Quantitative modelling of human health risks arising from pesticide use in Irish agriculture

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ABSTRACT: Pesticides have been identified as a growing concern due to the risk they pose to human health. Risk models can be used to predict pesticide concentrations in drinking water and analyse the associated health risks and hence allow for informed decision making and risk management. However, the majority of pesticide models currently in use are deterministic in nature, and do not reflect the inherent uncertainties and variabilities in pesticide transmission. Probabilistic-based approaches offer a powerful tool in this context as they can account for uncertainty and variability through the use of statistical input parameters to produce distributions of pesticide concentrations and health risks. This study develops a model which enables probabilistic assessment of risk posed to human health from pesticide runoff into water supplies. A Monte-Carlo-based approach was applied to enable the analysis of the probabilities of exceedance of chronic exposure levels to humans arising from the modelled transport of pesticides to drinking water. This model is applied to an Irish case study to allow for the identification of pesticides that could pose significant risks to drinking water. This may inform the implementation of risk mitigation measures to reduce pesticide pollution at source and/or identification of less harmful pesticides for use.

1. INTRODUCTION
Pesticides play a vital role in food production by improving crop yields (Carvalho, 2017). Approximately 2.7 million tonnes of pesticides are used globally each year (FAO, 2023), however it is estimated that only 1% to 10% of pesticides reach target organisms (Ali et al., 2019). Pesticides have been detected in groundwater and surface water with a growing number of studies highlighting the negative impact pesticides have...
on the environment and human health (Thompson et al., 2020). Guidelines have been put in place by governments to regulate pesticide use and address the risks posed by pesticides. These guidelines layout a framework for pesticide regulation consisting of risk-screening and detailed risk assessment to reduce potential harm to human health due to contaminated water (Fargnoli et al., 2019).

The risk assessment stage requires an estimate of the likelihood of exposure using transport models and an analysis of the consequence of this exposure. Many models, such as SWAT and PRZM, have been developed to model the transport of pesticides and are widely applied in research and regulation (Kolupaeva et al., 2022). However, such models tend to be deterministic and use point estimates or average values as inputs to calculate a single concentration for risk analysis. Therefore, the majority of pesticide risk assessments fail to fully consider the uncertainty and variability associated with real-world data and pesticide risk (Troldborg et al., 2022).

The European Food Safety Authority (EFSA) has suggested that it is critical that probabilistic approaches are applied to generate more meaningful results (EFSA and BfR, 2019). Probabilistic modelling has become more common in assessing the risk posed by pesticide residues in food (Kennedy et al., 2020), but are relatively limited when assessing risk via drinking water. The data-hungry and complex computational nature of many existing models limits the suitability of probabilistic approaches. Therefore existing studies either apply probabilistic distributions to one or two key parameters in complex transport models (Tasdighi et al., 2018) or have adapted simpler models for probabilistic modelling (Troldborg et al., 2022).

This study aims to develop a framework for the quantitative assessment of human health risk arising from contamination of surface water by pesticides. An existing empirical runoff model will be modified to improve on existing studies and a probabilistic approach will be applied to account for uncertainty and variability. This framework offers a relatively user-friendly probabilistic approach, which may be used by catchment and risk managers to make more risk-informed decisions when discussing pesticide selection and regulation.

2. METHODOLOGY

The model involves two stages: a) Section 2.1 describes a probabilistic model based on existing literature used to predict the concentration of pesticides in water due to runoff, b) Section 2.2 used the predicted pesticide concentrations to assess the level of risk from consumption of contaminated water using a risk quotient method. Having developed the modelling framework, Section 2.3 discusses how Monte Carlo methods were used to incorporate uncertainty and variability into the analysis.

2.1. Predicted Environmental Concentration

The model used in this study is a modified ‘Simplified Formula for Indirect loading caused by runoff’ (SFIL) from the OECD (2000) used to calculate pesticide concentration in surface water. This model was selected due to its simplicity and applicability while still considering a range of influencing factors a process involved in pesticide transport. The formula outlined by the OECD estimated the percentage of pesticide that will be lost to runoff (L%):

$$L\% = \frac{Q}{P} \times e^{\frac{3\ln 2}{DT_{50s}} \times x f_1 x f_2 x f_3}$$  \hspace{1cm} (1)

where Q is the runoff volume (mm) and P is the daily average rainfall (mm), DT_{50s} is the pesticide half-life in soil (days), K_d is the ratio of dissolved to sorbed pesticide:

$$K_d = \frac{K_{oc} \times OC}{100}$$  \hspace{1cm} (2)

where K_{oc} is the adsorption coefficient (l/kg), and OC is the soil organic carbon content (%).

Previously runoff volume (Q) was obtained from methods described by Lutz and Maniak (1984; 1992), which were only available for two soil types: sandy and loamy, and three site
scenarios: 1. Bare soil (i.e. pre-emergence) with high moisture content, 2. Bare soil with low moisture, and 3. Covered soil (i.e. crops in growth stages) with low moisture. Therefore, the SFIL was unable to realistically represent the various site combinations that exist in reality. To address this limitation, the US Soil Conservation Service Curve Number method, as used in more complex models such as PRZM and SWAT, was applied in this study. This novel approach was applied in order to provide more site-specific estimations of runoff volumes by considering soil texture, land use, and growth stage. A curve number was given to the site based on a specific hydrologic soil-cover complex, a combination of hydrologic soil group land use and crop cover type (USDA, 2004).

This method assumes that runoff will not occur until precipitation exceeds 20% of the soil’s maximum potential retention, and if this level is reached the resulting runoff volume was calculated as follows:

\[ Q = \frac{(P - 0.25)^2}{(P + 0.85)} \]  

where S is the maximum potential retention (mm), and was calculated using the site curve number:

\[ S = \frac{25400}{CN} - 254 \]

The correction factors \( f_1 \), \( f_2 \), \( f_3 \) were applied to account for site conditions and agricultural practices. Correction factor \( f_1 \) was used to account for the effects of site slope on potential runoff, whereby \( f_1 \) for slopes greater than 20% was assumed to be 1 and for slopes less than 20% it was calculated as follows:

\[ f_1 = 0.02153 \text{slope} + 0.001423 \text{slope}^2 \]

The reduction of pesticide loss due to the distance to waterbody was considered using the correction factor \( f_2 \):

\[ f_2 = 0.83^z \]

where \( z \) (m) is the distance to waterbody.

The amount of applied pesticide available for runoff after crop interception was included using factor \( f_3 \), whereby:

\[ f_3 = 1 - \frac{\text{PI}}{100} \]

where PI is the plant interception factor (%), which is based on the type of crop and its growth stage, developed by FOCUS (2002).

In order to obtain the predicted environmental concentration in surface water based on percentage loss, the modified SFIL described by Berenzen et al. (2005) was then applied.

2.2. Human Health Risk

Estimated daily intake of pesticides was calculated based on FAO and WHO guidelines (FAO and WHO, 1997):

\[ EDI = \frac{\text{PEC} \times \text{WC}}{\text{BW} \times 1000} \]

where EDI is the estimated daily intake (mg/kg/day), PEC is the predicted environmental concentration (µg/l), WC is daily water consumption (l/day), BW is the body weight (kg) and 1000 is a conversion factor to convert micrograms to milligrams.

A risk quotient method was used to determine the risk posed by the intake of pesticide. This was selected as it is widely used in literature and is recommended for risk assessments by the EU and WHO. The risk quotient for chronic exposure was calculated as a simple ratio of EDI to acceptable daily intake (ADI) whereby the ADI (mg/kg/day) is the level of exposure at which no adverse effects are expected.

If the risk quotient is greater than one, the risk associated with the level of pesticide exposure was deemed unacceptable and adverse health effects are expected to occur.

2.3. Probabilistic Modelling Method

A Monte-Carlo technique was utilised due to its applicability, reliability and simplicity for probabilistic assessment and uncertainty analysis. The statistical distribution of outputs is useful in a health risk study as it allows for the quantitative assessment of risks posed by low-level, median, and extreme exposure to pesticides in a population. The model in this study was
developed from first principles using MATLAB®, and the Monte-Carlo simulation was run for 1,000,000 iterations to ensure statistical stability of the model outputs. Statistical parameters were developed for several input parameters based on distributions selected from best-fit analysis and a review of distributions used in literature. The statistical parameters selected for the model are discussed in Section 3.

3. CASE STUDY

An Irish case study is presented to demonstrate model implementation. A hypothetical site was developed to represent an “average” Irish agricultural scenario. The site conditions were selected based on an analysis of the EPA’s Irish Soils Information Systems, stream flow database and agricultural reports (Cawkwell et al., 2017; EPA, 2021a, b), and climate conditions were established from Met Éireann data (2022). A grassland site, with soil conditions typical of hydrologic group C was then used for model simulations. The statistical parameters for this site are presented in Table 1.

The distributions of Irish adult and child body weight and water consumption rates were obtained from the National Adult Nutrition Survey and the National Children’s Food Survey II respectively (IUNA, 2011, 2021). The statistical parameters for the population data are presented in Table 2.

Pesticide selection was based on annual use obtained from the Irish Department of Agriculture’s National Crop Surveys (DAFM, 2016, 2017) and an initial risk screening process (Harmon O’Driscoll et al., 2022). The five pesticides selected make up over 30% of overall annual pesticide use. Relevant pesticide properties for the modelling process were obtained from EFSA reports which are presented in Table 3.

Table 1: Site Properties and Statistical Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Distribution</th>
<th>Statistical Parameters</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Precipitation</td>
<td>mm/day</td>
<td>Gamma</td>
<td>a = 0.255, b = 10.457</td>
<td>(Met Éireann, 2022)</td>
</tr>
<tr>
<td>Slope</td>
<td>%</td>
<td>Fixed</td>
<td>3</td>
<td>(Clarke et al., 2016)</td>
</tr>
<tr>
<td>z</td>
<td>m</td>
<td>Fixed</td>
<td>0</td>
<td>(Berenzen et al., 2005)</td>
</tr>
<tr>
<td>PI</td>
<td>%</td>
<td>Uniform</td>
<td>Min = 0, Max = 70</td>
<td>(FOCUS, 2015)</td>
</tr>
<tr>
<td>OC</td>
<td>%</td>
<td>Normal</td>
<td>µ = 2.36, σ = 2.79</td>
<td>(Fay et al., 2007; EPA, 2021b)</td>
</tr>
<tr>
<td>Q_stream</td>
<td>l/s</td>
<td>Lognormal</td>
<td>µ = 78.0, σ = 16.5</td>
<td>(WMO, 1989; EPA, 2021a)</td>
</tr>
</tbody>
</table>

Table 2: Population Data and Statistical Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Distribution</th>
<th>Statistical Parameters</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adult Body Weight</td>
<td>kg</td>
<td>Normal</td>
<td>µ = 78.0, σ = 16.5</td>
<td>(IUNA, 2011)</td>
</tr>
<tr>
<td>Child Body Weight</td>
<td>kg</td>
<td>Normal</td>
<td>µ = 32.5, σ = 11.4</td>
<td>(IUNA, 2021)</td>
</tr>
<tr>
<td>Adult Water Intake*</td>
<td>l/day</td>
<td>Lognormal</td>
<td>µ = 1.2, σ = 0.68</td>
<td>(USEPA, 2004; IUNA, 2011)</td>
</tr>
<tr>
<td>Child Water Intake*</td>
<td>l/day</td>
<td>Lognormal</td>
<td>µ = 0.5, σ = 0.32</td>
<td>(USEPA, 2004; IUNA, 2021)</td>
</tr>
</tbody>
</table>

* Includes water-based drinks such as tea and coffee
Table 3: Pesticide Properties and Statistical Parameters

<table>
<thead>
<tr>
<th>Pesticide</th>
<th>$K_{OC}$ (l/kg)</th>
<th>DT$_{50,5}$ (days)</th>
<th>$P_a$ (g/ha)</th>
<th>ADI (mg/kg/day)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glyphosate</td>
<td>Normal: $\mu = 14396.7$, $\sigma = 15102.9$</td>
<td>Normal: $\mu = 45.1$, $\sigma = 93.9$</td>
<td>Fixed: 2160</td>
<td>Fixed: 0.5</td>
<td>(EFSA, 2021)</td>
</tr>
<tr>
<td>MCPA</td>
<td>Normal: $\mu = 75.2$, $\sigma = 42.2$</td>
<td>Normal: $\mu = 21.4$, $\sigma = 42.7$</td>
<td>Fixed: 1800</td>
<td>Fixed: 0.05</td>
<td></td>
</tr>
<tr>
<td>Fluroxypyr</td>
<td>Normal: $\mu = 68$, $\sigma = 10.9$</td>
<td>Normal: $\mu = 19.6$, $\sigma = 15.2$</td>
<td>Fixed: 400</td>
<td>Fixed: 0.8</td>
<td></td>
</tr>
<tr>
<td>Mecoprop</td>
<td>Normal: $\mu = 56.3$, $\sigma = 56.3$</td>
<td>Normal: $\mu = 6.4$, $\sigma = 10.1$</td>
<td>Fixed: 1200</td>
<td>Fixed: 0.01</td>
<td></td>
</tr>
<tr>
<td>2,4-D</td>
<td>Normal: $\mu = 41.7$, $\sigma = 18.6$</td>
<td>Normal: $\mu = 18.8$, $\sigma = 25.5$</td>
<td>Fixed: 750</td>
<td>Fixed: 0.05</td>
<td></td>
</tr>
</tbody>
</table>

4. RESULTS AND DISCUSSION

The risk assessment proposed in this study allows for pesticide risk to be analysed and compared under a number of criteria. Pesticide exposure and resulting health risks are only predicted to occur when rainfall exceeds a retention level dependent on the site scenario. For the presented case study, daily rainfall of 8.3 mm is required for a runoff event. Based on historical rainfall data, it is predicted that such a rainfall event will occur 36 days each year on average. To allow for the examination and comparison of pesticide risk resulting from runoff, the model results in Section 0 and Section 4.2 are truncated to exclude zero-runoff rainfall events i.e. days with <8.3mm rainfall. Therefore the following results should be considered in the context that only 10% of model runs in this study result in runoff.

4.1. Predicted Environmental Concentration

The distribution of predicted environmental concentration for the modelled pesticides for days with runoff is presented in Figure 1. MCPA was predicted to occur in the highest concentrations followed by mecoprop. All pesticides selected for modelling, except glyphosate, have very low $K_{OC}$ and therefore are not likely to adsorb to the soil making them available for runoff and resulting in relatively high predicted concentrations. Conversely, the highly adsorbent glyphosate only exceeds the EU limit of 0.1 µg/l at the 80th percentile concentration. The modelled results have a good agreement with a monitoring programme of 14 pesticides in 144 rivers in Ireland over a five-year period which found that MCPA was the most widely detected pesticide in Irish rivers, followed by mecoprop and 2,4-D (EPA, 2019). The concentrations for MCPA, Mecoprop, and 2,4-D presented in Figure 1 are relatively high when compared to the legal limit of 0.1µg/l. However, as a successful runoff event only occurs in 10% of model runs, the likelihood of exposure to such levels is relatively low. In that context, MCPA’s 95th percentile concentration of 29.95 µg/l is predicted to occur less than twice a year.

4.2. Human Health Risk

The EDI and the RQs resulting from the 5th, median and 95th percentile daily intake of pesticide for Irish adults and children are presented in Table 4. No pesticide analysed in this study exceeded a risk quotient of one for any level of pesticide intake from drinking water. Therefore, the results indicate a low level of risk to human health. The level of risk posed to children is slightly higher than to adults as the rate of water intake is higher relative to body weight. According to the risk quotient, mecoprop has the highest risk level with a 95th percentile RQ of 0.045 in adults and 0.057 in children due to its comparatively high ADI (Table 3). These RQ values are still well below a RQ of one which suggests that there is negligible risk. In fact, for an adult or child of average weight and average daily water intake from Table 2, the environmental concentration of mecoprop would have to be 650
µg/l in order for the RQ to exceed an acceptable level. This level of exposure is extremely unlikely to occur under legal agricultural practices as this is almost 40 times mecoprop’s 95th percentile of predicted environmental concentration.

Despite the fact that the median predicted concentrations during runoff days for four of the five pesticides exceed the EU legal limit of 0.1 µg/l, none of the pesticides approach a level of concern for chronic human health risks. Dekant et al. (2010) have stated that the near zero pesticide limits were set with little consideration of a pesticide’s evaluated toxicological significance. The findings herein support this and could facilitate discussion around adjusting the EU legal limits for individual pesticides in place of a single limit for all pesticide irrespective of their toxicity, at least in the case of human health.

Figure 1: Distribution of predicted environmental concentrations for runoff days (Truncated at 5 µg/l for clarity).

Table 4: Summary of estimated daily intake (EDI) and risk quotient (RQ) for adult and child population

<table>
<thead>
<tr>
<th>Pesticide</th>
<th>5th</th>
<th>EDI_{Adult}</th>
<th>95th</th>
<th>5th</th>
<th>EDI_{Child}</th>
<th>95th</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glyphosate</td>
<td>8.0E-10</td>
<td>1.9E-7</td>
<td>1.2E-5</td>
<td>1.1E-9</td>
<td>2.4E-7</td>
<td>1.5E-5</td>
</tr>
<tr>
<td>MCPA</td>
<td>1.0E-7</td>
<td>2.0E-5</td>
<td>8.1E-4</td>
<td>1.3E-7</td>
<td>2.4E-6</td>
<td>1.0E-3</td>
</tr>
<tr>
<td>Fluroxypyr</td>
<td>2.4E-8</td>
<td>4.3E-6</td>
<td>1.8E-4</td>
<td>3.1E-8</td>
<td>2.4E-5</td>
<td>1.0E-3</td>
</tr>
<tr>
<td>2,4-D</td>
<td>5.8E-8</td>
<td>1.1E-5</td>
<td>4.3E-4</td>
<td>7.5E-8</td>
<td>1.3E-5</td>
<td>5.5E-4</td>
</tr>
<tr>
<td>Mecoprop</td>
<td>5.6E-8</td>
<td>1.1E-5</td>
<td>4.5E-4</td>
<td>7.1E-8</td>
<td>1.4E-5</td>
<td>5.7E-4</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Pesticide</th>
<th>5th</th>
<th>RQ_{Adult}</th>
<th>95th</th>
<th>5th</th>
<th>RQ_{Child}</th>
<th>95th</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glyphosate</td>
<td>1.8E-9</td>
<td>3.7E-7</td>
<td>2.4E-5</td>
<td>2.3E-9</td>
<td>4.8E-7</td>
<td>3.1E-5</td>
</tr>
<tr>
<td>MCPA</td>
<td>2.0E-6</td>
<td>3.9E-4</td>
<td>1.6E-2</td>
<td>2.6E-6</td>
<td>4.9E-4</td>
<td>2.0E-2</td>
</tr>
<tr>
<td>Fluroxypyr</td>
<td>3.1E-8</td>
<td>5.4E-6</td>
<td>2.2E-4</td>
<td>3.9E-8</td>
<td>6.8E-5</td>
<td>2.8E-4</td>
</tr>
<tr>
<td>2,4-D</td>
<td>1.2E-6</td>
<td>2.1E-4</td>
<td>8.9E-3</td>
<td>1.5E-6</td>
<td>2.7E-4</td>
<td>1.1E-2</td>
</tr>
<tr>
<td>Mecoprop</td>
<td>5.6E-6</td>
<td>1.1E-3</td>
<td>4.5E-2</td>
<td>7.1E-6</td>
<td>1.4E-3</td>
<td>5.7E-2</td>
</tr>
</tbody>
</table>
5. **CONCLUSION**

The present study has developed a probabilistic modelling framework and a case study to demonstrate that the agricultural use of pesticides can result in high levels of pesticides in drinking water. In this study, however, levels of pesticide exposure do not indicate that there is perceivable risk to human health at predicted concentrations.

This model provides a useful step in the use of probabilistic analysis for pesticide risk in drinking water. The model can be applied for pesticide assessment by a range of users such as catchment managers and farm advisors. Due to the relatively simplistic nature of the model, it can be adapted to include more pesticide processes such as portioning in water and hydrolysis, to assess different pesticide use patterns or to account for the impact climate change may have on pesticide risk.

6. **REFERENCES**


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IUNA, 2021. (Irish Universities Nutrition Alliance) National Children's Food Survey II.


