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# Novel two-tiered approach of ecological risk assessment for pesticide mixtures based on joint effects

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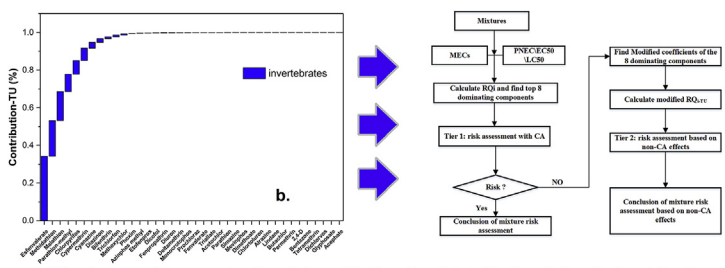
## HIGHLIGHTS

- A novel approach was proposed to assess the mixture risks based on joint effects.
- Top 8 dominating components explained 95.5%–99.8% of mixture risks in this study.
- The algae is the most sensitive in the three tropical levels.

## GRAPHICAL ABSTRACT




Novel approach to assessing the mixture risks based on joint effects and chemical contributions

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## ABSTRACT

Ecological risk assessments for mixtures have attracted considerable attention. In this study, 38 pesticides in the real environment were taken as objects and their toxicities to different organisms from three trophic levels were employed to assess the ecological risk of the mixture. The first tier assessment was based on the CA effect and the obtained sum of risk quotients ($SRQ_{species\text{-}CA}$) were 3.06–9.22. The second tier assessment was based on non-CA effects and the calculated $SRQ_{species\text{-}TU}$ are 5.37–9.29 using joint effects ($TU_{sum}$) as modified coefficients, which is higher than $SRQ_{species\text{-}CA}$ and indicates that ignoring joint effects might run the risk of underestimating the actual impact of pesticide mixtures. Due to the influences of synergistic and antagonistic effects, risk contribution of components to mixture risks based on non-CA effects are different from those based on the CA effect. Moreover, it was found that the top 8 dominating components explained 95.5%–99.8% of mixture risks in this study. The dominating components are similar in the two tiers for a given species. Accordingly, a novel two-tiered approach was proposed to assess the ecological risks of mixtures based on joint effects. This study provides new insights for ecological risk assessments with the consideration of joint effects of components in the pesticide mixtures.

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## 1. Introduction

Pesticides are widely used in the world, especially in the United States and China. Various pesticides are usually used simultaneously in order to strengthen their effectiveness. These pesticides residues are readily gathered through the surface runoff and mixed

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together in the surface water. Kreuger (1998) found that streams in Sweden contained between 10 and 22 pesticide residues. Kolpin et al. (2002) investigated the contamination of 139 rivers in the US and found one third of the rivers contained 10 or more different pollutants. As pesticides are usually present as chemical mixtures, mixture risk assessments are indispensable for environmentally actual risk assessments.

Most current research has assessed risks of mixtures based on concentration additive (CA) method, that is, the risk of a mixture can be obtained by summing up all risks of components in the mixture (Bundschuh et al., 2014). However, the behavior of components in a mixture may not always correspond to CA effects (Silva et al., 2002). Various researches have reported non-CA effects for a wide range of mixtures including sulfonylurea herbicides (Junghans et al., 2003a), chloroacetanilide herbicides (Junghans et al., 2003b), atrazine and organophosphate insecticides (Pape-Lindstrom and Lydy, 1997), cyanazine and chlorpyrifos (Lydy and Linck, 2003), triazine herbicides and organophosphate insecticides (Schuler et al., 2005; Belden et al., 2007). Cedergreen et al. found only 10% of 158 pesticide mixtures could be predicted by CA concept and synergy occurred in 7% of the 194 binary pesticide mixtures (Cedergreen et al., 2008; Nørgaard and Cedergreen, 2010; Cedergreen, 2014). Although most of these studies focused on binary mixtures and multicomponent mixtures were seldom involved, it can be inferred that a multicomponent mixture might yield non-CA effect when the joint effect of two dominating components is synergy or antagonism based on a variation rule of joint effects (Tian et al., 2012; Li et al., 2014). Consequently, we should take joint effects of pesticide components into account in mixture risk assessments (Białk-Bielińska et al., 2017).

Few methods can be used to assess the ecological risks for mixtures. Backhaus et al. proposed a tiered strategy for risk assessments of pharmaceuticals mixtures based on the CA and IA (Independent Action) concept (Backhaus and Faust, 2012; Backhaus and Karlsson, 2014). They suggested applying CA as a precautious first tier for a conservative risk assessment of mixtures and IA as a second tier risk estimate for a potential risk. However, such a sequential method relies on a classification of components into two groups of similar and dissimilar mechanisms of action. Ma et al. also predicted the mixture toxicity of coking wastewater based on the CA concept (Ma et al., 2017). These previous studies took additional effects (including CA and IA) as reference models and the method is not applicable for mixtures with non-addition effects (including synergistic and antagonistic effects). Therefore, it still remains unclear how to assessment the risks for mixtures with non-CA

**Table 1**
Single toxicities of 38 pesticides to algae, invertebrates and fish ($EC_{50}$ or $LC_{50}$, $mg \cdot L^{-1}$).

| NO. | Pesticides | Algae | Invertebrates (*Chironomus/Daphnia*) | Fish |
|---|---|---|---|---|
| 1 | Prochloraz | 0.24[1] | 0.041[2] | 1.68[b] |
| 2 | Lindane | 4.22[1] | 1.93[a] | 0.11[b] |
| 3 | Parathion | 7.86[1], 3.88[3] | 0.039[4] | 2.80[4] |
| 4 | Chlorotoluron | 0.023[1] | 67[a] | 6[b] |
| 5 | Simazine | 0.073[1] | 1[5] | 12.6[b] |
| 6 | Triallate | 3.87[1] | 1.23[6] | 1.8[b] |
| 7 | Bentazone | 42.5[1] | 34.4[7] | 600[b] |
| 8 | 2,4-D | 88.9[1] | 236[8] | 270[9] |
| 9 | Glyphosate | 377[1], 7.25[10] | 673.4[a] | 2.9[b] |
| 10 | Azinphos methyl | 1.67[b] | 0.00032[5] | 3.20[b] |
| 11 | Chlorpyrifos | 0.521[11], 6.34[10] | 0.00017[5], 0.0053[12], 0.00016[13] | 0.00154[12], 0.0872[12], 0.2[13] |
| 12 | Cyanazine | 0.0159[14] | 1[5] | 1.3[b] |
| 13 | Diazinon | 10.82[b] | 0.019[5] | 45[b] |
| 14 | Diuron | 0.0108[b] | 1[5] | 6.3[b] |
| 15 | Methidathion | 21.95[b] | 0.00123[5] | 6.8[b] |
| 16 | Esfenvalerate | 0.00083[b] | 0.00021[13] | 0.00044[13] |
| 17 | Atrazine | 0.0147[15] | 20[16] | 1.8[b] |
| 18 | Machette | 2.31[15] | 2.4[b] | 1.69[b] |
| 19 | Dicofol | 0.454[11] | 1.4[a] | 0.90[b] |
| 20 | Acetochlor | 0.0856[11] | 8.4[b] | 1.20[b] |
| 21 | Malathion | 0.019[16] | 0.0322[4] | 0.016[b] |
| 22 | Methoxychlor | 0.0033[16] | 0.1[a] | 0.038[b] |
| 23 | Parathion-methyl | 0.0323[16] | 0.0069[4] | 0.0015[4] |
| 24 | Mevinphos | 0.0088[16] | 0.18[17] | 0.30[b] |
| 25 | Chlorophos | 0.0899[16], 59.53[10] | 0.01[a] | 32.66[18] |
| 26 | Cypermethrin | 112.45[b] | 0.0026[a] | 2.61[19] |
| 27 | Phenvalerate | 0.0017[b] | 0.5[a] | 3.71[19] |
| 28 | Fenpropathrin | 0.03[20] | 0.014 [b] | 0.51[19], 3.314[18] |
| 29 | Deltamethrin | 2.56 [b] | 0.3[a] | 0.34[19] |
| 30 | Phoxim | 6.62[10], 0.77 [3], | 0.019[b] | 5.34[19] |
| 31 | Acephate | 50[b] | 54[21] | 5.44[19] |
| 32 | Tetramethrin | 1.12[22] | 0.47[a] | 0.046[23] |
| 33 | Bifenthrin | 0.00034[b] | 0.0052[a] | 0.0032[23] |
| 34 | Ethofenprox | 0.053[b] | 0.011[b] | 0.0791[23] |
| 35 | Dichlorvos | 82.12[10], 2.88[3] | 17.6[24] | 13[23] |
| 36 | Permethrin | 5[b] | 0.4[a] | 0.0025[23] |
| 37 | Dimethoate | 141.37[10] | 5.04[4] | 4.1 [4] |
| 38 | Monocrotophos | 0.1[3] | 0.161[25] | 6.5[b] |

[1](Faust et al., 1994), [2](Dalhoff et al., 2016), [3](Xie et al., 1999), [4](Lei et al., 2013), [5](Lydy and Austin, 2004), [6](Buhl and Faerber, 1989), [7](Overmyer, 2009), [8](Oris et al., 1991), [9](Neškovid et al., 1994), [10](Wang et al., 2012), [11](Chen et al., 2010), [12](Tyler Mehler et al., 2008), [13](Belden and Lydy, 2006), [14](Faust et al., 2001), [15](He et al., 2012), [16](Pape-Lindstrom and Lydy, 1997), [17](Mayer and Ellersieck, 1986), [18](Tang, 2012), [19](Zhao et al., 2008), [20](Shoaib et al., 2011), [21](Printes and Callaghan, 2004), [22](Chen et al., 1997), [23](Zhang et al., 2010), [24](Ankley and Collyard, 1995), [25](Wang et al., 2008).

[a] http://www.pesticideinfo.org/.

[b] http://cfpub.epa.gov/ecotox/.

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