Article

Daily Intake Estimation for Young Children’s Ingestion of Residential Dust and Soils Contaminated with Chlorpyrifos and Cypermethrin in Taiwan

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Abstract: We estimated the daily intakes of chlorpyrifos and cypermethrin via ingestion of indoor dust and outdoor soils using the Stochastic Human Exposure and Dose Simulation Model on a probabilistic approach for Taiwan’s homes. Variable information for the daily intake estimation, such as concentration, ingestion rate, body weight, was adopted from previous studies. Monte Carlo simulation was performed with 1,000,000 iterations to simulate a single daily intake, which was shown in terms of percentage of the Acceptable Daily Intake (ADI) of either insecticide. The daily intakes were minimal with a 99% probability; at 99.9th percentile, however, the total intakes leaped to 13.1% and 20.0% of the respective ADIs of chlorpyrifos and cypermethrin. The sensitivity analysis indicated that concentration was the most determinant variable. Compared to the data of daily intakes via dietary ingestion of vegetables derived from a previous study, the estimated intakes by this study were considerable at the highest percentile, which referred to insecticide residues few days after insecticide application. Consequently, the non-dietary ingestion exposure to either insecticide was negligible in most cases; nevertheless, for those with indoor insecticide applications, the daily intakes for young children could be of concern. Frequently home cleaning is recommended to reduce the exposure.

Keywords: chlorpyrifos; cypermethrin; daily intake; home environment; Monte Carlo simulation; non-dietary ingestion; SHEDS model; Taiwan

1. Introduction

Taiwan is a perennially warm and humid island where pesticides are frequently used for agriculture and vector-borne disease control. A previous report indicated that Taiwan was among the top countries around the world in pesticide consumption [1]. With such substantial consumption in quantity, residues of insecticides present in residential environments are nearly inevitable. A pilot study conducted in Taiwan has demonstrated high detection rates of chlorpyrifos and cypermethrin in house dust, reflecting frequent use of insecticides in the homes [2]. Residues of insecticides in the home environment serve as a potential health threat to young children, because their hand-to-mouth behaviors may enhance the non-dietary ingestion exposure to insecticides [3].

Chlorpyrifos and cypermethrin are different types of insecticides (i.e., organophosphate, pyrethroid), but both serve as neurotoxic agents to kill bugs. A number of studies have indicated that prenatal exposure to organophosphate and/or pyrethroid insecticides is associated with children’s behavioral disorders or neurodevelopmental problems [4-14]; moreover, recent studies...
have found that postnatal childhood exposure to insecticides may have similar effects [15-18], suggesting young children’s susceptibility to insecticides. The childhood exposure may occur via dietary ingestion, such as consumption of fruits and vegetables contaminated with insecticides, and via non-dietary ingestion (e.g., by mouthing behaviors), which is of great concern should the environment be highly contaminated. A previous study conducting an exposure assessment of chlorpyrifos on residential surfaces for young children at 3 – 6 years old one week after insecticide application has found the dose via non-dietary ingestion to be 126 µg/kg/day [3]. Another study using a probabilistic modeling framework to estimate children’s non-dietary ingestion of chlorpyrifos has indicated the daily ingested chlorpyrifos being approximately 1,000 µg/day (median value) and slightly lower than 100 µg/day (median value) within and after the first week of application, respectively [19]. These estimates that show high doses few days after insecticide application suggest that non-dietary ingestion could be an important route of exposure to pesticides for children in the residential environments.

The modeling used in the previous study [19] is the Stochastic Human Exposure and Dose Simulation Model (SHEDS) for multiple pollutants, which is a probabilistic human-activity-based physical model developed by the U.S. Environmental Protection Agency (EPA) [20]. It is designed to estimate dust and soil ingestion exposure to pollutants, with application of videographic data of children’s everyday activities indoors and outdoors to indicate frequencies and types of contact with various surfaces, instead of using tracer-element-based mass balance models [21]. In addition, this strategy of modeling predicts full variability distributions of dust and soil ingestion rates for age-specific groups (e.g., preschool children), which are better than traditional single-point estimates. Coupled with Monte Carlo simulation, a method using repeated random sampling to generate simulated data, SHEDS could provide a decent estimate of non-dietary dust and soil ingestion exposure to insecticides in home environment for young children, with variability and uncertainty being considered.

Since Taiwan uses large quantities of pesticides every year, it is interesting to know residential exposure to insecticide residues particularly for young children. The previous studies that estimated non-dietary ingestion exposure to insecticides were all conducted short after application, mimicking worst-case scenarios. This study intended to assess the exposure on a regular basis, and to include variability and uncertainty for the outcome. Thus, we introduced the concentration profiles of chlorpyrifos and cypermethrin previously measured from residential environments by our research team [2] and the dust and soil ingestion rates for 3 – 6 year old children estimated by SHEDS [21] to Monte Carlo simulation to predict the non-dietary ingestion exposure to chlorpyrifos and cypermethrin for young children in the home environments of Taiwan. To our best knowledge, this is the first study that estimates residential exposure via non-dietary ingestion of insecticides for children in Taiwan.

2. Materials and Methods

The formula for ingestion of dust or soils is as follows:

\[
\text{Daily intake} = \frac{IR \times C \times CF}{BW} \quad (1)
\]

Where

- \(IR\): Ingestion rate of dust or soils (mg/day)
- \(C\): Concentration of insecticide in dust or soils (chlorpyrifos or cypermethrin) (µg/g)
- \(CF\): Conversion factor (10\(^{-3}\) g/mg)
- \(BW\): Body weight of children (kg)

Distributions of the above variables are listed in Table 1. Ingestion rates of dust or soils adopted herein were modeled specifically for 3 – 6 year old children using SHEDS, because of high...
frequency of mouthing behaviors and availability of adequate exposure data for this age group [21]. Concentrations of insecticides (chlorpyrifos, cypermethrin) for indoor dust and outdoor soils, provided by our previous work [2], could be used for calculation with ingestion rates of dust and soils, respectively. For those dust samples under limits of detection (LOD), halves of LODs were used in place for dose estimation. Children’s body weight data were adopted from the new growth charts suggested and developed for Taiwanese children and adolescents [22]. The dust and soil ingestion fits a lognormal distribution, as stated by Özkaynak et al. [21]; concentrations of insecticides from our previous study were distributed lognormally, as shown by most common environmental data. Each of the body weight distributions for different ages on the growth charts appears to be skewed to the right, suggesting a lognormal distribution.

Table 1. Distributions of variables used in daily intake estimation for ingestion of dust and soils

<table>
<thead>
<tr>
<th>Variable</th>
<th>N</th>
<th>Mean</th>
<th>SD</th>
<th>P5</th>
<th>P25</th>
<th>P50</th>
<th>P75</th>
<th>P95</th>
<th>Maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ingestion rate of dust (mg/day) a</td>
<td>1,000</td>
<td>26.65</td>
<td>36.54</td>
<td>0.66</td>
<td>4.06</td>
<td>10.80</td>
<td>28.72</td>
<td>100.97</td>
<td>901.96</td>
</tr>
<tr>
<td>Ingestion rate of soils (mg/day) a</td>
<td>1,000</td>
<td>40.96</td>
<td>78.29</td>
<td>0.15</td>
<td>5.26</td>
<td>15.34</td>
<td>44.85</td>
<td>175.60</td>
<td>1367.37</td>
</tr>
<tr>
<td>Indoor concentration of chlorpyrifos in dust (µg/g) b</td>
<td>52</td>
<td>2.32</td>
<td>15.41</td>
<td>&lt; LOD</td>
<td>0.08</td>
<td>0.11</td>
<td>0.30</td>
<td>0.91</td>
<td>112.34</td>
</tr>
<tr>
<td>Outdoor concentration of chlorpyrifos in soils (µg/g) b</td>
<td>57</td>
<td>4.27</td>
<td>23.10</td>
<td>&lt; LOD</td>
<td>&lt; LOD</td>
<td>&lt; LOD</td>
<td>0.11</td>
<td>16.19</td>
<td>134.60</td>
</tr>
<tr>
<td>Indoor concentration of cypermethrin in dust (µg/g) b</td>
<td>52</td>
<td>18.33</td>
<td>55.92</td>
<td>&lt; LOD</td>
<td>0.11</td>
<td>0.37</td>
<td>0.83</td>
<td>105.79</td>
<td>343.27</td>
</tr>
<tr>
<td>Outdoor concentration of cypermethrin in soils(µg/g) b</td>
<td>57</td>
<td>4.29</td>
<td>22.32</td>
<td>&lt; LOD</td>
<td>&lt; LOD</td>
<td>&lt; LOD</td>
<td>0.11</td>
<td>16.47</td>
<td>134.20</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Variable</th>
<th>P3</th>
<th>P15</th>
<th>P25</th>
<th>P50</th>
<th>P75</th>
<th>P95</th>
</tr>
</thead>
<tbody>
<tr>
<td>Body weight of 3 – 6 year old children at (kg) c</td>
<td>13.5</td>
<td>15.1</td>
<td>15.8</td>
<td>17.3</td>
<td>19.0</td>
<td>20.1</td>
</tr>
</tbody>
</table>

SD, standard deviation; P#, #th percentile; < LOD, under limit of detection.

a Derived from Özkaynak et al. [21]; ingestion rate of dust is a sum of dust via hand-to-mouth and object-to-mouth behaviors.
b Derived from Hung et al. [2].
c Derived from Chen et al. [22]; distributions between 3 and 6 years old are combined.

We applied Monte Carlo simulation as probabilistic modeling using Oracle® Crystal Ball (Fusion Edition, Release 11.1.2.3.000), an add-on software to Microsoft® Excel 2010. The software allows users to set up a distribution of data, instead of a value of mean or median, as input of a variable. At each trial, data are randomly selected following the given distributions of variables to compute an outcome value; such a trial can be repeated up to a hundred million times to derive cumulative outcome values, which form a distribution as well with full simulated statistical information (e.g., mean, median, standard deviation) (Figure 1). For estimation of the daily intakes in this study, the variables of IR, C and BW were set as lognormal distributions with parameters listed in Table 1, and doses of chlorpyrifos and cypermethrin via indoor dust and outdoor soil ingestion were separately derived. Each simulation for dose estimation was performed with 1,000,000 iterations, and 30 replicates were completed to get better estimates.

For risk assessment of children’s exposure to both insecticides, we followed a previous study that expressed the estimated daily intakes as percentages of the Acceptable Daily Dose (ADI) values for chlorpyrifos and cypermethrin [23], which were 0.01 and 0.02 mg/kg/day, in accordance with World Health Organization (WHO) [24].

3. Results

The simulation results of daily intakes of chlorpyrifos and cypermethrin via ingestion of indoor dust and outdoor soils for 3 – 6 year old children are presented in Table 2. As all variables in the model appeared to be lognormal distributions, the simulation data were obviously accumulated to form a lognormal distribution (Figure 1d). At the 90th percentile of the daily intake, neither insecticide resulted in more than 0.1% of the respective ADI, indicating that the daily intake of
chlorpyrifos or cypermethrin for young children in the residential environment was negligible with a probability more than 90%. The percentage of ADI increased as the percentile went up; at the P99, the total daily intake of chlorpyrifos was raised to 1.30% of ADI (exposure to indoor dust and outdoor soils), whereas that of cypermethrin was approximately 2.35% of ADI. In the extreme case of P99.9, the total intakes leaped to 13.1% and 20.0% for chlorpyrifos and cypermethrin, respectively.

Figure 1. Distributions of variables (a), (b) and (c), and result (d) for estimation of daily intake of chlorpyrifos

Monte Carlo simulation also conducted sensitivity analysis of modeling, determining which variables had the greatest impact on the model. For the modeling of indoor dust ingestion, the variable of C (concentration of either insecticide) was the one with the greatest impact, bearing a value of contribution to variance around 82%, whereas IR (ingestion rate) only resulted in approximately 18% and BW (body weight) had nearly zero impact (data not shown in table). Ingestion of outdoor soils demonstrated a similar pattern with 91.4% and 8.5% for C and IR, respectively, and no effect for BW. The sensitivity analysis indicated that concentration of insecticide (C) was the principal factor that determined the daily intake.

Table 2. Non-dietary ingestion exposures in terms of percentage of ADI for young children

<table>
<thead>
<tr>
<th>Percentile</th>
<th>Chlorpyrifos via indoor dust</th>
<th>Chlorpyrifos via outdoor soils</th>
<th>Cypermethrin via indoor dust</th>
<th>Cypermethrin via outdoor soils</th>
</tr>
</thead>
<tbody>
<tr>
<td>P50</td>
<td>&lt;0.001% (&lt;0.001%, &lt;0.001%)</td>
<td>&lt;0.001% (&lt;0.001%, &lt;0.001%)</td>
<td>&lt;0.001% (&lt;0.001%, &lt;0.001%)</td>
<td>&lt;0.001% (&lt;0.001%, &lt;0.001%)</td>
</tr>
<tr>
<td>P75</td>
<td>0.002% (0.002%, 0.002%)</td>
<td>0.001% (0.001%, 0.001%)</td>
<td>0.004% (0.004%, 0.004%)</td>
<td>&lt;0.001% (&lt;0.001%, &lt;0.001%)</td>
</tr>
<tr>
<td>P90</td>
<td>0.019% (0.019%, 0.019%)</td>
<td>0.007% (0.007%, 0.007%)</td>
<td>0.052% (0.052%, 0.053%)</td>
<td>0.005% (0.005%, 0.005%)</td>
</tr>
<tr>
<td>P95</td>
<td>0.079% (0.079%, 0.079%)</td>
<td>0.029% (0.029%, 0.029%)</td>
<td>0.208% (0.207%, 0.209%)</td>
<td>0.023% (0.023%, 0.023%)</td>
</tr>
<tr>
<td>P97.5</td>
<td>0.254% (0.253%, 0.255%)</td>
<td>0.102% (0.101%, 0.102%)</td>
<td>0.629% (0.627%, 0.631%)</td>
<td>0.080% (0.079%, 0.080%)</td>
</tr>
<tr>
<td>P99</td>
<td>0.884% (0.880%, 0.887%)</td>
<td>0.412% (0.410%, 0.414%)</td>
<td>2.042% (2.035%, 2.049%)</td>
<td>0.310% (0.308%, 0.311%)</td>
</tr>
<tr>
<td>P99.9</td>
<td>8.313% (8.234%, 8.392%)</td>
<td>4.819% (4.782%, 4.856%)</td>
<td>16.893% (16.745%, 17.042%)</td>
<td>3.155% (3.124%, 3.186%)</td>
</tr>
</tbody>
</table>

Numbers in parenthesis represent the 95% confidence intervals.
As the sensitivity analysis shows, the estimated daily intake is mostly determined by the variable of insecticide concentration in dust or soils (C), data of which are distributed unevenly with the majority at the low end but few with extremely high values. The second influential variable, ingestion rate (IR), appears to be less dispersed than C with a relative standard deviation (RSD = SD/mean) of 1.37 (= 36.54/26.65), compared to an RSD of 6.64 (= 15.41/2.32) for C of chlorpyrifos. The least variable with impact is body weight (BW), which results in an RSD of 0.086, calculated from simulation modeling. It is apparent that variables with large variability turn to be the determinant factors of simulation models, as is C in this modeling of daily intake.

The simulation modeling indicates that the daily intake of chlorpyrifos or cypermethrin is minimal for the most occasions, suggesting that pesticide contamination in Taiwan’s home environments may not need to worry. The concentration profiles, adopted from our previous study [2], monitored pesticide concentrations in residential environments using composite sampling. By definition, a composite sample refers to “a physical mix of individual sample units or a batch of unblended individual sample units that are tested as a group;” in such a way, the sample is “as homogenous as possible” [25]. That is, the concentration profiles built by composite samples were actually groups of mean insecticide concentrations of homes, implying concentrations higher than mean values certainly occurring in “hot spots,” such as rooms after application of insecticides. Thus, our focus should lie on those estimates on the high end (e.g., P99, P99.9), the occurrence of which may be sporadic but needs attention to be paid to.

Even at P99 or P99.9, the estimate daily intake of chlorpyrifos or cypermethrin is considered safe, because the predicted results (<20% of ADI) are lower than ADI suggested by WHO [24]. Nevertheless, let us not forget that non-dietary ingestion is not the only route of exposure to insecticides; dietary ingestion of vegetables or agricultural produce is a major route of insecticide exposure. A Chinese study analyzing 2,083 vegetables for chlorpyrifos and cypermethrin in Zhejiang Province has completed a risk assessment for daily intakes of both insecticides via dietary ingestion on a probabilistic approach [23], providing valuable data for comparison with ours of non-dietary ingestion (Table 3). Because those vegetables are also commonly consumed in Taiwan and the regulations for pesticide residues are similar, we assume that the data of risk assessment could be used as replacement of dietary ingestion exposure to insecticides in Taiwan. As seen in Table 3, with a 99% probability dietary ingestion exposure to either insecticide outweighs non-dietary ingestion exposure by a large margin; at P99.9, however, the ratio of dietary to non-dietary ingestion exposure shrinks to 3.06 for chlorpyrifos and 1.30 for cypermethrin, suggesting that home environment highly contaminated with insecticides may contribute considerable doses to daily intakes for young children, compared to that via dietary ingestion.

### Table 3. Comparison of non-dietary and dietary ingestion exposures in terms of percentage of ADI

<table>
<thead>
<tr>
<th>Percentile</th>
<th>Chlorpyrifos via non-dietary ingestion</th>
<th>Chlorpyrifos via dietary ingestion</th>
<th>Cypermethrin via non-dietary ingestion</th>
<th>Cypermethrin via dietary ingestion</th>
</tr>
</thead>
<tbody>
<tr>
<td>P50</td>
<td>&lt;0.001% (&lt;0.001%, &lt;0.001%)</td>
<td>1.39% (1.35%, 1.42%)</td>
<td>&lt;0.001% (&lt;0.001%, &lt;0.001%)</td>
<td>1.67% (1.64%, 1.70%)</td>
</tr>
<tr>
<td>P75</td>
<td>0.003% (0.003%, 0.003%)</td>
<td>NA</td>
<td>0.004% (0.004%, 0.004%)</td>
<td>NA</td>
</tr>
<tr>
<td>P90</td>
<td>0.026% (0.026%, 0.026%)</td>
<td>15.52% (15.35%, 15.70%)</td>
<td>0.057% (0.057%, 0.058%)</td>
<td>10.55% (10.44%, 10.67%)</td>
</tr>
<tr>
<td>P95</td>
<td>0.108% (0.108%, 0.108%)</td>
<td>NA</td>
<td>0.231% (0.230%, 0.232%)</td>
<td>NA</td>
</tr>
<tr>
<td>P97.5</td>
<td>0.356% (0.354%, 0.357%)</td>
<td>24.07% (23.69%, 24.47%)</td>
<td>0.709% (0.706%, 0.711%)</td>
<td>15.94% (15.68%, 16.19%)</td>
</tr>
<tr>
<td>P99</td>
<td>1.296% (1.290%, 0.887%)</td>
<td>29.03% (28.43%, 29.66%)</td>
<td>2.352% (2.343%, 2.360%)</td>
<td>19.09% (18.70%, 19.52%)</td>
</tr>
<tr>
<td>P99.9</td>
<td>13.095% (13.016%, 13.248%)</td>
<td>40.16% (38.39%, 42.41%)</td>
<td>20.048% (19.869%, 20.228%)</td>
<td>26.07% (24.87%, 27.42%)</td>
</tr>
</tbody>
</table>

NA, not available.

Numbers in parenthesis represent the 95% confidence intervals.

* Data for 2 – 6 years old in the whole province of Zhejiang, derived from Yuan et al. [23].
The comparison in Table 3 also indicates that daily intake of dietary ingestion of either insecticide raises gradually as the percentile goes up, unlike that of non-dietary ingestion remaining at very low levels until reaching the highest percentile. As discussed before, the estimation of daily intake is mostly determined by insecticide concentration; thus, the difference between the two ingestion routes is probably due to much smaller variability for insecticide residues in vegetables than that in dust or soils. Yuan et al. [23] reported the maximum concentrations of either insecticide in vegetables to be <10 \( \mu g/g \), whereas the environmental data used in this study showed the highest levels above 100 \( \mu g/g \) (Table 1). This fact can be reasonably explained because insecticide application for vegetable farming is regulated, meaning that residues of insecticides are supposedly kept under the standards, and variability of the distribution of insecticide residues is limited. In contrast, concentrations of insecticides in dust may be largely different, from none to extremely high levels. For instance, an indoor application of insecticides could sustain much higher levels than that of residues in vegetables for weeks [3, 19]. Even though the ingestion rate of dust is minimal compared to that of vegetables, the estimated daily intake of dust/soil ingestion is as significant as that of vegetable ingestion in a home environment that is highly contaminated with insecticides. Therefore, for homes with need of indoor applications of insecticides, the daily intakes for young children should not be overlooked, particularly for the first few days after application.

The trend that dietary ingestion is the major route of insecticide exposure with non-dietary ingestion catching up at the high percentiles is in support of the work of Xue et al. [26], which used EPA’s SHEDS-multimedia model to estimate pyrethroid intakes for 3 – 5 years old. They estimated the doses for the general population and specifically for people with pyrethroid use in the homes (i.e., residential use population), and found that exposure via non-dietary ingestion prevailed over that via dietary ingestion above the 95th and 75th percentiles for the general population and residential use population, respectively. The difference between our result and theirs is that the estimated daily intakes of non-dietary ingestion from this study do not top that of dietary ingestion of vegetables at any chance, indicating that the residential concentrations of insecticides in Taiwan were lower than those measured from American homes. This is supported by a comparison of pyrethroid concentrations in dust among various studies made in our previous study [2], which shows the lowest median (0.37 \( \mu g/g \)) and P75 values (0.83 \( \mu g/g \)) for Taiwan’s home environments. Despite the slight difference between this work and other western studies, non-dietary ingestion exposure to insecticides could be considerable as the environmental levels are elevated. Frequent home cleaning is recommended to reduce insecticide residues as well as the intakes for young children.

The environmental data used herein comes from a pilot study, and may not fully reflect the distribution of insecticide residues in Taiwan’s houses. In southern cities and counties of Taiwan, insecticide applications are more frequently conducted than that in other parts of the island for vector control (e.g., dengue fever) during the warm and hot months, and the residential insecticide concentrations are expected to be elevated; therefore, the non-dietary ingestion exposure to insecticides could be of special concern in those homes with 3 – 6 year old children.

5. Conclusions

Concentrations of residential insecticide residues are the major determinant factor of the estimation of daily intake via non-dietary ingestion for young children. The daily intake is rarely of concern until the environment is highly contaminated with insecticides (e.g., after an indoor application). It is estimated that the occurrence of such a contamination could bring the intake of non-dietary ingestion exposure close to that of dietary ingestion exposure in the homes of Taiwan, and the total intake would approach the recommended ADI. Frequent home cleaning is necessary for homes with need of insecticide applications to lower the exposure of young children.

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Author Contributions: Ya-Qing Yang conceived the study concept and worked on preliminary trials. Lih-Ming Yin contributed to study design, performed data analysis and wrote the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.
References


