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J. Thomas Sanderson, Academic Editor PeerJ

Dear Dr. Sanderson,

My co-authors and I would like to thank you for allowing us the opportunity to resubmit our manuscript entitled EVALUATION OF THE GLOBAL IMPACTS OF MITIGATION ON PERSISTENT, BIOACCUMULATIVE AND TOXIC POLLUTANTS IN MARINE FISH to PeerJ. We believe that the comments from the reviewers as well as your own comments have helped us to significantly improve this paper. We have responded to all comments in detail below and have updated our manuscript accordingly.

Here we quantify and evaluate patterns of published concentrations of five classes of persistent pollutants in marine fish globally: mercury, chlordanes, DDTs, PBDEs, and PCBs. The goals of our study were to collate all published, peer reviewed data to: a) synthesize global concentration levels and distribution of PBTs, b) investigate the extent of regional, trophic, or habitat-related behavior patterning in data, and c) evaluate effectiveness of PBT mitigation efforts. **Our results provide the first global assessment of PBTs in the edible tissue of marine finfish, and we show that for all classes of pollutants reviewed, there is a significant global decline in concentration through time. We do see evidence for some regional patterning across the global landscape for CHLs and PCBs, and fairly consistent regional ranking of pollutant concentrations. Our review is timely as it highlights the need for continuous and**

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standardized pollutant monitoring to properly assess and manage PBT levels in the environment and elucidate patterns and trends more reliably.

We hope that you find our revisions satisfactory and we look forward to hearing from you soon.

Sincerely,

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Lindsay T. Bonito

General Comments:

We found the comments from both the Editor and Reviewer 1 to be very helpful and generally minor. In the attached manuscript, we have made a number of revisions in response, with details of these changes below.

The primary concerns of the Editor were related to our use of a linear regression to describe temporal decline in PBT concentrations. We completely agree that high variability exists in the data, and R2 values are low. We have challenged the data to explore and, in our perspective, to confirm the suitability of the linear model. Specific responses to Editor's concerns can be found below, including details regarding the statistical tests.

Reviewer 1 had useful comments regarding general pollutant transport and had good grasp of and insight into the out literature, from which we made numerous throughout the manuscript. Nearly all recommendations were translated into changes in the main text, particularly throughout the introduction and discussion sections. Note that the line numbers have been edited to reflect the correct numbering found in the revised version.

Response to Editor's Comments:

Editorial Comments:

This manuscript describes a detailed analysis of a large amount of environmental pollution data which provides information for the scientific and regulatory community. The data collection and statistical analyses are as thorough as can be expected given the experimental conditions. The results should be discussed more critically. **(1)** For example, the regression analyses of environmental levels over time (Fig 5) appear to be highly influenced by one may be two data point for their statistical significance. **(2)** Also, despite statistical significance, given the shallow slopes and huge variability, how much have levels actually gone down between 1960 and now? **(3)** Have levels in certain areas gone up (e.g. arctic – **(4)** it is also surprising that the investigators could not find arctic contamination data given the region has an active monitoring program-fish, seals, polar bears). **(5)** Also, why was linear regression used. What if levels initially increased and only recently started coming down? A U shaped trend is entirely possible given the cloudy nature of the data.

(6) Please redefine, replace or remove vague terms or jargon such as 'patterning', 'legacy', etc...as their meaning will not be apparent to all readers. Sentences are often convoluted, lack precision or are too vague.

e.g.: 'Mercury levels revealed the strongest patterns across trophic and habitat types.' or ' ..variability in pollutants..' or 'mean values of pollutants' What do these phrases mean?

(7) Tidy-up expressions such as: concentration levels (=concentrations or levels).

(8) Propositions should be corrected: at (time-scale) = on (or sometimes 'over'); through (time) = over. Propositions and other words are occasionally missing. Manuscript needs thorough editorial review.

Author Response:

We thank the editor for the positive comments and will respond directly to what he defines as the limitations of our study as follows. **(1)** In reviewing Figure 5 and the associated linear regression, we believe that no single or pair of data points are driving the signal we see in the regression. There are hundreds of points in each plot, with a substantial amount of scatter. Furthermore, we performed the linear regression using only a subset of data (years 1990-2012) where the bulk of the data lie, to see if the signal changed. This regression on the more recent data alone can be found in Supplemental Table 6. All the regressions, with the exception of DDT (p-value 0.62) returned similar slopes and statistical significance (p-values). Finally, when performing a linear regression with "cloudy data", the value of the Mean Squares for Error (variance of residuals) increases in denominator of the F-statistic calculation, thereby increasing the chance of committing a Type II error (failing to reject a null hypothesis that is actually false). Seeing a signal in the cloudy data confirms that the signal is rather robust. **(2)** It is important to note that Figure 5 is displayed in a logarithmic scale, making the slopes seem shallow upon first inspection. However, the proportional decline in concentrations ranges from 15-30% across PBT groups per decade.

(3) Every region, and nearly every PBT group, showed declined in concentrations through time (1960-2012). Additional supplemental figure was created to look at the linear regression within regions (Supplemental Figure 3). (4) Although there is a large amount of studies monitoring POP trends in the Arctic, a significant portion sample only homeotherms. Of the fish species sampled, it is predominantly Arctic cod. These data were collected when possible. (5) A linear regression was used to answer the simple question of decline through time. Importantly, due to the scatter in the data, tests of more subtle non-linear trends will lack statistical power. We opt for the most simple linear model (which, based on the data transformation describes logarithmic decline) as the most straightforward and parsimonious way to describe the directional trend in the data. Again, further support for the simple linear model comes as we see the same decreasing linear trend when shifting the time period window. Similar declines are seen whether we look at the whole time period (1960-2012) or the recent window (1990-2012). We have revised the wording and grammar throughout the manuscript to address the remainder of the editorial comments (6-8).

REVIEWER #1

Basic Reporting

Reviewer Comments:

In this paper a large dataset consisting of 2662 measurements of persistent organic pollutants (POPs) and mercury in marine fish from 303 papers were assembled. The data were then evaluated statistically to assess broad spatial and temporal trends as well as effects of habitat use and trophic level. These goals were quite ambitious and somewhat unrealistic given that data had to be aggregated from many

species and geographic areas (eg the Atlantic Ocean is one region), there being almost no global studies of a single marine fish species. The only nearly global study that this reviewer has been able to find are the papers by Ueno et al on POPs in skipjack tuna (one is cited).

Author Response:

We appreciate the Reviewer acknowledging the large dataset and associated difficulties in assessing the data. Although the paper does take on some assumptions and generalities (eg treating the Atlantic Ocean as one region), we remained transparent in our methods as to not deceive readers of the complicated nature of the dataset.

Experimental design

Reviewer Comments:

Despite this limitation the study largely succeeds in addressing these goals although the wide ranges of concentrations within and across regions limit the ability to demonstrate the importance of factors such as trophic level and habitat. The assembling of such a large dataset is unique and potentially useful if only to draw attention to the limited ability to assess global trends of pollutants which have undergone (eg PCBs, chlordane) or are undergoing (eg mercury) global bans or emission reductions. In this regard, the study, perhaps inadvertently, also demonstrates that studies focussed on spatial and temporal trends in the same species yield much more statistically powerful trends. Meta analyses of such studies (eg Riget et al (2010; 2011) for POPs and mercury) which included several datasets for marine fish provide more definitive results. Also Domingo & Bocio'sreview PCDD/Fs and PCBs in marine fish used a meta-data and assessment type of approach (Environ Int 2007). However both reviews were somewhat limited geographically given the preponderance of contaminants data from Europe, North America and the Arctic. **(1)** The decision not to include contaminants trends in Baltic Sea was surprising considering that very strong datasets are available eg for herring (reported in HELCOM contaminant assessments). **(2)** The authors also fail to note that assessment of global trends of POPs is a goal of the Stockholm Convention's global monitoring program for effectiveness evaluation

(http://chm.pops.int/Implementation/GlobalMonitoringPlan/Overview/tabid/83/Default.aspx) although admittedly it is not targeted at marine fish.

Author Response:

We are happy to see that the Reviewer felt the study succeeded in addressing the research questions and managed to see the ultimate goal of the manuscript – to draw attention to the limited ability to assess global trends of pollutants. (1) We do agree that a well-established monitoring program in Baltic Sea (HELCOM) is well reported in both HELCOM assessments as well as in the peer-reviewed literature. Because of this, we didn't want to duplicate and re-report data from the region. However, the Baltic data extracted in our own dataset was included in the global temporal evaluation (linear regression – Figure 5). (2) The Reviewer is correct in noting we did not explicitly say that the Stockholm Convention does have a goal of evaluating global POP trends. We have added in text in the introduction to resolve this oversight (lines 72-73).

Validity of the findings *Reviewer Comments:*

The weakest aspects of the paper are the sweeping statementts about the knowledge of emissions, sources and biogeochemistry of the contaminants. The authors appear to be unfamiliar with some of the literature on global distribution and modelling of POPs and mercury. **(1)** For example on line 47 it is stated that "estimates of global emissions and secondary sources of PBTs are poor or non-existent". This is not the case for PCBs (see Brevik et al Environ Poll 2004; Sci Total Environ 2002) or for mercury (eg Mason et al (Environ Res 2012), Pacyna et al Atmos Environ 2010). Li et al (Environ Monit Assess 2005; other papers) have also estimated global inventories and emissions of chlorinated pesticides. The suggestion (line 76) that " no study has attempted to resolve the question of spatial patterning across ocean basins" ignores the studies that have examined the oceanic distribution of POPs eg. Iwata et al ES&T 1993; Lohmann et al ES&T 2001; JGR 2006) and many global modelling studies which have included ocean waters (eg Lammel & Stemmler Atmos Chem Phys 2012).

Author Response:

We acknowledge that the general overview provided in the introduction does include some broad and sweeping statements. As this study isn't focused directly on the inputs and circulation of pollutants, rather, how they accumulate in marine fish and patterns within this more narrow scope, we chose to spend less time delving into the biogeochemistry of the contaminants. Furthermore, Reviewer 1 should note that the statement **(1)** "estimates of global emissions and secondary sources of PBTs are poor or non-existent" continued to say "prior to 2000" (line 18). The references listed by Reviewer 1 do make valiant attempt to create mass budget estimates of PCBs, yet themselves acknowledge "For legacy and emerging persistent organic pollutants (POPs), surprisingly little is still known in quantitative terms about their global sources and emissions...Similarly, our current understanding of POPs' global transport and fate remains sketchy" (Lohmann et al Environmental Pollution, 2007). That being said, we have edited the introduction to remove some of these statements and include references acknowledging these studies (lines 17-18).

Comments for the author (Line by line)

- Reviewer Comment: Line 7-8 (formerly line 40). These references that support the statement that a "significant proportion of the world's population is exposed to PBTs" are very USA centric. Yet there are many more globally oriented papers (eg for mercury see Chen et al EHP 2008; Environ Res 2012) and Domingo &Bocio (Environ Int 2007) for PCDD/Fs and PCBs.
 Author Response: We thank the Reviewer for noticing the limited scope of the references here and providing some further reading. Domingo & Bocio 2007 has been added to the introduction; however the Chen references were excluded as they focus on air or water exposure, rather than fish consumption.
- Reviewer Comment: Line 42 (formerly line 73). "rapidly volatilize" is not a correct term for describing the behavior of POPs such as most PCBs, DDT and chlordane compounds. Normally they are described as semi-volatile with the ability to be sorbed on surface and to revolatilize. Author Response: We have changed the wording of the sentence to be more accurate in the description of the general behavior of POPs.

- Reviewer Comment: Line 51-52 (formerly line 82-83). Awkward sentence, seems to be combining POPs and mercury meaning is unclear.
 Author Response: We have edited this sentence to be less convoluted to readers.
- 4. **Reviewer Comment:** Line 60 (formerly line 91). The authors need to be more specific here re "patterns among marine taxa" because there are many examples of biomagnification of POPs and mercury in marine food webs but perhaps far less if homeotherms are left out of the calculation eg. Borga et al ET&C 2004.

Author Response: We agree that there are numerous examples of biomagnification in marine food webs, particularly in webs that include homeotherms. However, we were specifically interested in studies that investigated transfer within a fish-dominated web, fish to fish transfer excluding homeotherms. Far fewer of these studies exist, and results are generally non-conclusive. However, the reviewer's comments were taken into consideration and a revision in the manuscript has been made. Furthermore, Borga, 2004 was a useful addition to references in the introduction.

5. **Reviewer Comment:** Line 116 (formerly line 147). The "ICES 7" congeners are not the ones "commonly reported in toxicological studies" but rather are the ones used for monitoring PCBs in foods (for example). The co-planar PCBs would be examples of ones reported in toxicological studies.

Author Response: We have made a change in the manuscript to include this detail.

6. **Reviewer Comment:** Line 235 (formerly line 266). *Shouldn't results be limited to 2 or 3 significant figures given the known uncertainty or imprecision of measurement, especially of POPs.*

Author Response: We have changed the significant digits throughout the results in the main text and Table 1 to reflect 1-2 significant digits.

- 7. Reviewer Comment: Line 254 (formerly line 285). Why not use ng/g everywhere rather than "ppb" to be consistent with concentration data?
 Author Response: We have made changes in this paragraph to keep consistent units (ng/g) throughout the text.
- 8. **Reviewer Comment:** Line 405-407 (formerly line 436-438). The sentence implies that the Stockholm Convention has been successful because of the observed decline "over the past 50 years". The SC only came into force in 2004 so cannot be credited with this decline, which is obviously due to national regulatory actions starting in the 1970s. Also it should be made clear that this is for POPs. Similarly global declines in mercury are attributable to national or regional actions.

Author Response: We agree with the comment and have made the appropriate changes in the manuscript.

9. Reviewer Comment: Line 490. Missing information for this reference

Author Response: Due to the mysterious line number issue, we are unsure which reference the Reviewer is commenting on...