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Anticoagulant Rodenticides in fish: Spatial and Temporal Distribution in German Freshwater Aquatic Systems

MATTHIAS KOTTHOFF [1], HEINZ RÜDEL[1], MARK BÜCKING[1], ANTON FRIESEN[2], JAN KOSCHORREK[2]

[1] Fraunhofer Institute for Molecular Biology and Applied Ecology (Fraunhofer IME), 57392 Schmallenberg, Germany, matthias.kotthoff@ime.fraunhofer.de
[2] German Environment Agency (Umweltbundesamt), 06844 Dessau-Rosslau, Germany

Anticoagulant rodenticides (AR) are effective poisons used for rodent control. Active substances of marketed second-generation anticoagulant rodenticides have been identified as bioaccumulative, persistent and toxic (PBT) substances in regulatory approval process for biocides. Knowledge about the exposure of the environment including food webs is needed but few data are available, especially for aquatic systems. To fill this gap, we analysed liver samples of bream (*Abramis brama*) and suspended particulate matter (SPM) from the German Environmental Specimen Bank (ESB). An appropriate method was developed for the determination of eight different ARs (Flocoumafen, Bromadiolon, Brodifacoum, Difenacoum, Warfarin, Chlorophacinone, Coumatetralyl, and Difethialon) in fish muscle and liver comprising both, first- and second-generation anticoagulant rodenticides. The final protocol applied liquid chromatographic separation coupled with high-resolution mass spectrometric detection in tandem mode and yielded a limit of quantification (LOQ) ranging in a substance and matrix specific manner from 0.2 to 2 µg kg^{-1} wet weight. Applying the final method to bream liver samples from 17 or 18 sampling locations of the years 2011 or 2015, respectively, five out of eight ARs were found at levels above LOQ. For 2015, brodifacoum was detected in 88 % of the samples with a maximum concentration of 12.5 µg kg^{-1}. Moreover, difenacoum, bromadiolone, difethialon, and flocoumafen were detected in some samples above LOQ. In contrast, the first-generation AR warfarin, coumatetralyl, and chlorophacinone were not detected in the ESB fish samples. To our knowledge, this is the first evidence for AR in fish from wildlife.
In contrast to fish samples, only bromadiolone could be detected in 56 % of the SPM samples at levels up to 9.24 µg $kg^{-1.}$ No other AR was found in SPM samples from the 16 ESB sampling locations.
A temporal trend analysis of bream liver from two sampling locations over a period of up to 23 years revealed no significant trend for the ARs detected in this study.
Spatial exposure analysis of 17 fish samples from 2011 and 2015 indicate two hot spots showing high levels and a comparatively broad occurrence of AR in the upper Elbe region in eastern Germany and the Saar region in Germany's west.
This study provides for the importance of further monitoring of AR in freshwater aquatic systems and food webs.

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