# HISTORICAL TRENDS OF DIOXINS AND FURANS IN SEDIMENTS AND FISH FROM THE GREAT LAKES

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## OBJECTIVES

- Provide a method to evaluate historical concentrations of dioxin in fish from the Great Lakes basins
- Investigate historical variations of the congener specific contribution to the TEQ

## INTRODUCTION

A significant fraction of body burdens of dioxin is thought to remain in individuals as a consequence of long half-lives in the human body and past elevated environmental levels. This work provides a trend specifically calibrated for reflecting historical trends in fish from the Great Lakes. In particular, we derive a mathematical function describing the trend of the concentrations of 2,3,7,8-TCDD in these water basins. Until potential future availability of congener, region, and species-specific concentration data, we suggest application of the provided historical functions calibrated for a particular species concentration at a certain time, as a basis for calculating historical trends of dioxins and furans in fish species from the Great Lakes.

## DATA AND METHODS

- Concentrations of 2,3,7,8-TCDD in multiple fishes species and trout eggs from the Great Lakes and the Tittabawassee River over various periods of time have been assembled and normalized using 1978 as a reference year (fig. 1).[1-2-3]
- Trends of total PCDDs and PCDFs in sediment from the beginning of the 20th century and up to 1970 (ascending part) from Lake Michigan, Lake Huron, and Lake Ontario[4] were rescaled using the value in 1978 as a reference value and used for the final regression.
- A least square regression for all data (1935-2002 range) was performed using an altered version of the equation previously suggested by Pinsky and Lorber[5](bell shaped).
- Changes of relative contributions of 16 congeners to the TEQ (WHO2005 weighted TEFs) in trout[6] and from an assembly of human cohorts[7] have been investigated.

## RESULTS

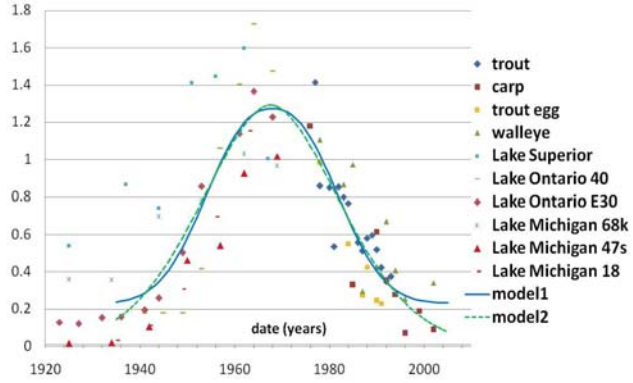


**Figure 1.** Historical trend for models with all parameters fitted without constraints (model 1) and forced to tend towards 0 (model 2).

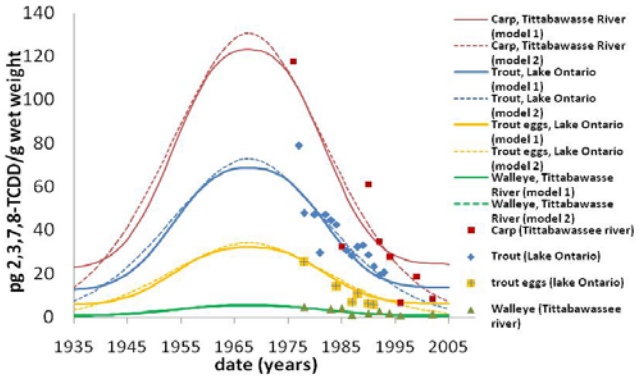


**Figure 2.** Predicted fish and trout eggs historical concentrations in several Lake Ontario and Tittabawassee River using original data from Huestis et al.[1] and Woodburn et al.[2]

**Table 1.** Congener specific parameters estimates and p values for the % change in WHO2005-TEQ contribution per year between 1977 and 1991. The last row provides the estimates for the total 2005-WHOTEQ variation/year during the period.

| | Lake Ontario Trout[a] | | U.S. Pop. blood[b] | | | Lake Ontario Trout[a] | | U.S. Pop. blood[b] | |
|---|---|---|---|---|---|---|---|---|---|
| Congener | $\beta_{\%}$ ($CI_{95\%}$) | p value | $\beta_{\%}$ ($CI_{95\%}$) | p value | Congener | $\beta_{\%}$ ($CI_{95\%}$) | p value | $\beta_{\%}$ ($CI_{95\%}$) | p value |
| 2,3,7,8-TCDD | -8.5 (-11.6,-5.4) x $10^{-3}$ | <0.001 | -4.4 (-8.0,-0.8) x $10^{-3}$ | 0.022 | 2,3,4,7,8-PeCDF | 2.1 (0.6,3.5) x $10^{-3}$ | 0.010 | -2.9 (-6,0.1) x $10^{-3}$ | 0.057 |
| 1,2,3,7,8-PeCDD | 3.2 (1.8,4.6) x $10^{-3}$ | <0.001 | 6.9 (-0.2,14.0) x $10^{-3}$ | 0.056 | 1,2,3,4,7,8-HxCDF | -1.0 (-6.9,4.9) x $10^{-4}$ | 0.715 | -5.7 (-,-) x $10^{-7}$ | 0.999 |
| 1,2,3,4,7,8-HxCDD | | n/a | | | 1,2,3,6,7,8-HxCDF | 3.5 (2.2,4.7) x $10^{-4}$ | <0.001 | 4.8 (-1.6,11.2) x $10^{-4}$ | 0.127 |
| 1,2,3,6,7,8-HxCDD | 1.8 (0.8,2.9) x $10^{-4}$ | 0.002 | -1.2 (-6.4,4.1) x $10^{-3}$ | 0.634 | 1,2,3,7,8,9-HxCDF | | n/a | | |
| 1,2,3,7,8,9-HxCDD | 8.5 (3.4,13.5) x $10^{-5}$ | 0.003 | 1.3 (0.2,2.4) x $10^{-3}$ | 0.024 | 2,3,4,6,7,8-HxCDF | 1.3 (0.7,1.9) x $10^{-4}$ | 0.001 | 3 (1.0,5.1) x $10^{-4}$ | 0.010 |
| 1,2,3,4,6,7,8-HpCDD | 9.0 (-1.3,19.3) x $10^{-6}$ | 0.081 | -2.5 (-15.7,10.7) x $10^{-4}$ | 0.686 | 1,2,3,4,6,7,8-HpCDF | 1.1 (0.1,2.1) x $10^{-5}$ | 0.040 | 1.1 (-1.5,3.6) x $10^{-4}$ | 0.374 |
| OCDD | -7.8 (-35.8,20.1) x $10^{-7}$ | 0.556 | 2.1 (-0.2,4.5) x $10^{-4}$ | 0.070 | 1,2,3,4,7,8,9-HpCDF | | n/a | | |
| 2,3,7,8-TCDF | 2.4 (0.6,4.1) x $10^{-3}$ | 0.013 | 4.4 (3.0,5.8) x $10^{-4}$ | <0.001 | OCDF | 9 (-3.7,21.7) x $10^{-7}$ | 0.150 | 4.2 (-0.3,8.7) x $10^{-6}$ | 0.061 |
| 1,2,3,7,8-PeCDF | 1.6 (0.9,2.4) x $10^{-4}$ | <0.001 | 1.2 (0.7,1.6) x $10^{-4}$ | 0.001 | 2005-WHOTEQ | -3.1 (-4.5,-1.7) | <0.001 | -2.5 (-3.6,-1.4) | <0.001 |

[a]Huestis et al.

[b]Dioxin reassessment report

The best fits for both models lead to similar peak dates (1968) and analogous overall shapes except for the ancient and recent times where a divergence is observed. As shown in table 1, most of the congeners exhibit the same tendency over time in trout and in human blood. For example 2,3,7,8-TCDD is shown to decrease at a non statistically distinguishable rate in both sample types.

## CONCLUSIONS

- The fitted trend function represented in figure 1 is consistent with reports from the literature
- The absolute concentrations widely differ between fish species, but the trend relative to a reference year is similar
- Additional data on recent concentrations is needed to fully assess recent trends
- The analysis of congener specific trends suggests that contribution of some of the most important congeners for the TEQ were varying over time

[1] Huestis, S. Y.; Servos, M. R.; Whittle, D. M.; Van Den Heuveli, M.; Dixon, D. G., Environmental Toxicology and Chemistry 16, 154 1997. [2] Woodburn, K.; Budinsky, R.; Blankenship, A., Organohalogen Compounds, 57 2003. [3] Cook, P. M.; Robbins, J. A.; Endicott, D. D.; Lodge, K. B.; Guiney, P. D.; Walker, M. K.; Zabel, E. W.; Peterson, R. E., Environmental Science and Technology 37, 3864 2003. [4] Pearson, R. F.; Swackhamer, D. L.; Eisenreich, S. J.; Long, D. T., Environmental Science and Technology 31, 2903 1997. [5] Pinsky, P. F.; Lorber, M. N., Journal of Exposure Analysis and Environmental Epidemiology 8, 187, 1998. [6]Huestis, S. Y.; Servos, M. R.; Whittle, D. M.; Van Den Heuveli, M.; Dixon, D. G., Environmental Toxicology and Chemistry 16, 154 1997. [7]United States Environmental Protection Agency (USEPA). Exposure and Human Health Reassessment of 2,3,7,8-Tetrachlorodibenzo-p-Dioxin and Related Compounds. Draft Final, EPA/600/P-00/001Be, National Center for Environmental Assessment, US Environmental Protection Agency: Washington, DC, 2000. [8]sad

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