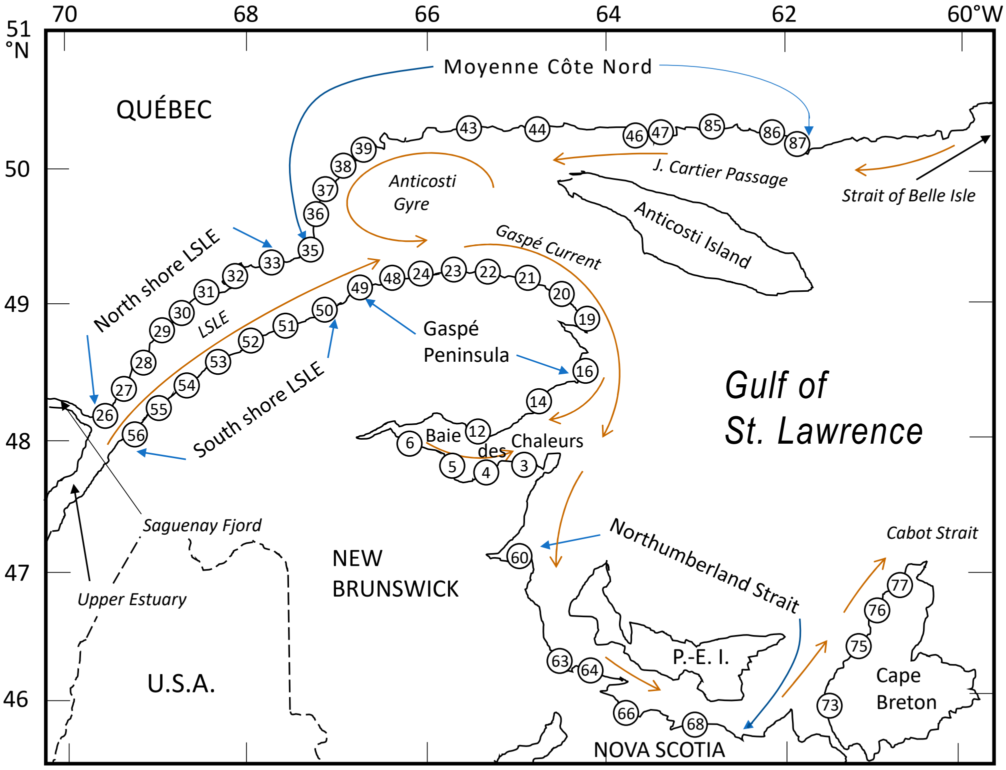

Figure 1. Location of the sampling sites and hydro-ecological regions. LSLE: Lower St. Lawrence Estuary. P.-E.I. Prince Edward Island. Brown arrows indicate main surface water circulation.

4. Results and Discussion

4.1. Geographical Patterns of the Growth

Total shell length (L), width (l), height (h), and tissue mass (M) measurements on mussel allow us to calculate allometric growth indices, that may vary depending on the *Mytilus* species and growth conditions [29,30]. In the St. Lawrence Estuary and Gulf, L, l, and h are linearly correlated, whereas L and M are better related by power functions [17]. These findings are confirmed by the present data (Table 1, Figure 2). In this study (2016–2019), allometric growth indices (h/L, l/L ratios, and the parameters of the L vs M power functions) were similar to the earlier sampling (1977–1979) (Table 1). This similarity in the mussel morphometry suggests that the possible environmental changes in the St Lawrence Estuary and Gulf [31] during the last decades, such as temperature, did not generate shell shape differences despite the high plasticity of the mussel shell [32]. Nevertheless, the parameters of the L vs M equations, derived for each of the six regions, display some variations (Figure 2, Table 1). Such slight regional morphological shell differences are consistent with the small regional differences in the absolute growth rate estimated in 1977–1979 [17]. We can infer from these allometric index comparisons that mussel growth conditions have not differed significantly for the last forty years, with slight regional differences in the L *vs* M index persisting (Table 1). This finding allows us to compare the Hg load of the mussel soft tissues between the two time periods without any significant bias affecting Hg bioaccumulation due to the change of mussel growth rates.

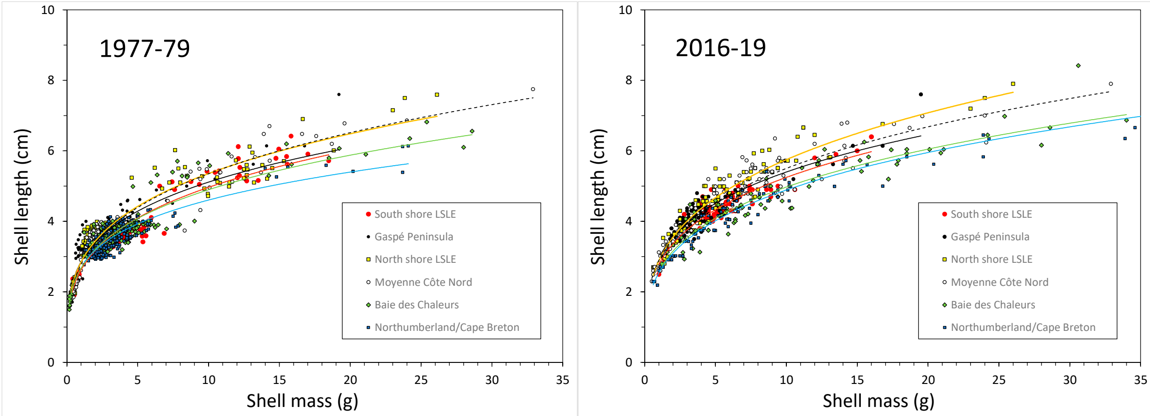

Figure 2. Length-mass relationship of *Mytilus edulis* shell from six hydro-ecological regions of the St. Lawrence Estuary and Gulf (Figure 1). Mussels sampled in 1977–1979 (n = 1222) and in 2016–2019 (n = 504) periods.

Table 1. Allometric relationships in the mussel shell shape and their changes between 1977–1979 and 2016–2019 periods. L: length (cm), l: width (cm); h: height (cm); M: mass (g). All the relationships were statistically significant with a $p < 0.001$. AGR: Absolute growth rate (cm y^{-1}) from ref. [17]. 1977–1979 statistics were calculated on 1222 individual mussels vs. 504 in 2016–2019.

Period	h/L	l/L	L vs M	AGR
		South shore LSLE		
1977–1979	0.52 ± 0.03	0.40 ± 0.07	$L = 2.62\ M^{0.28}$	0.57
2016–2019	0.49 ± 0.03	0.42 ± 0.04	$L = 2.92\ M^{0.25}$	–
		Gaspé Peninsula		
1977–1979	0.50 ± 0.05	0.42 ± 0.03	$L = 2.85\ M^{0.25}$	–
2016–2019	0.49 ± 0.05	0.41 ± 0.04	$L = 2.97\ M^{0.26}$	–
		North shore LSLE		
1977–1979	0.50 ± 0.05	0.42 ± 0.03	$L = 2.85\ M^{0.25}$	0.52–0.60
2016–2019	0.51 ± 0.03	0.42 ± 0.07	$L = 2.88\ M^{0.29}$	
		Moyenne Côte Nord		
1977–1979	0.52 ± 0.04	0.42 ± 0.05	$L = 2.78\ M^{0.28}$	0.57–0.65
2016–2019	0.52 ± 0.04	0.42 ± 0.03	$L = 2.89\ M^{0.28}$	–
		Baie des Chaleurs		
1977–1979	0.52 ± 0.04	0.42 ± 0.05	$L = 2.67\ M^{0.26}$	0.56–0.67
2016–2019	0.56 ± 0.04	0.43 ± 0.04	$L = 2.60\ M^{0.28}$	–
		Northumberland/Cape Breton		
1977–1979	0.57 ± 0.06	0.42 ± 0.05	$L = 2.72\ M^{0.23}$	0.40–0.57
2016–2019	0.57 ± 0.06	0.42 ± 0.04	$L = 2.54\ M^{0.28}$	–
		Estuary and Gulf of St Lawrence		
1977–1979	0.53 ± 0.06	0.43 ± 0.06	$L = 2.80\ M^{0.26}$	0.40–0.57
2016–2019	0.52 ± 0.07	0.43 ± 0.04	$L = 2.83\ M^{0.27}$	–

4.2. Estuary and Gulf of St. Lawrence Mussel Watch in the North Atlantic Context

The Hg concentrations in mussel soft tissues ranged from 0.06 to 0.51 µg g^{-1} dw (n = 504), with a grand mean of 0.17 ± 0.08 µg g^{-1} dw (± 1 standard deviation), and a median of 0.16 µg g^{-1} dw. In 1977–1979, the Hg concentrations were very similar with a mean of 0.16 ± 0.05 µg g^{-1} dw, and a median of 0.15 µg g^{-1} dw (n = 442) [33]. Very few Mussel Watch programs have been carried out uninterrupted for longer than a few years with the notable exception of the US and French monitoring programs that have been running yearly since the 1980s [34,35]. The median concentration in the St. Lawrence system is slightly higher than the most recent available medians: 0.11 µg g^{-1} dw (2005–2012, n = 298) for the US Mussel Watch and 0.12 µg g^{-1} dw (2000–2004, n = 303) for the French Mussel Watch programs [34,35]. It is similar to the annual median concentrations of the French coasts

Mussel Watch for the period 1980–94 (0.15 µg g^{-1} dw, n = 937) [36,37]. In summary, the current median Hg concentrations of the St. Lawrence Mussel Watch is similar to the long-term representative surveys carried out along the Atlantic shores in the last 40 years.

4.3. Temporal and Regional Hg Trends

To reach an optimal discriminating capacity for detecting temporal and regional trends, we used a normalization procedure minimizing bias due to biological factors (see above). The relationships between the Hg content of the 504 analyzed mussels and the dry mass of their soft tissue (TW) or shell length (L) were highly significant ($p < 0.001$). The Equation used for the normalization model built with the entire sampling set is:

$$\log Hg = 1.39 \pm 0.15 \log L + 0.49 \pm 0.04 \log TW - 0.36 \ (R^2 = 0.87; n = 504) \quad (1)$$

compared to:

$$\log Hg = 1.43 \pm 0.49 \log L + 0.34 \pm 0.16 \log TW - 0.48 \ (R^2 = 0.91; n = 143) \quad (2)$$

obtained for the St Lawrence Mussel Watch performed in 1977–1979 [1]. Regression coefficients, obtained forty years apart, are not statistically different ($p < 0.01$).

Normalized Hg content (individual of 35 mm length and 0.17 g dry weight) varied from 10.9 to 66.6 ng per individual, with a grand mean of 42.0 ± 2.1 ng (±1 standard deviation), and median of 44.2 ng, for the 504 individuals. Expressed as Hg concentration in the soft tissue, the range becomes 0.06–0.39 µg g^{-1} (dw). A similar normalization procedure applied to the 1977–1979 data, gave a very similar range: 12 to 64 ng per individual mussel [1] for the same stations. This finding strongly suggests that the Hg bioavailable to marine mussels in the study area has not changed over 40 years. At first glance, these observations are surprising since a decrease in Hg inputs to the St. Lawrence Estuary and Gulf waters could be expected (see below).

First, riverine Hg inputs to the St. Lawrence Estuary should have decreased due to the implementation of the International regulation on the Great Lakes basin [38–40]. Few time-series observations of Hg inputs from riverine sources, but a high-frequency sampling experiment of water was performed over 18 months in 1995–96 in the St. Lawrence River. Mercury export to the LSLE was estimated at ~1.2 Mg y^{-1}, for a mean dissolved Hg concentration of 0.60 ± 0.46 ng L^{-1} [41]. Since that period, no systematic study has been published on temporal variations of Hg concentrations in the St. Lawrence River. According to a recent government report, the Hg flux to the St. Lawrence River has not changed between 1995–1996 and 2004–2008 [42]. A comparison of mussel mean Hg contents at stations from the head of the LSLE (Stations 53–56), where freshwater influence is maximum, fails to show a statistically significant difference ($p < 0.01$) in Hg tissue levels between 1977–1979 (44.6 ± 6.9 ng, n = 5) and 2016–2019 (37.4 ± 6.6 ng, n = 5). It could be interpreted to imply that Hg bioavailability in freshwaters of the LSLE has not changed in the last 40 years.

Secondly, atmospheric Hg concentrations and wet deposition in Eastern North America are thought to have declined during the 1990–2010 period [43–45]. Regulations for reducing Hg emissions were implemented in New England and Eastern Canada [46]. In an assessment of Hg sources and fate in a marine environment very close to the Gulf of St Lawrence (The Gulf of Maine), Sunderland et al. [47] reported that: "Temporal patterns in sentinel species (mussels and birds) have in some cases declined in response to localized point source mercury reductions but overall Hg trends do not show consistent declines". Likewise, studies of coastal Massachusetts, New Hampshire, and Maine, reveal that no significant temporal trends in mussel Hg concentrations were found between 1990 and 2010 at 12 of the 15 monitored stations [19]. Our results are also consistent with those of Hg trends in herring gull eggs from Atlantic Canada collected between 1972 and 2008 [48]. These authors reported that, after adjusting Hg trends for dietary shifts, environmental Hg in coastal ecosystems had remained relatively constant in Eastern Canada over the previous 36 years. According to Sunderland et al. [43], reductions in atmospheric Hg deposition from North American sources could have been offset by increased deposition from global Hg sources.

Figure 3 illustrates the station-to-station variability in mussel tissue concentration. Strikingly, the continuum on the South shore of the LSLE- Gaspé Peninsula (Stations 56 to 14) exhibits a gradual Hg decrease from the brackish estuarine water originating from the St Lawrence Estuary to the Gaspesian coast as they are diluted by the Gulf waters originating from the Anticosti Gyre. This pattern strongly suggests that St. Lawrence River waters are a significant source of bioavailable Hg. Seaward, along the Gaspé Current, mussels exhibit very low Hg contents and variability (Stations 48, 49, 16 to 24, Figures 1 and 3). This would imply that Gaspé Current waters are impoverished in bioavailable Hg compared to waters of the LSLE. Increasing Hg reduction and evasion in the atmosphere in the productive waters (Anticosti Gyre and Gaspé Current, see Section 3) may favor this Hg depletion. It should be noted that the Gaspesian coastline is also an area where riverine inputs are small compared to other LSLE and Gulf shores. Another Hg dilution structure is visible along the Baie des Chaleurs (Stations 6 to 3, Figures 1 and 3) where riverine inputs are also important (see Section 3). Interesting to note is that, during the 1077–79 Mussel Watch, high Hg contents in mussel soft tissue were also observed at stations along the southern shores of the LSLE and of the Baie des Chaleurs where brackish waters are present (see Figure 4 in Ref. [1]).

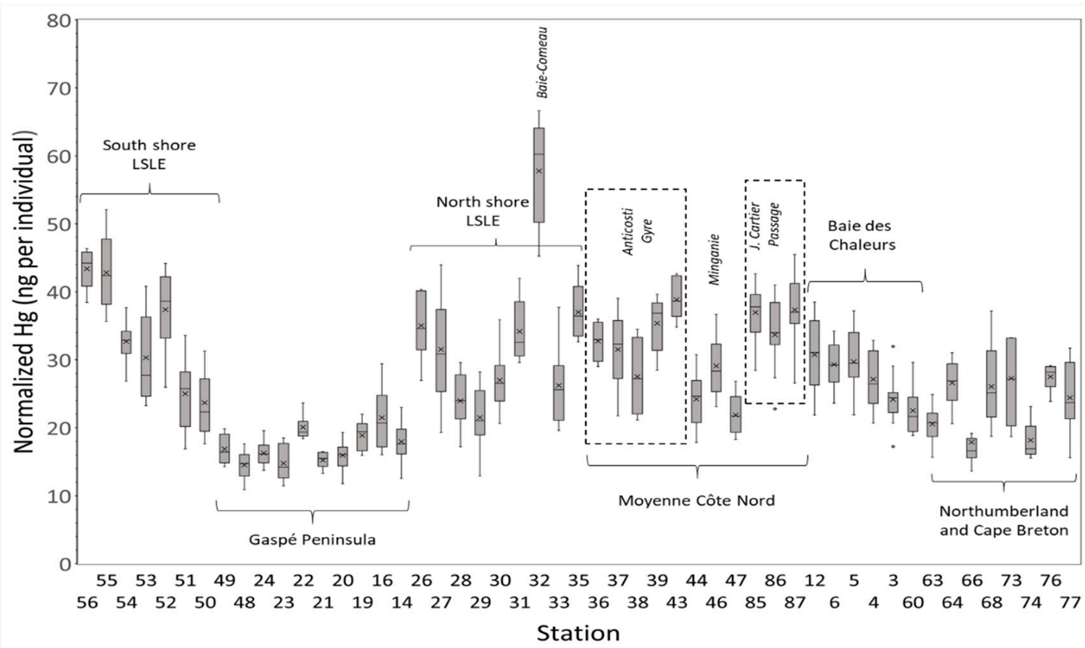

Figure 3. Mercury content (ng) of individual mussels after normalization to 35 mm-long and 0.17 g virtual specimens. Mean contents are indicated by a horizontal bar and the median by a cross. Each station combines 10 to 12 individual mussels. Stations are eastward distributed on the x-axis.

The North Shore of the LSLE and the Moyenne Côte Nord coastlines display inter-station large variations in mussel tissue Hg content, even within the same oceanographic entity (e.g., Moyenne Côte Nord, Figures 1 and 3). These fluctuating distributions are difficult to interpret since no steady geographical trend can be observed in Hg content. Nevertheless, some hypotheses can be put forward to explain these. Mercury content in the coastal waters of the North Shore of the St. Lawrence Estuary and Gulf are most likely influenced by upwellings and organic-rich river waters that drain through the Canadian Shield soils [49,50]. The organic matter of these circumneutral pH waters contains strong Hg-binding functional groups (e.g., thiol) that favor Hg transport in solution [41,51–53]. On the other hand, upwellings bring saline low Hg waters of Atlantic origin to the surface, especially in the Minganie region (Stations 44–47, Figures 1 and 3) [54]. These Atlantic waters enter the Gulf through

Cabot Strait and are known to contain low picomolar Hg levels [55]. Conversely, waters entering the Northeastern Gulf through the Strait of Belle Isle originate from the Labrador Sea Current, known for its relative high Hg concentrations due to the inputs of the organic-rich rivers outflowing the Arctic Canadian Archipelago [56]. These waters, brought by the Labrador Sea Current, are transported westward along the J. Cartier Passage, a remote region without any known trace-metal sources but with relatively high Hg content in sampled mussels (Figures 1 and 3).

It is interesting to note that low Hg content mussels are located along the Gaspé Peninsula (Stations 48, 49, 16–24) and Minganie (Stations 44–47) coasts, where commercial or experimental mussel farms are installed (mapaq.gouv.qc.ca/fr/Peche/aquaculture) [57,58]. This latter area is known for its low-temperature upwellings in summer [54].

4.4. Current and Former Hot Spots

Industrial areas distributed along the Estuary and Gulf coastlines are likely to shelter polluted sites. These would include a former chlor-alkali plant, pulp and paper mills, and smelters [33]. The highest Hg concentrations of the present survey were found at Station 32 near the city of Baie Comeau. The city hosts an active industrial complex and the bay sediments are enriched in trace metals from current and legacy sources [59,60]. Our study fails to highlight other potential hotspots that had been identified by previous Mussel Watch Programs. A chlor-alkali plant located near the mouth of the Restigouche River in the Baie des Chaleurs has been shown to generate local Zn and Hg contamination [61], but its impact on mussel tissue Hg contents was not revealed by our study. This might be explained by may be the result of the 20-km distance between Station 6 and the plant and/or the fact that it was shut down in 2008 [62]. Likewise, the previously reported Hg contamination [20] at the mouth of the Saguenay Fjord (Station 26), is no longer being observed, as the main Hg source, the Arvida chlor-alkali plant was shut down 40 years ago [63].

5. Summary and Conclusions

A Hg Mussel Watch Program was carried out at 52 stations distributed along the intertidal zone of the Estuary and Gulf of St. Lawrence, in the summers of 2016, 2018, and 2019. This survey took place forty years after a previous Mussel Watch survey was performed in this area (1977–1979), at almost all stations currently sampled. The same normalization procedure applied 40 years ago was also applied to the current data set. It allows the minimization of the effects of biological factors on the Hg content of the mussel soft tissues and optimizes the interpretation of Hg distribution in terms of the availability of Hg in coastal waters. Current Hg concentrations in the soft tissue of mussels from the St. Lawrence System are similar to those of the perennial Mussel Watch Programs implemented along the US and French Atlantic coasts [34,35]. The results indicate that the normalized Hg contents of mussels in the Estuary and the Gulf of St Lawrence were similar at sampling times 40 years apart. In addition, sub-regional Hg distribution patterns indicate a gradual decrease of Hg content in mussels downstream from freshwater inputs, which suggests that the spatial distribution of Hg concentrations in the soft tissues of marine mussels is, in part, governed by riverine Hg sources. Atmospheric Hg deposition and concentration in marine waters of the Atlantic Ocean are known to have decreased in the last decades [44,64]. However, in coastal environments, the response to these changes does not seem to be rapid. This probably results from the long residence time of Hg in soil and land cover before being exported to coastal areas with freshwater discharges. Indeed, different environmental reservoirs have different time scales as regards Hg mobility [65,66]. Furthermore, coastal sediment resuspension/sedimentation cycling may also contribute to a longer retention time of Hg in the intertidal marine environments, making mussel habitats conducive to the retention of legacy Hg. The changes in atmospheric Hg inputs are thus damped in terms of Hg availability for sessile benthic animals. A renewed Mussel Watch survey in a few decades may elucidate the long-time trend of Hg load in the St Lawrence Coastal System.

Author Contributions: D.C. designed the experiment, performed sampling and analyses, and wrote the article. A.-M.T. participated in sampling and sample preparation. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare that there is no conflict of interest regarding the publication of this paper.

Appendix A

Table A1. Station coordinates. See also map in Figure 1. Mean normalized Hg content of (±1 SD) in ng per individual.

Station	Name	Longitude (W)	Latitude (N)	Hg Content
South shore of the LSLE/Gaspé Peninsula				
56	Rivière-Trois-Pistoles	69°13′18.1″	48°06′22.3″	43.4 ± 2.8
55	St-Simon-sur-Mer	69°03′39.4″	48°13′10.5″	41.9 ± 5.9
54	Cap du Corbeau (Bic)	68°43′38.2″	48°22′44.6″	32.7 ± 3.1
53	Anse aux coques (St Luce)	68°22′55.8″	48°33′05.3″	30.3 ± 6.4
52	Pointe-Mitis	68°01′55.5″	48°40′52.4″	37.4 ± 6.0
51	Petite rivière Blanche (St Ulric)	67°41′31.6″	48°47′36.5″	25.0 ± 5.2
50	Anse à la Croix	67°18′00.7″	48°54′27.1″	23.7 ± 4.7
49	Baie des Capucins	66°50′37.8″	49°03′09.4″	16.8 ± 2.2
48	Anse de l’Eglise (Tourelle)	66°25′12.6″	49°09′27.6″	14.5 ± 2.2
24	Cap à la Martre	66°10′19.0″	49°12′26.7″	16.3 ± 1.8
23	Anse de Mont-St-Pierre	65°47′32.0″	49°13′40.0″	14.8 ± 2.5
22	Rivière-la-Madeleine	65°17′45.0″	49°14′30.0″	20.1 ± 1.9
21	Pointe-à-la-Frégate	64°56′25.6″	49°12′28.0″	15.2 ± 1.1
20	Pointe Jaune	64°30′17.3″	49°04′07.2″	16.0 ± 2.2
19	Cap des Rosiers	64°12′08.0″	48°51′21.0″	18.9 ± 2.2
16	Rocher Percé	64°12′27.7″	48°31′30.8″	21.5 ± 4.5
North shore of the LSLE				
26	Pointe aux Vaches (Tadoussac)	69°41′18.9″	48°08′08.9″	35.0 ± 4.9
27	Grandes-Bergeronnes (Marina)	69°33′09.9″	48°13′42.9″	31.5 ± 7.9
28	Baie St Onge (Les Escoumins)	69°23′48.5″	48°21′03.2″	23.9 ± 3.8
29	Pointe Verte (Forestville)	69°03′13.4″	48°44′21.9″	21.5 ± 4.6
30	Anse du Cap Colombier	68°52′50.6″	48°49′23.5″	27.0 ± 4.9
31	Baie Verte (Ragueneau)	68°34′33.6″	49°03′38.7″	34.2 ± 4.8
32	Baie-Comeau	68°08′25.6″	49°13′12.2″	57.8 ± 8.1
33	Franquelin	67°53′46.0″	49°17′28.7″	26.2 ± 6.0
Moyenne Côte Nord				
35	Pointe des Monts	67°22′14.6″	49°19′30.5″	37.0 ± 4.3
36	Pointe aux Anglais	67°12′09.0″	49°38′14.1″	32.8 ± 3.0
37	Anse au Pot	67°08′57.1″	49°48′04.0″	31.5 ± 5.4
38	La Grande Anse	66°56′56.1″	49°59′00.3″	27.5 ± 5.8
39	Parc Île Paterson (Port Cartier)	66°52′12.8″	50°01′05.3″	35.4 ± 4.0
43	Rivière Pigou	65°30′37.9″	50°16′59.6″	38.8 ± 3.0
44	Rivière-au-Tonnerre	64°45′42.0″	50°16′22.4″	24.2 ± 3.9
46	Île Quarry	63°50′11.8″	50°12′28.3″	29.1 ± 4.4
47	Havre-St-Pierre	63°36′08.5″	50°14′12.5″	21.9 ± 2.7
85	Baie Johan Beetz	62°48′26.9″	50°17′10.1″	37.0 ± 3.9
86	Île Michon	62°01′53.9″	50°13′21.8″	33.7 ± 5.2
87	Le Petit Havre (Natashquan)	61°50′26.6″	50°11′19.9″	37.4 ± 5.4

Table A1. *Cont.*

Station	Name	Longitude (W)	Latitude (N)	Hg Content
		Baie des Chaleurs		
14	Point Newport	64°43′42.6″	48°17′08.6″	18.0 ± 3.0
12	Pointe Bonaventure	65°27′33.1″	48°00′25.8″	30.8 ± 5.8
6	Nash Creek	66°07′48.7″	45°55′08.3″	29.3 ± 3.2
5	Petit-Rocher	65°42′37.0″	47°47′02.6″	29.7 ± 4.2
4	Stonehaven	65°21′43.5″	47°45′14.6″	27.1 ± 4.4
3	Anse-Bleue	65°06′13.4″	47°49′46.7″	24.2 ± 3.9
		Northumberland/Cape Breton		
60	Escuminac	64°54′47.2″	47°04′39.7″	22.5 ± 3.8
63	Cap Bimet	64°27′31.9″	46°14′14.8″	20.6 ± 2.8
64	Cap Pelé (Trois-Ruisseaux)	64°13′35.2″	46°13′16.6″	26.6 ± 3.3
66	Wild Rose (Linden)	63°47′02.3″	45°53′17.8″	17.9 ± 4.8
68	Horn Point (Seafoam)	63°00′30.3″	45°47′40.0″	26.1 ± 5.7
73	Little Judique Ponds	61°30′44.9″	45°54′28.3″	27.3 ± 6.1
75	Whale Cove (Margaree)	61°08′18.2″	46°25′28.2″	18.2 ± 2.6
76	Pointe Enragée (Chéticamp)	61°01′37.9″	46°38′58.2″	27.5 ± 1.9
77A	Pleasant Bay (Harbour)	60°47′52.0″	46°49′53.2″	24.4 ± 5.1
77B	MacKenzies River/Pleasant Bay	60°49′51.5″	46°49′25.0″	

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