

1 **Title:**

2 Remediating Indoor Pesticide Contamination from Improper Pest Control  
3 Treatments: Persistence and Decontamination Studies

4  
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29 **Abstract**

30

31 The improper and excessive use of pesticides in indoor environments can result in adverse  
32 human health effects, sometimes necessitating decontamination of residential or commercial  
33 buildings. A lack of information on effective approaches to remediate pesticide residues  
34 prompted the decontamination and persistence studies described in this study.

35 Decontamination studies evaluated the effectiveness of liquid-based surface decontaminants  
36 against pesticides on indoor surfaces. Building materials were contaminated with 25-2,400  
37  $\mu\text{g}/100\text{cm}^2$  of the pesticides malathion, carbaryl, fipronil, deltamethrin, and permethrin.

38 Decontaminants included both off-the-shelf and specialized solutions representing various  
39 chemistries. Pesticides included in this study were found to be highly persistent in a dark indoor  
40 environment with surface concentrations virtually unchanged after 140 days. Indoor light  
41 conditions degraded some of the pesticides, but estimated half-lives exceeded the study

42 period. Decontamination efficacy results indicated that the application of household bleach or a  
43 hydrogen peroxide-based decontaminant offered the highest efficacy, reducing malathion,  
44 fipronil, and deltamethrin by >94-99% on some surfaces. Bleach effectively degraded  
45 permethrin (>94%), but not carbaryl (<70%) while the hydrogen peroxide containing products  
46 degraded carbaryl (>71-99%) but not permethrin (<54%). These results will inform responders,  
47 the general public and public health officials on potential decontamination solutions to  
48 remediate indoor surfaces.

49 **Keywords**

50 pesticide

- 51 misuse
- 52 decontamination
- 53 cleanup
- 54 persistence

## 55 **1. Introduction**

56 Under the U.S. Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA), it is a violation to  
57 use a pesticide in a manner inconsistent with the product's labeling. Applying pesticides in ways  
58 that deviate from the label directions is illegal and considered a misuse or a misapplication.  
59 Misusing pesticides can include the application of products not registered by the U.S  
60 Environmental Protection Agency (EPA) [1], off-label application in areas where the product is  
61 not intended, or at higher rates or concentrations (mass per unit area) than specified on the  
62 product label. Misapplications also include disregarding safety instructions and applying  
63 restricted-use pesticides without proper applicator certification.

64 Misapplying pesticides in homes, schools, businesses or other indoor environments can lead  
65 to adverse health effects and contamination [2-8], often impacting vulnerable populations.  
66 Building occupants and federal, state and local agencies responding to pesticide misuse  
67 incidents seek information about whether pesticide residues present exposure risk and how to  
68 clean treated surfaces to reduce pesticide levels, if necessary [7-10]. This study does not  
69 attempt to evaluate whether pesticide levels are unsafe but provides decontamination  
70 information should remediation be desired or deemed appropriate.

71 Tools to determine the potential risk to occupants from misused pesticides are limited, and  
72 there are few known effective cleaning procedures to reduce pesticide levels in affected  
73 structures. Proper cleanup can also be very costly, presenting significant challenges for  
74 individuals with limited economic resources. As a result, occupants could continue to inhabit  
75 contaminated buildings, could be forced to vacate contaminated properties, or could attempt  
76 to remediate on their own, possibly creating toxic byproducts or further spreading pesticides

77 residues. Science based remediation methods are needed to safely reduce occupant exposure  
78 following pesticide misapplication incidents.

79 Although the fate and transport of pesticides in the environment has been studied  
80 extensively including those completed for pesticide registration [11], there is very limited  
81 information on the persistence of pesticides in indoor environments [12-15] where degradation  
82 mechanisms, such as direct sunlight, water, and soil microbes, do not exist. Numerous studies  
83 have documented the presence of many different pesticides inside homes and day care centers  
84 in indoor air, in dust and on surfaces [16-21] indicating long-term persistence. Few studies  
85 evaluate indoor pesticide fate for extended periods but results from previous pesticide misuse  
86 cases (reference 8 and personal communication) suggest that pesticide residues persist indoors  
87 due to the absence of the primary degradation factors found outdoors. To confirm that  
88 pesticides of interest (malathion, carbaryl, deltamethrin, fipronil, and permethrin) do persist in  
89 the indoor environments, the present study included persistence tests conducted in a  
90 controlled environment under dark and indoor light conditions, to assess the rate of dissipation  
91 of the pesticides from the surface via volatilization and/or degradation.

92 Managing outdoor pesticide spills [22] and remediating pesticide contaminated soil or  
93 water traditionally focus on control, containment and various cleanup technologies. The  
94 cleanup procedures following an outdoor release tend to focus on the physical removal of the  
95 contamination and, if applicable, leaving pesticide residues associated with normal application  
96 conditions in place to breakdown naturally. Leaving residues to degrade naturally indoors may  
97 not be a suitable approach due to extended indoor persistence (reference 8 and verbal

98 communication). Additionally, when pesticide misuse results in an unsafe environment or  
99 occupant evacuation, more immediate and proactive residue removal or reduction is required.

100 Information about decontamination approaches for pesticides on indoor surfaces is almost  
101 nonexistent and knowledge on complete chemical degradation pathways is limited to general  
102 concepts derived from processes observed in water or soils. Initial attempts to remediate  
103 pesticide contaminated surfaces through general washing and physical removal are rarely  
104 successful [7-9] and physical removal may not be possible for all surfaces. Pesticide  
105 manufacturer labels or safety data sheets (SDS) identify decontaminants such as chlorine  
106 bleach, caustic soda, or lime without evidence or reference to degradation rates. Use of these  
107 products by building occupants may pose health risks and may not be practical for use on  
108 common household items or for extensive use throughout grossly contaminated residential or  
109 business settings. Further, potentially toxic byproducts may be formed during decontamination,  
110 which would require additional chemical analysis and costs in order to verify a successful  
111 cleanup.

112 To inform safer and effective application of decontaminants, we conducted a series of  
113 decontamination experiments on malathion, carbaryl, deltamethrin, fipronil, and permethrin  
114 contaminated materials. Because these studies were the first of their kind, they focused strictly  
115 on chemical interactions to degrade pesticides on indoor surfaces to better understand efficacy  
116 independent of various scrubbing, rinsing and other physical removal procedures. In addition,  
117 physical removal processes introduce numerous variables beyond the scope of this  
118 investigation.

119 The efficacies of selected commercially available decontamination solutions (Spic and Span®  
120 cleaner, Clorox® Bleach, EasyDECON DF200®, and Sterilex® Ultra-Kleen) were examined for  
121 their ability to degrade the pesticides under operationally realistic conditions such as  
122 application of a representative decontaminant volume per surface area and contact time. These  
123 solutions were selected based on their observed effectiveness (Sterilex® Ultra-Kleen) for  
124 remediating organophosphate pesticide (methyl parathion) contaminated homes [8], and on  
125 the chlorine-based oxidation chemistry (Clorox® Bleach) [23], because they were designed to  
126 degrade organophosphate chemical warfare agents (EasyDECON DF200®) [24], or because they  
127 are a conventional, commercially available detergent solution (Spic and Span® cleaner). This  
128 included a measurement of the efficacy of decontamination solutions on three building  
129 materials (stainless-steel, plywood, and vinyl flooring) using representative decontamination  
130 solution dwell times on the pesticide contaminated surfaces and contamination levels  
131 measured in pesticide misuse incidents (25–2,400 µg/100cm<sup>2</sup>). The measured contamination  
132 levels vary due to (1) the amount and concentration of the product applied at the site; (2) the  
133 surface types sampled and sampling methods used; (3) the time that passed between the  
134 application and the sampling; or (4) previous applications and residues from other sources,  
135 such as being tracked-in from outdoors or from use of pet products containing the same  
136 pesticides. SDS and other health and safety information on any decontamination solution  
137 should be reviewed as the decontamination solution itself may introduce an additional  
138 exposure risk to personnel. This study included a semi-quantitative analysis for known  
139 byproducts of noticeable toxicity following the decontamination process which were identified  
140 for two pesticides, malathion and fipronil.

## 141 2. Materials and Methods

### 142 2.1 Pesticides

143 Targeted pesticides were technical grade malathion (CAS Number 121-75-5), carbaryl (CAS  
144 Number 63-25-2), fipronil (CAS Number 120068-37-3), deltamethrin (CAS Number 52918-63-5),  
145 and permethrin (CAS Number 52645-53-1) as well as two commercial pesticide formulations,  
146 Ortho® MAX® Malathion Inspect Spray Concentrate [Ortho MAX] (The Scotts Company LLC,  
147 Huntsville, TX) and Sevin® Carbaryl Insecticide [Sevin] (TechPac, LLC, Atlanta, GA). All pesticides  
148 and pesticide containing formulations that were part of this study are tabulated in Table 1 and  
149 were procured from either a commercial source (Sigma Aldrich, St. Louis, MO) at 97% or higher  
150 purity or as commercial formulations from local vendors. Labeled internal standards of  
151 pesticides were purchased from Cambridge Isotope Laboratories (Tewksbury, MA, USA), CDN  
152 Isotopes (Pointe-Claire, Quebec, Canada) and Dr. Ehrenstorfer® GmbH (Augsburg, Germany).

153 The pesticides selected are common use, registered insecticides that have been misused  
154 indoors [2-8]. They also represent a range of pesticide classes and physiochemical properties as  
155 to evaluate their behaviors on surfaces and chemical interactions with decontamination  
156 solutions. Surface contamination levels, as tabulated in Table 1, were different for each  
157 pesticide based on the highest observed surface concentrations reported in several pesticide  
158 misuse investigations conducted by state pesticide regulatory agencies and shared with US EPA.  
159 This study utilized these pesticide surface concentrations because a high level of surface  
160 contamination is likely more difficult to cleanup than a lower level of surface contamination.

161 Technical grade pesticides were dissolved in n-hexane (>98.5%, mixture of hexane isomers,  
162 HPLC, GC, pesticide residue analysis grade) or dichloromethane (>99.9% HPLC, GC, pesticide



163 residue analysis grade) at stock concentration values that allowed for the application of a single  
164 10 µL droplet of the dissolved pesticide on the targeted surfaces to reach the intended surface  
165 concentration. After chemical analysis of the concentration of target chemicals in commercial  
166 pesticide formulations, the Ortho MAX malathion-containing product was mixed with deionized  
167 water (1:125 ratio) to create the same malathion concentration on the surface as tested using  
168 the technical grade. Similarly, the Sevin carbaryl containing product was diluted 8.3-fold with  
169 deionized water to create the same carbaryl surface concentration for a direct comparison with  
170 the technical grade decontamination tests.

## 171 2.2 Test surface materials

172 Test materials were selected to represent a variety of commonly encountered indoor  
173 surfaces with potential different degrees of permeability to pesticides. The materials selection  
174 was based on its likely use as a subflooring material (plywood) under carpet or hardwood  
175 flooring, or actual flooring material (vinyl) in a residence. Stainless-steel was representative of a  
176 nonporous surface and served as a reference material since treatment and removal methods  
177 are expected to perform more efficiently on this material. Materials were also selected for  
178 relative simplicity to minimize potential interactions due to surface material interferences. For  
179 example, painted or stained surfaces commonly found in homes may interact with the  
180 pesticides and decontamination solutions and complicate interpretation of results. Further fate  
181 and transport research are needed on more complex surfaces before decontamination  
182 approaches can be developed. All test substrates were spiked with a pesticide and underwent  
183 decontamination treatments. Large sections or panels of each material were obtained from  
184 suppliers (stainless-steel, 304 Grade; McMaster-Carr; plywood, untreated pine plywood, Lowe's

185 Home Improvement; and vinyl, TrafficMaster Allure, Lowe's Home Improvement). Small  
186 rectangular coupons (2.5 × 4.0 cm dimensions) were cut from the larger materials and were  
187 used as substrates for the application of the pesticides and subsequent decontamination steps.  
188 All materials were cleaned prior to use by removal of any dust followed by a surface cleaning  
189 with methanol. Material coupons were spiked with a single 10 µL droplet, using a gas tight  
190 micro-syringe (SGE Analytical Science, Melbourne, Australia), of the targeted pesticide at  
191 pesticide specific stock concentrations in hexane or dichloromethane.

### 192 2.3 Decontamination solutions

193 Four decontamination products were initially chosen for testing. EasyDECON® DF200  
194 (hereafter, DF200) solution was prepared by proportional mixing of DF200 Parts 1-3 (hydrogen  
195 peroxide solution as the active ingredient with surfactants; pH = 9.8). DF200 Part 1 is composed  
196 of cationic detergents and fatty alcohols; DF200 Part 2 is an 8% hydrogen peroxide stabilized  
197 solution; and DF200 Part 3 contains diacetin. Sterilex Ultra Kleen Solution 1&2 (hereafter,  
198 Sterilex) was prepared by proportional mixing of Solution 1 and 2 (peroxide solution, pH = 11).  
199 Sterilex Solution 1 contains hydrogen peroxide (6-6.6% by weight) as the active ingredient,  
200 quaternary ammonium compounds and ammonium salts while Solution 2 contains sodium- and  
201 potassium carbonates (approximately 6% by weight). Clorox® Concentrated Germicidal Bleach  
202 (8% sodium hypochlorite as the active ingredient, pH = 11.4) (hereafter, bleach) was applied as  
203 received without dilution. Spic and Span® Liquid Multi-Surface and Floor Cleaner (hereafter,  
204 Spic&Span) solution was prepared per manufacturer instructions (sodium carbonate as the  
205 active ingredient and surfactant, pH = 9.4-9.7). All decontamination solutions were prepared  
206 immediately prior to use. The use of full-strength bleach without dilution to clean surfaces is

207 not recommended as per the manufacturer's label. Previous decontamination testing using ten-  
208 fold diluted bleach on materials contaminated with organophosphate nerve agents suggested  
209 low efficacy [23]. Therefore, this bench-scale study used full strength bleach to evaluate  
210 whether a concentrated solution would degrade the pesticides more completely without  
211 leaving toxic byproducts.

212 After a 5-min contact time of the pesticide solution with the flooring or nonporous  
213 reference material (to allow the solvent to evaporate), 75  $\mu$ L of the decontamination solution  
214 was applied as a single droplet over the contaminant using a gas tight micro-syringe (SGE  
215 Analytical Science, Melbourne, Australia). Considering the small coupon sizes, the  
216 decontamination solution was applied as a single droplet that was large enough to cover the  
217 pesticide contaminated area on a coupon. This volume is representative of the volume of  
218 decontaminant solution applied to a surface from a back-pack type sprayer [23]. The  
219 decontamination solution was allowed to interact with the pesticide on the surface for 18 hours  
220 (representing an overnight drying of the decontamination solution). For a second set of test  
221 coupons, the same decontaminant with the same volume was reapplied after 90 minutes  
222 followed by 16.5 hours contact time. Both sets of test coupons, as well as the positive controls  
223 (see below), were extracted at the same time (18 hr after contamination). Each test point  
224 consisted of two sets of three replicate test coupons (contaminated and decontaminated  
225 material). Each test point also included two positive controls that were contaminated with a  
226 pesticide but not decontaminated; one procedural blank that was not contaminated with a  
227 pesticide, but the decontamination solution was applied; and one laboratory blank that was not  
228 treated with either pesticides or decontamination solutions.

229 The decontamination testing started with all the decontaminants (DF200, Sterilex, bleach,  
230 Spic&Span) applied against the malathion contaminated materials (stainless-steel, plywood,  
231 and vinyl). Subsequent decontamination testing for other pesticide contaminated materials was  
232 limited to the better performing decontaminants and to a lower number of materials (see Table  
233 2).

#### 234 2.4 Pesticide persistence tests

235 Persistence tests were conducted for the targeted pesticides as applied to stainless-steel  
236 coupons (10 cm<sup>2</sup> surface area). Fourteen sets of triplicate stainless-steel coupons plus one  
237 triplicate Day 0 set were contaminated with a pesticide mixture containing malathion, carbaryl,  
238 fipronil, deltamethrin, and permethrin at concentrations to reach the surface concentration in  
239 Table 1. All contaminated coupons, as well as single procedural blank coupons (no pesticide  
240 applied), were placed in two chambers with controlled temperature ( $24 \pm 3^\circ\text{C}$ ) and  $50 \pm 3\%$   
241 relative humidity (RH). Seven sets of contaminated coupons were placed in a dark chamber.  
242 The other seven sets were placed in the second chamber that simulated an indoor light  
243 environment. The indoor lighting was simulated using seven 4 Watt fluorescent warm white  
244 light fixtures (color temperature 3000 K). Light fixtures were present in close distance to the  
245 coupons (2-5 inch range). The air exchange rate for both chambers was controlled at one air  
246 exchange per hour representing a common air change rate for standard residences [25]. At  
247 seven intermediate time points up to 140 days post pesticide application, coupon sets including  
248 the procedural blank coupon were removed from both chambers and extracted to quantify  
249 residual pesticide mass. One set of triplicate coupons was extracted immediately after spiking  
250 with the pesticide and functioned as the Day 0 starting pesticide mass.

251 2.5 Extraction of pesticides from materials and neutralization of residual decontaminant

252 After a 30-min contact time between the pesticide and surface, residual pesticide amounts  
253 were recovered by transfer of the coupon into an extraction vial containing 50 mL of n-hexane  
254 as the extraction solvent. Vials were then sonicated for 10 min. Following sonication, a 10 mL  
255 aliquot of the extract was solvent exchanged with a 10 mL 3:7 water: methanol ratio (v/v)  
256 solvent mixture for LC-MS/MS analysis. Extracts that were to be analyzed by GC-MS remained  
257 in the hexane solvent. Extracts were diluted as needed, spiked with internal standard (See Table  
258 A, Supporting Materials) and stored in a refrigerator (4°C) or freezer (-20°C) for interim storage  
259 until chemical analysis. Extraction efficiencies, defined as the percent ratio between recovered  
260 pesticide mass from a coupon material and that from a spike control into the same extraction  
261 solvent, were determined for all material / pesticide combinations prior to the decontamination  
262 study. Almost all recoveries (see Table A, Supplemental Materials) using this method exceeded  
263 the initial targeted recovery value (better than 70%, but not higher than 120%, recovered mass)  
264 with a low coefficient of variance among test coupons (less than 30%). In general, extraction  
265 efficiency increased for the extraction of pesticides from plywood to vinyl to stainless-steel.  
266 This can be attributed to the more porous nature of plywood and vinyl versus stainless-steel.  
267 Extraction of both permethrin and deltamethrin from vinyl and plywood did not reach the 70%  
268 target extraction efficiency but were in the 47-57% range, which were still deemed acceptable  
269 as lower recoveries would be associated with both positive controls and test coupons leading to  
270 a limited impact in calculated decontamination efficacy.

271 It is critical that the extract is neutralized as residual decontaminant that is part of the  
272 extract may continue to degrade the pesticide during the extraction and handling of the extract

273 prior to the analysis. This would bias the recovered pesticide amount leading to artificially  
274 higher decontamination efficacy values than those associated with just the degradation of the  
275 pesticide on the surface with the decontaminant. The selection of hexane as a nonpolar solvent  
276 simplified the needed quenching of the decontamination reaction in the extract as the  
277 pesticides partitioned into this solvent layer while the residual decontaminant formed a  
278 separate aqueous layer. By taking an aliquot from the solvent phase rather than the reactive  
279 ingredient-containing aqueous phase, a further separation of reactant (the residual  
280 decontaminant) and pesticide is guaranteed. This was verified prior to the actual  
281 decontamination testing through spiking of a pesticide into a simulated extract.

## 282 2.6 Analytical methods

283 Chemical analysis of the coupon extracts occurred using two analytical methods, gas  
284 chromatography/mass spectrometry (GC/MS) for malathion (decontamination tests only) and  
285 liquid chromatography/tandem mass spectrometry (LC-MS/MS) for malathion (persistence  
286 tests only), carbaryl, fipronil, deltamethrin, and permethrin. Details on the analytical methods  
287 are provided in the Supplemental Materials. The lowest recovered pesticide mass is reported at  
288 the method quantification level (MQL), which was defined in this study as the lowest calibration  
289 curve standard for each pesticide in the study. All malathion extracts were screened for the  
290 presence of malaoxon, a toxic oxidation byproduct of malathion [26,27]. For the fipronil  
291 decontamination tests, extracts were screened for the presence of fipronil sulfone, fipronil  
292 amide, and fipronil desulfinyl as fipronil byproducts of noticeable toxicity [28,29].

## 293 2.7 Decontamination test matrix, calculations, and statistical analysis

294 Table 2 summarizes the overall decontamination test matrix. Not all decontaminant-  
295 pesticide combinations were evaluated. Results from the first pesticide (malathion)  
296 decontamination tests were considered in the selection of decontamination solutions for the  
297 second pesticide (carbaryl) with two decontaminants being selected for the other three  
298 pesticides. Decontaminants that were excluded from the test matrix in this study should not *a*  
299 *priori* be considered ineffective unless explicitly noted. This decontamination study focused on  
300 the identification and efficacy measurement of decontamination solutions that were expected  
301 to be efficacious based on the limited literature on surface decontamination via chemical  
302 degradation of similar pollutants [2,24]. Decontamination efficacy tests were also executed for  
303 two commercial pesticide products applied (at the same pesticide surface concentration) to  
304 address impacts of product formulation on the observed efficacy.

305 The decontamination efficacy was defined as the percentage decrease in the mean  
306 pesticide mass recovered from the decontaminated (test) coupons ( $\bar{M}_{TC}$ ) compared to the  
307 mean pesticide mass applied to the test coupons (positive controls, ( $\bar{M}_{PC}$ )). To account for  
308 possible losses, e.g., due to volatilization or natural degradation, the positive control coupons  
309 (contaminated but not decontaminated) were extracted at the same time as the test coupons  
310 (i.e., after 18 hr) and were used to account for such a decrease. For a given test coupon (TC),  
311 the response variable calculated for this analysis was:

$$312 \quad \text{Decontamination Efficacy} = 100 \times \frac{\bar{M}_{PC} - \bar{M}_{TC}}{\bar{M}_{PC}}$$

313 The standard deviation in the mean mass recovered from test and positive control coupons was  
314 used to derive the standard deviation in decontamination efficacy through propagation of  
315 error.

316 Two-tailed Student's t-tests were used to compare the means of the residual pesticide mass  
317 following the application of the decontamination solution. Unequal variance between the  
318 populations was assumed. A p-value is the result of such comparison and results are considered  
319 significantly different if  $p < 0.05$ . The statistical analysis included comparisons between pesticide  
320 residuals following different decontamination solution applications per pesticide; comparisons  
321 in pesticide residuals between single and double decontamination solution applications; and  
322 comparisons in residuals among the three materials per decontamination solution.

### 323 **3. Results**

#### 324 3.1 Persistence of pesticides in simulated indoor environment

325 The mean recovered mass for carbaryl, fipronil, deltamethrin, and permethrin from a  
326 stainless-steel material surface as function of time (up to 140 days) is shown in Figure 1.  
327 Persistence test results for malathion are not reported due to high inconsistencies in the  
328 extracted malathion within the first two weeks of the experiment. This was attributed to the  
329 high dilution ratio of the extract prior to analysis leading to concentrations close to the MQL.  
330 The malathion persistence test was not repeated. Analysis of the deltamethrin samples was  
331 limited to five time points as extracts were lost due to a solvent extraction step error for two  
332 time points.

333 None of the pesticides were detected on any of the procedural and laboratory blanks. In a  
334 dark environment, all pesticides were found to be highly persistent with near equal amounts of  
335 pesticide recovered after 140 days compared to the contamination level at the start. Initial  
336 contamination levels were based on observed surface concentrations from various pesticide



337 misuse investigations. In the presence of simulated indoor light conditions, some degradation  
338 of most of the pesticides was observed. Estimated half-life values on a nonporous stainless-  
339 steel surface in an indoor light environment were derived from a single first-order kinetics  
340 model which specifies that the rate of concentration decline is proportional to the  
341 concentration in the system. Fitted half-life values are tabulated in Table 3. Half-life values  
342 range from 166 days for fipronil and deltamethrin to 286 days for permethrin. A linear fit to the  
343 dark indoor condition persistence data for carbaryl, fipronil and deltamethrin resulted in slope  
344 values that were not statistically significantly different from zero ( $p < 0.05$ ). Hence, half-life  
345 values in dark environments could not be calculated based on these data but are expected to  
346 exceed 500 days. No appreciable degradation was observed for carbaryl on stainless-steel over  
347 the 140-day period in both dark and indoor light environments. The initial increase (Day 1 to  
348 Day 21) in recovered amounts of carbaryl from the stainless-steel coupons in both the dark and  
349 indoor light environment is probably due to an analytical bias in the highly diluted carbaryl  
350 samples. For comparison, Table 3 includes reported half-life values for water and soil. Reported  
351 ranges in water and soil half-life values are mainly due to their dependence on pH in water or  
352 the soil type and depth [30-36]. The analysis of the extracts by LC-MS/MS did not include  
353 identification of possible degradation products except for the fipronil persistence test where  
354 fipronil sulfone was included in the analysis. Fipronil sulfone (data provided in Supplemental  
355 Materials) was present in each sample (approximately 200 ng/coupon or less than 2% of  
356 applied fipronil mass) from Day 0 to Day 140 with no noticeable change in mass recovered as  
357 function of time and independent on dark/indoor light conditions.

358 3.2 Decontamination results for indoor building materials contaminated with malathion

359 Decontamination efficacy values for a single or double application of the decontamination  
360 solution followed by an 18 hr contact time (simulating an overnight drying) are shown in Figure  
361 2 for indoor materials contaminated with 400  $\mu\text{g}/100\text{cm}^2$  malathion. For malathion,  
362 decontamination using Spic&Span left more than 50% of the initial malathion applied on the  
363 surface after an 18 hr contact time. All other tested decontaminants (DF200, Sterilex, and  
364 bleach) yielded greater than 96% reduction in malathion on stainless-steel and vinyl.  
365 Decontamination of plywood was noticeably less efficacious than that of stainless-steel or vinyl  
366 except when bleach was used. A second application of the same decontamination solution 90  
367 min after the first application improved the efficacy for plywood. For the other two materials,  
368 residual malathion levels were already at or just above the MQL (equivalent to 2.5  $\mu\text{g}/100\text{cm}^2$   
369 malathion) after one application of DF200, Sterilex, and bleach. Hence, the impact of a second  
370 application for these two materials could not be established. Residual malathion levels after a  
371 single or double application are tabulated in Table 4. Malathion was not detected on any of the  
372 procedural and laboratory blanks.

373 Residual malathion amounts were highest when using Spic&Span followed by Sterilex,  
374 DF200 and bleach for the stainless-steel plywood, and vinyl materials. A statistical comparison  
375 of residual malathion amounts following decontamination showed that residue levels were only  
376 significantly different ( $p < 0.05$ ) between those following the Spic&Span application and any of  
377 the other three decontaminants, while residuals among the three decontaminants were not  
378 significantly different statistically ( $p > 0.05$ ). The observed reduction in residual malathion  
379 associated with two applications of decontamination solution were not significantly different  
380 from the results following a single application of decontamination solution ( $p > 0.05$ ). When

381 comparing malathion residuals after a double decontamination application, plywood generally  
382 had the highest amount of malathion remaining followed by vinyl and stainless-steel. However,  
383 the residual malathion on plywood was not significantly different statistically ( $p>0.05$ ) from that  
384 on stainless-steel or vinyl. The only exception was for the Spic&Span product where the  
385 residual amount of malathion on plywood was significantly different from that on vinyl or  
386 stainless-steel.

387 All malathion extracts were screened for the presence of malaoxon, a toxic oxidation  
388 byproduct of malathion. One extract out of three replicates associated with the bleach  
389 decontamination of the plywood contained malaoxon. The recovered malaoxon mass from this  
390 single coupon was not quantified. No malaoxon was observed in extracts following  
391 decontamination with the other decontaminants DF200, Sterilex, and Spic&Span.

392 The Spic&Span product was not evaluated further against the other pesticides based on its  
393 poor decontamination performance against malathion and the lack of an active oxidative  
394 ingredient.

### 395 3.3 Decontamination results for indoor building materials contaminated with carbaryl

396 Figure 3 summarizes the decontamination efficacies as observed when applying DF200,  
397 Sterilex, and bleach for single and double applications onto the three materials contaminated  
398 with carbaryl (at a  $2400 \mu\text{g}/100\text{cm}^2$  surface concentration). Efficacies were highest when using  
399 the DF200 product followed by Sterilex and bleach. For carbaryl, efficacies exceeding 99% were  
400 only observed for the DF200 solution when applied to stainless-steel and vinyl. This material  
401 dependence was not observed with bleach although bleach efficacies never exceeded 70%  
402 across all materials. Residual carbaryl levels after one or two decontaminant applications are

403 tabulated in Table 4. Carbaryl was not detected on any of the procedural and laboratory blanks.  
404 As was the case for malathion contaminated materials, a double application of  
405 decontamination solution did not yield significantly different ( $p>0.05$ ) residual carbaryl levels  
406 except for bleach on stainless-steel. Here, a significant reduction ( $p=0.02$ ) in residual carbaryl  
407 was observed when bleach was applied twice. Although residuals on plywood after any of the  
408 decontamination tests were higher than those on other materials, these differences were not  
409 significantly different statistically ( $p>0.05$ ) except when comparing carbaryl residuals on  
410 plywood against those on vinyl after decontamination with Sterilex.

#### 411 3.4 Decontamination results for indoor building materials contaminated with fipronil

412 The decontamination efficacy results for fipronil contaminated materials are shown in  
413 Figure 4. These decontamination tests were limited to the DF200 and bleach solutions. Both  
414 solutions accomplished a better than 99% reduction in fipronil mass (applied at 150  
415  $\mu\text{g}/100\text{cm}^2$ ), even after a single application except when applied to plywood. The repeated  
416 application did not improve decontamination efficacy appreciably for both decontamination  
417 solutions. Residual fipronil amounts after decontamination are tabulated in Table 4. Fipronil  
418 was not detected on any of the procedural and laboratory blanks. Residual amounts of fipronil  
419 after one application were at or just above the MQL (equivalent to  $0.5 \mu\text{g}/100\text{cm}^2$  for fipronil)  
420 for both decontaminants on stainless-steel and vinyl. In decontamination tests in which residual  
421 fipronil was above the MQL, none of the residuals were found to be significantly different  
422 ( $p>0.05$ ) in a direct comparison between the two decontaminants. The changes in residual  
423 amount of fipronil after a repeated application of DF200 and bleach were not statistically  
424 significant ( $p>0.05$ ). Plywood was more difficult to decontaminate with bleach with statistically

425 significant higher amounts of fipronil recovered after a single application. Residuals on plywood  
426 after a second application of bleach were not significantly different compared to the residuals  
427 on the other two materials.

428 All extracts were screened for the presence of fipronil amide, fipronil desulfinyl, fipronil  
429 sulfone as three possible fipronil degradation byproducts [28,29]. All three byproducts were  
430 detected in small quantities (up to 0.015 µg fipronil amide; up to 0.05 µg for fipronil desulfinyl  
431 and up to 0.25 µg fipronil sulfone per extract / 10 cm<sup>2</sup> size coupon) in the test coupon extracts  
432 of all materials and across both decontaminants. They were also detected in the positive  
433 control extracts at the same levels/concentrations. Hence, it was impossible to attribute the  
434 detection of these three byproducts to their formation during the decontamination process  
435 with either decontaminant. Further, these byproduct concentrations were in the same order of  
436 magnitude to a factor 10 lower than residual fipronil following decontamination of the  
437 materials.

438 3.5 Decontamination results for indoor building materials contaminated with pyrethroids

439 The efficacy testing for materials contaminated with the pyrethroids deltamethrin and  
440 permethrin (at 25 and 500 µg/100cm<sup>2</sup>, respectively) are summarized in Figure 5.

441 Decontamination testing for bleach was limited to stainless-steel material only. For  
442 deltamethrin, stainless-steel was the easiest material to decontaminate (better than 94%  
443 efficacy) with both DF200 and bleach leading to residuals near or below the MQL of 0.5  
444 µg/100cm<sup>2</sup>. Where low efficacies were observed, a second application of the same  
445 decontaminant resulted in a slightly improved efficacy. The results for permethrin are different  
446 from deltamethrin as efficacy values with the DF200 product never reached 55%. Residual

447 contamination levels for these two pyrethroid pesticides after decontamination with DF200 and  
448 bleach are summarized in Table 4. Both pyrethroids were not detected on any of the procedural  
449 and laboratory blanks. Deltamethrin residuals following decontamination with DF200 were  
450 below the MQL (0.05 µg/coupon) for stainless-steel while higher residuals were detected on  
451 vinyl and plywood. These higher residuals were not significantly different ( $p>0.05$ ) than those  
452 for stainless-steel. Deltamethrin residuals following decontamination with bleach were limited  
453 to the stainless-steel material. The repeated application of DF200 or bleach did not result in  
454 statistically significantly lower residuals of deltamethrin. This statement is biased since the MQL  
455 for residual deltamethrin was already reached for stainless-steel after a single application of  
456 both DF200 and bleach. The differences in residual deltamethrin levels across the three  
457 materials were not significantly different statistically ( $p>0.05$ ). Permethrin residuals following  
458 decontamination with DF200 were noticeably higher than those following bleach  
459 decontamination although there is no direct overlap in the tested materials. The repeated  
460 application did not significantly reduce residuals ( $p>0.05$ ) for either decontaminant on tested  
461 materials. Residuals pyrethroids on the plywood and vinyl surfaces following DF200  
462 decontamination were not significantly different statistically ( $p>0.05$ ).

463 3.6 Decontamination results for indoor building materials contaminated with commercial  
464 pesticide formulations

465 Decontamination efficacy results shown so far were based on the use of a technical grade  
466 pesticide product. In most pesticide misuse cases, a commercial formulation would have been  
467 applied. Figure 6 shows the observed efficacies for the DF200 and bleach products when  
468 decontaminating a stainless-steel surface contaminated with the formulated pesticide products

469 Ortho MAX (containing 25-50% malathion) and Sevin (containing 43% carbaryl by weight) at the  
470 same pesticide loading used for the other decontamination studies. Decontamination efficacies  
471 were better than 99.2% with no detectable residual malathion after DF200 or bleach  
472 applications on stainless steel surfaces that were contaminated with either technical grade  
473 malathion or Ortho Max. Efficacies using the DF200 product were somewhat lower when  
474 decontaminating stainless steel contaminated with Sevin (88%) than with technical grade  
475 carbaryl (>99.9%). Efficacies when using bleach to decontaminate stainless steel contaminated  
476 with Sevin was 74% which is slightly higher than when contaminated with technical grade  
477 carbaryl (70%).

#### 478 **4. Discussion**

479 The persistence test results show that pesticide mass on the tested surfaces does not  
480 readily dissipate in indoor environments. Calculated half-life values from a first order  
481 exponential decay of the recovered mass data are noticeably longer than half-life in water and  
482 comparable to those in soil. The lack of liquid water, sunlight, or only low levels of (different)  
483 microbes in an indoor environment will limit the natural degradation of these pesticides. This is  
484 consistent with field observation of pesticide residues from applications that were made  
485 several years earlier [15]. Further research should consider whether any toxic byproducts are  
486 formed during the prolonged presence of these pesticides on these indoor materials.

487 The decontamination efficacy tests show that effective decontamination approaches exist  
488 that can reduce the initial pesticide mass by more than 95%. The hydrogen peroxide chemistry  
489 existing in DF200 and Sterilex products was found to be highly effective for degrading  
490 malathion, carbaryl, fipronil, deltamethrin, and permethrin on stainless-steel, plywood, and

491 vinyl. Hydrogen peroxide is a strong oxidant with a pH dependent electrochemical oxidation  
492 potential ranging between 0.87 and 1.80 V [37]. Similarly, the hypochlorite oxidation (oxidation  
493 potential 0.95 V at pH 9.5 [38]) in bleach effectively reduced the mass of all tested pesticides  
494 except for permethrin. A comprehensive understanding of the decontamination  
495 mechanisms/reactions cannot be derived from this study alone. Such effort requires liquid  
496 reactor chemistry experiments that utilize a comprehensive suite of chemical analysis  
497 instrumentation that were beyond the scope of this study.

498       Based on the observed degradation of malathion in the environment, malathion is expected  
499 to have been initially oxidized at the P=S bond to its P=O oxon analog, malaaxon, which is more  
500 toxic than the parent compound but would have likely degraded further to O,S-Dimethyl  
501 phosphorothioate and diethyl succinate [39]. Malaaxon was detected as a malathion  
502 degradation product in one of the seventy-two decontamination test samples. The very limited  
503 detection of malaaxon as an oxidation byproduct can be attributed to the continued  
504 degradation of malaaxon by the same decontaminant, especially under higher pH conditions  
505 [40]. Similarly, malathion can also rapidly hydrolyze without malaaxon formation at more  
506 alkaline pH levels as present in bleach (pH >11) [41].

507       Degradation of carbaryl is likely to have involved the hydrolysis to 1-naphthol and  
508 methylamine [42] followed by further degradation into more benign byproducts. The current  
509 toxicological profile for carbaryl [43,44] does not identify degradation byproducts of significant  
510 toxicity that should be avoided or minimized in the carbaryl degradation process.

511       Degradation of fipronil in the presence of hydrogen peroxide or hypochlorite is expected to  
512 lead to the intermediate formation of fipronil sulfone and fipronil-desulfinyl (oxidation) and



513 fipronil-amide (hydrolysis) [28,29]. These byproducts were detected in extracts following  
514 decontamination of a fipronil-contaminated surface but also in the not-decontaminated  
515 positive control extracts. There was no noticeable difference in byproduct concentration  
516 between test coupon and positive control coupon extracts which suggests that no additional  
517 byproducts were created during the degradation by bleach or DF200 of fipronil on these  
518 surfaces.

519 The degradation of the pyrethroids deltamethrin and permethrin is expected to follow a  
520 combined hydrolysis and/or oxidation process [45]. The hydrolysis is enhanced under alkaline  
521 conditions such as observed when using bleach. The current toxicological profile for these  
522 pyrethroids [45] does not identify degradation byproducts of significant toxicity that should be  
523 avoided or minimized in the permethrin or deltamethrin degradation process.

524 The use of full-strength bleach without any dilution to clean surfaces is not recommended  
525 under normal conditions. Further research is needed to address whether a diluted bleach  
526 product can still be as efficacious as observed in this study. Such effort would also need to  
527 verify whether the degradation reaction may become incomplete leaving malaoxon as a  
528 persistent toxic degradation byproduct on the surface.

529 The limited improvement in efficacy and associated limited reduction in residual pesticides  
530 on these surfaces by including a second application suggests that the amount of decontaminant  
531 on a molar basis in a single application is enough to reach high efficacy. Efficacy for bleach was  
532 not significantly affected by the presence of other ingredients that are included in the tested  
533 commercial formulations Ortho Max and Sevin. When DF200 was used to decontaminate the  
534 Sevin product on stainless steel, more unreacted carbaryl remained on the surface in

535 comparison to the decontamination of technical grade carbaryl on the same surface. This  
536 suggests that other ingredients in the Sevin product provide demand for the DF200 active  
537 ingredients which prevents a full degradation of carbaryl on the surface. Such loss may be  
538 overcome through reapplication of the decontamination product,

539       Investigations of the efficacy of surface decontaminants, against VX, a highly persistent OP  
540 compound, produced comparable results. Efficacy values for bleach against most of the tested  
541 pesticides were similar or higher than those obtained for VX [24]. In that study, the reported  
542 efficacies were 42-67% after a 24 hr contact time, depending on the material. A 10x diluted  
543 bleach solution with an added surface wetting agent (trisodiumphosphate) was used and the  
544 starting concentration of VX was higher (10 mg/100cm<sup>2</sup> range) in comparison to the pesticide  
545 surface concentrations. This suggests that the use of diluted bleach in the presence of high  
546 levels of contamination may not be as efficacious, however, such decontamination efficacy is  
547 chemical specific and can be material dependent. The VX study also evaluated DF200 and the  
548 efficacy results are similar to those observed in this pesticide study. In the VX study, efficacies  
549 were 75-99% after a 24-hour contact time depending on the material, with one exception of  
550 noticeably lower efficacy for vinyl tile material (20%). Consistent high efficacy values on vinyl  
551 material were observed in the pesticide study.

552       In this study, initial pesticide surface concentrations were representative of the pesticide-  
553 specific levels measured during field investigations involving misapplications of pesticides in  
554 homes or businesses. These concentrations varied widely from 25 µg/100cm<sup>2</sup> (deltamethrin) to  
555 2,400 µg/100cm<sup>2</sup> for carbaryl yielding molar ratios of active ingredient in the decontamination  
556 solution over the pesticide solution ranging from approximately 40 (Sterilex/carbaryl) to over

557 22,000 (DF200/deltamethrin). Although this study was limited to one surface concentration per  
558 pesticide, it is expected that surfaces contaminated with a lower pesticide surface  
559 concentration would be equal or easier to clean as this molar ratio would be larger. For higher  
560 pesticide surface concentrations, the molar ratio may become close to the stoichiometric ratio  
561 likely resulting in lower decontamination efficacy values.

562 The long contact time (18 hr), simulating an overnight drying of the decontamination  
563 solution, allows for degradation reactions to continue potentially longer than if the  
564 decontaminant was rinsed or neutralized shortly after application. However, it is expected that  
565 the eventual drying of the decontaminant through evaporation (occurring in time frame of  
566 several hours; no information collected) will limit further degradation beyond the time of  
567 decontamination solution evaporation. Among the materials tested, residual pesticide amounts  
568 on plywood were in most cases higher than those on stainless-steel and vinyl. In addition, the  
569 variability in recovered pesticide mass from plywood was noticeably higher than those from  
570 stainless-steel and vinyl. This may be due to the nonhomogeneous nature of plywood that can  
571 lead to variations in the degree of pesticide permeation. Water-based decontaminants applied  
572 to the surface would be unable to access pesticides that permeate into the plywood.

## 573 **5. Conclusions**

574 The bench scale decontamination efficacy test results reported here are a first step in  
575 identifying and developing decontamination strategies for indoor environments following  
576 pesticide misuse. Further research is needed to evaluate the potential effects of  
577 decontamination methods used in the field, including material compatibility, safety concerns,  
578 how decontaminant solutions are applied, and the use of physical removal processes, such as

579 scrubbing and/or rinsing of surfaces. Also, the decontamination efficacy values are not  
580 connected to achieving a particular cleanup level or health-based criteria. As noticed when  
581 decontaminating plywood and as observed in other studies [46] for a painted drywall material,  
582 the transport of a pesticide into a permeable material or into paint/coating covering a material  
583 plays a significant role in whether these decontamination technologies are equally successful in  
584 cleaning surfaces that are abundant in an indoor environment. The presence of these  
585 permeable materials in an environment where pesticide misuse occurs may drive the decision  
586 to forego *in situ* cleaning and instead remove the material to meet the health-risk based  
587 cleanup standard. Such clearance goals are pesticide specific and may be depend on the extent  
588 of pesticide application and the potential exposure to the affected indoor surfaces.

589 This study was conducted to provide information to federal, state, tribal, and local agencies  
590 about decontamination approaches for residential or commercial buildings to reduce occupant  
591 exposure following pesticide misuse incidents. The mechanistic details of the degradation  
592 chemistries are pesticide specific, although the same degradation approach may be applicable  
593 by first approximation for the same class of pesticides such as pyrethroids, organophosphate or  
594 carbamates. Decontamination practices should be tailored to the specific pesticide(s) involved  
595 to maximize its efficacy and minimize the risk of transferring contamination within and from the  
596 site. Further, formation of toxic decontamination byproducts should be avoided. This may need  
597 to be evaluated on a case-by case basis.

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607 products or services.

## 608 **Appendix A. Supplementary Data**

609 Supplementary data associated with this article can be found, in the online version, at  
610 *<insert link>*

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761

762 **Tables and Table Captions**

763 **Table 1. Targeted Pesticides**

Pesticide	CAS #	Pesticide Family	Purpose	Target Surface Concentration ( $\mu\text{g}/100\text{cm}^2$ ) <sup>1</sup>
Malathion	121-75-5	Organophosphate	Insecticide	400
Carbaryl	63-25-2	Carbamate	Insecticide	2400
Fipronil	120068-37-3	Phenylpyrazole	Insecticide	150
Deltamethrin	2918-63-5	Pyrethroid	Insecticide	25
Permethrin	52341-32-9	Pyrethroid	Insecticide	500
Ortho® MAX® Malathion Inspect Spray Concentrate	N/A	Organophosphate	Commercial insecticide product (50% malathion)	400
Sevin® Carbaryl Insecticide	N/A	Carbamate	Commercial insecticide product (43% carbaryl)	2400

764 N/A: Not Applicable

765 <sup>1</sup> Based on highest observed surface concentration from specific pesticide misuse cases

766

767 **Table 2. Test Matrix**

Pesticide	Spic&Span	Bleach	DF200	Sterilex
Malathion	X	X	X	X
Carbaryl		X	X	X
Fipronil		X	X	
Deltamethrin		X	X	
Permethrin		X	X	

768

769 **Table 3. Persistence Half-Life values of Selected Pesticides on Stainless-Steel**

Pesticide	Half-Life on SS with Light $\pm$ SD (days)	Half-Life on SS in Dark (days)	Half-Life in Water (days)	Half-Life in Soil (days)	Soil/Water Reference
<b>Malathion</b>	Not measured	Not measured	1.7-17 <sup>1</sup>	1-17	27,30,31
<b>Carbaryl</b>	ND; >500	ND; >500	4 <sup>2</sup>	16 (at surface) -72 (in soil)	32
<b>Fipronil</b>	166 $\pm$ 62	ND; > 500	0.25-0.5 <sup>3</sup>	125	33,34
<b>Deltamethrin</b>	166 $\pm$ 42	ND; > 500		5.7- 209	35
<b>Permethrin</b>	286 $\pm$ 91	ND; > 500	0.8-1.1 <sup>5</sup>	40	36

770 SS: Stainless-Steel

771 ND: Not determined due to lack of decay

772 SD: Standard deviation in fitted half-life value

773 **Table 4. Pesticides Residuals after Decontamination of 10 cm<sup>2</sup> Surfaces.**

Pesticide	Decontaminant	Recovered mass from SS (Mean ± SD, µg) Number of Applications		Recovered mass from plywood (Mean ± SD, µg) Number of Applications		Recovered mass from vinyl (Mean ± SD, µg) Number of Applications	
		1	2	1	2	1	2
Malathion	DF200	0.25*	0.25*	3.8±3.2	3.6±2.2	1.1±0.5	0.6±0.1
	Sterilex	0.25*	0.25*	8.1±3.0	4.7±3.1	1.2±0.1	1.2±0.5
	Spic&Span	37±1.6	34±5.5	18±5.2	19±6.6	30±0.2	30±3.0
	Bleach	0.25*	0.25*	0.91±0.27	0.63±0.41	0.25*	0.25*
Carbaryl	DF200	5.0±0.0	0.08±0.02	37±20	31±22	0.05±0.02	0.04*
	Sterilex	49±10	25±12	129±38	83±32	36±19	3.3±1.9
	Bleach	158±20	97±21	141±41	103±23	122±25	107±28
Fipronil	DF200	0.05*	0.05*	0.97±0.47	1.5±0.95	0.07±0.02	0.056±0.001
	Bleach	0.05*	0.05*	3.9±1.5	3.2±2.5	0.05*	0.05*
Deltamethrin	DF200	0.05*	0.05*	0.24±0.12	0.14±0.09	0.15±0.17	0.17±0.18
	Bleach	0.05*	0.05*				
Permethrin	DF200			16±2.0	14±2.8	20±7.1	15±1.2
	Bleach	4.3±3.4	3.1±1.8				

774 SD: Standard deviation to the Mean

775 \*: Method quantification level (MQL) with no standard deviation; residuals were at or below this value  
776 and not quantified.

777

778 **Figure Captions.**

779 **Figure 1.** Recovered pesticide mass as function of time in dark and indoor light conditions on stainless-  
780 steel. Error bars are one standard deviation from the mean (n=3). Panel A: Carbaryl data. Panel  
781 B: Fipronil data. Panel C: Deltamethrin data. Panel D: Permethrin data.

782 **Figure 2.** Decontamination efficacies for tested decontaminants against malathion. Error bars in efficacy  
783 are one standard deviation from the mean (n=3).

784 **Figure 3.** Decontamination efficacies for tested decontaminants against carbaryl. Error bars in efficacy  
785 are one standard deviation from the mean (n=3).

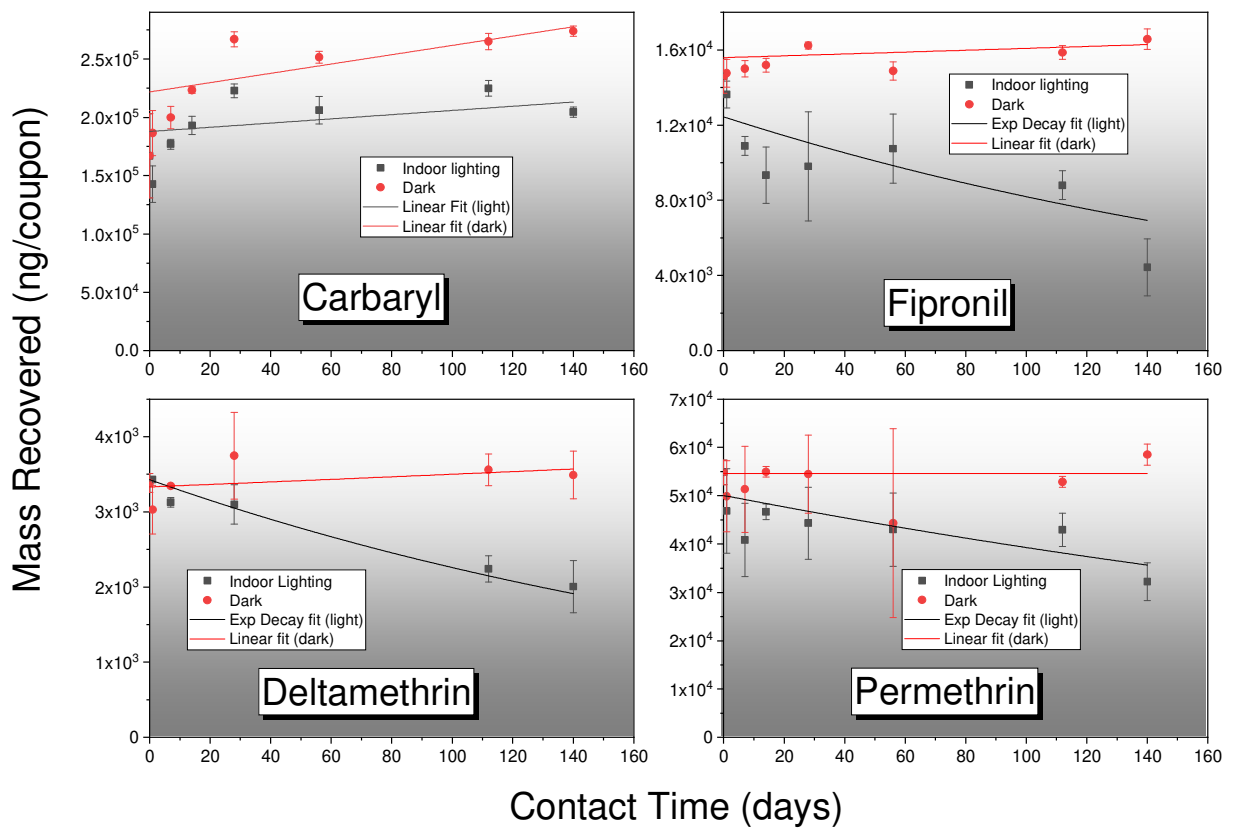
786 **Figure 4.** Decontamination efficacies for tested decontaminants against fipronil. Error bars in efficacy  
787 are one standard deviation from the mean (n=3).

788 **Figure 5.** Decontamination efficacies for tested decontaminants against deltamethrin (upper panel) and  
789 permethrin (lower panel). Error bars in efficacy are one standard deviation from the mean  
790 (n=3).

791 **Figure 6:** Decontamination efficacies for DF200 and bleach for decontamination of stainless-steel  
792 contaminated with malathion and carbaryl using a technical grade standard and commercial  
793 products Ortho MAX and Sevin, diluted to equal concentration of the technical grade standard.  
794

795 **Figures.**

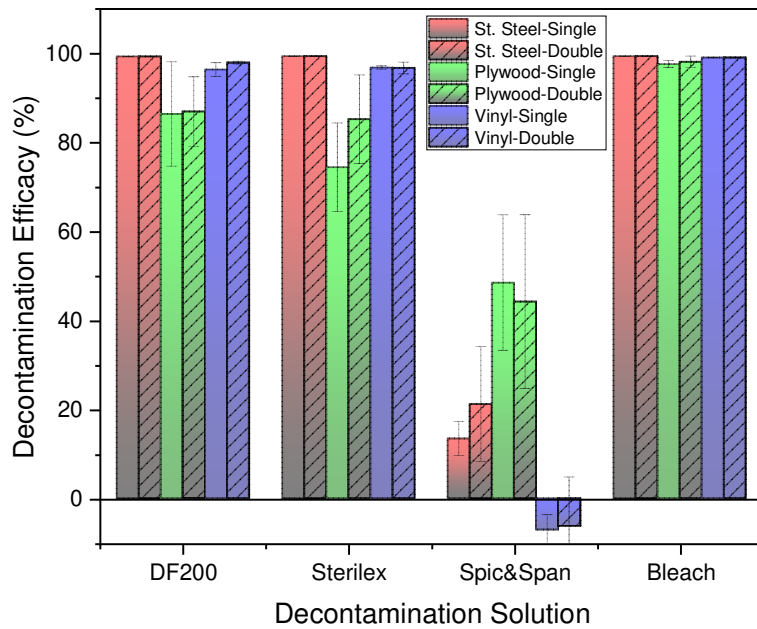
796 **Figure 1.**



797

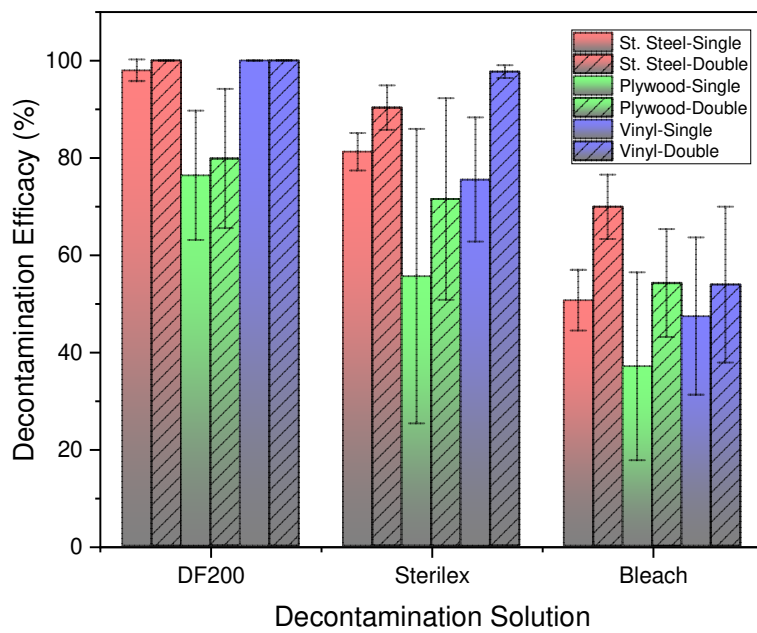
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799 **Figure 2.**



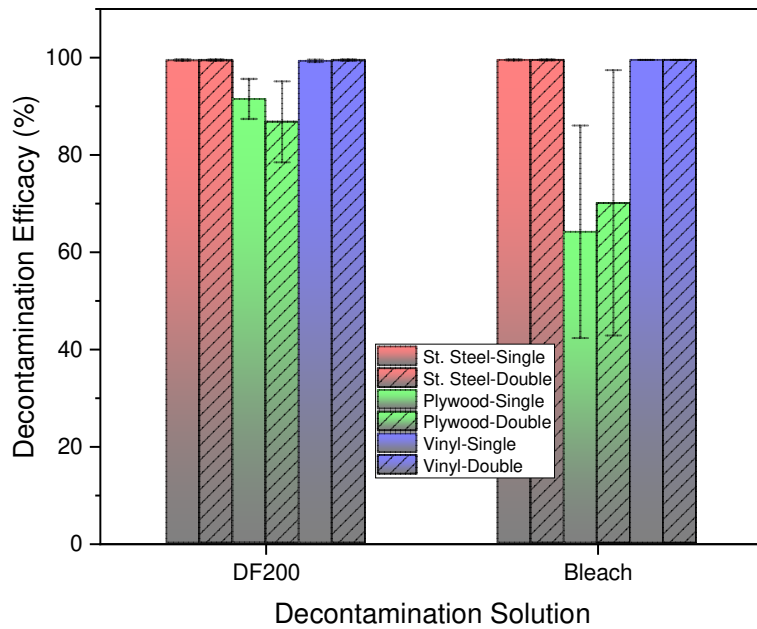
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801 **Figure 3.**

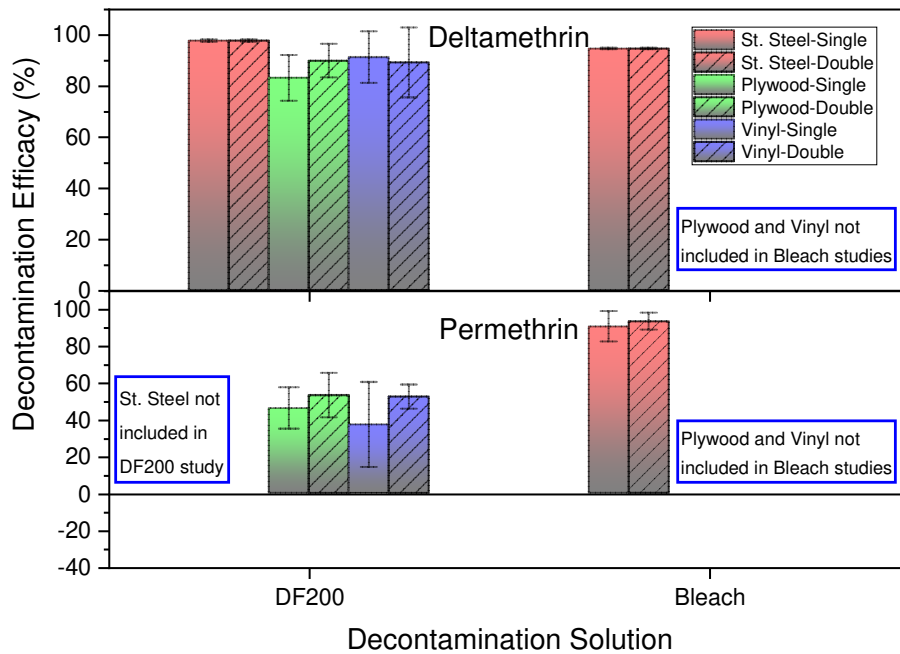


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803 **Figure 4.**

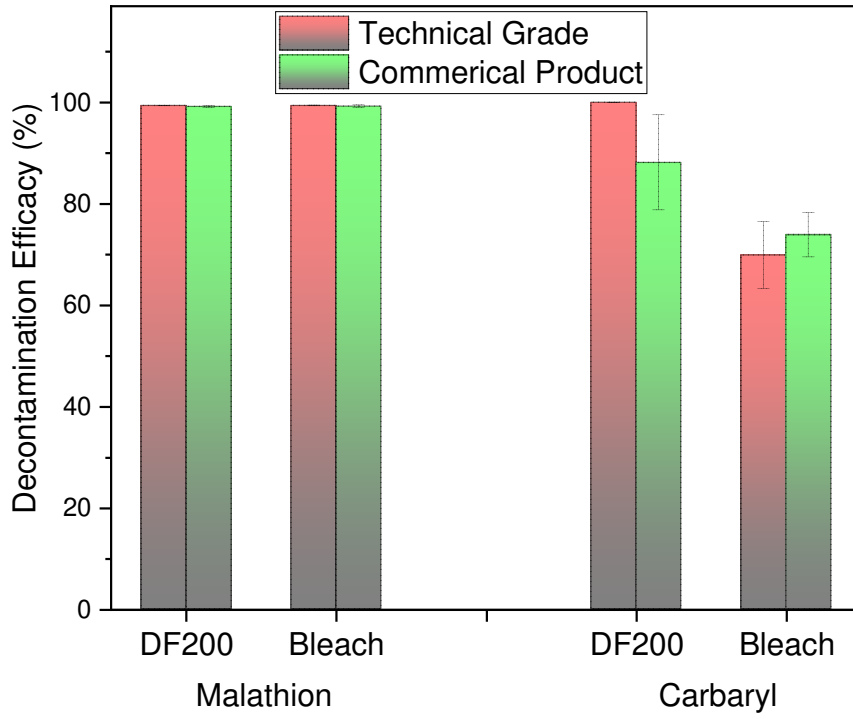


804  
805 **Figure 5.**



806

807 **Figure 6.**



808

809



810 **Supplemental Materials**

811

812 Table A: Extraction Efficiencies

813 Analytical Methods Information

814 Persistence data for fipronil sulfone

815

816 **Supplemental Materials**

817

818 Title: Remediating Indoor Pesticide Contamination from Improper Pest Control Treatments:  
819 Pesticide Persistence Studies and Decontamination Approaches

820

821 Authors: Lukas Oudejans, Amy Mysz, Emily Gibb Snyder, Barbara Wyrzykowska-Ceradini, Joshua  
822 Nardin, Dennis Tabor, James Starr, Daniel Stout II, and Paul Lemieux

823

824 **Extraction Efficiencies**

825 Extraction efficiencies were measured prior to the persistence and decontamination testing as  
826 to verify whether the extraction method was able to recover the pesticide from the surface.  
827 Efficiencies are tabulated in Table A.

828 **Table A: Extraction Efficiencies of Targeted Pesticides from Materials**

Pesticide	Mean Percentage of Pesticide Mass Recovered $\pm$ SD (n=3)		
	Stainless-Steel	Plywood	Vinyl
Malathion	103 $\pm$ 2	96 $\pm$ 16	139 $\pm$ 14
Carbaryl	98 $\pm$ 3	113 $\pm$ 57	79 $\pm$ 8
Fipronil	93 $\pm$ 7	77 $\pm$ 9	75 $\pm$ 3
Deltamethrin	75 $\pm$ 23	51 $\pm$ 14	51 $\pm$ 30
Permethrin	80 $\pm$ 21	47 $\pm$ 17	57 $\pm$ 42

829

830

831 **Description of Analytical Methods**

832 **GC/MS analysis of malathion (decontamination study only)**

833 A 1  $\mu$ L sample of the extract was analyzed by GC/MS using an Agilent 6980 gas chromatograph  
834 coupled with a low-resolution Agilent 5973 mass spectrometer. Injection port temperature was  
835 set at 150  $^{\circ}$ C. Analytes were chromatographically separated using a DB-5MS column  
836 (dimensions, 60-m x 0.25-mm i.d., 0.25  $\mu$ m film thickness) with a GC oven temperature profile  
837 starting at 110  $^{\circ}$ C (1 min hold) to 250  $^{\circ}$ C at 15  $^{\circ}$ C/min (5 min hold), to 300  $^{\circ}$ C at 15  $^{\circ}$ C/min (10  
838 min hold). The mass spectrometer was operated in selective ion monitoring (SIM) mode  
839 collecting chromatograms at masses 127 m/z and 158 m/z (malathion identification), 173  
840 (malathion quantification), 183 (malathion-d10 internal standard), and 99 m/z (malaoxon).

841 **LC-MS/MS analysis of malathion (persistence tests only), carbaryl, fipronil, deltamethrin, and**  
 842 **permethrin**

843 Analysis of malathion, carbaryl, permethrin, deltamethrin, fipronil, fipronil sulfone, fipronil  
 844 amide, and fipronil desulfinyl was done using an Agilent (Santa Clara, CA) model 1100 LC with  
 845 an AB Sciex (Foster City, CA) API 4000 tandem mass spectrometer. All compounds were  
 846 separated using an Agilent Zorbax® C18 column (3.5  $\mu$ m, 3 x 150 mm) and a mobile phase flow  
 847 rate of 400  $\mu$ L/min. Compound specific LC-MS/MS settings are provided in Table B:

848 **Table B: LC-MS/MS settings**

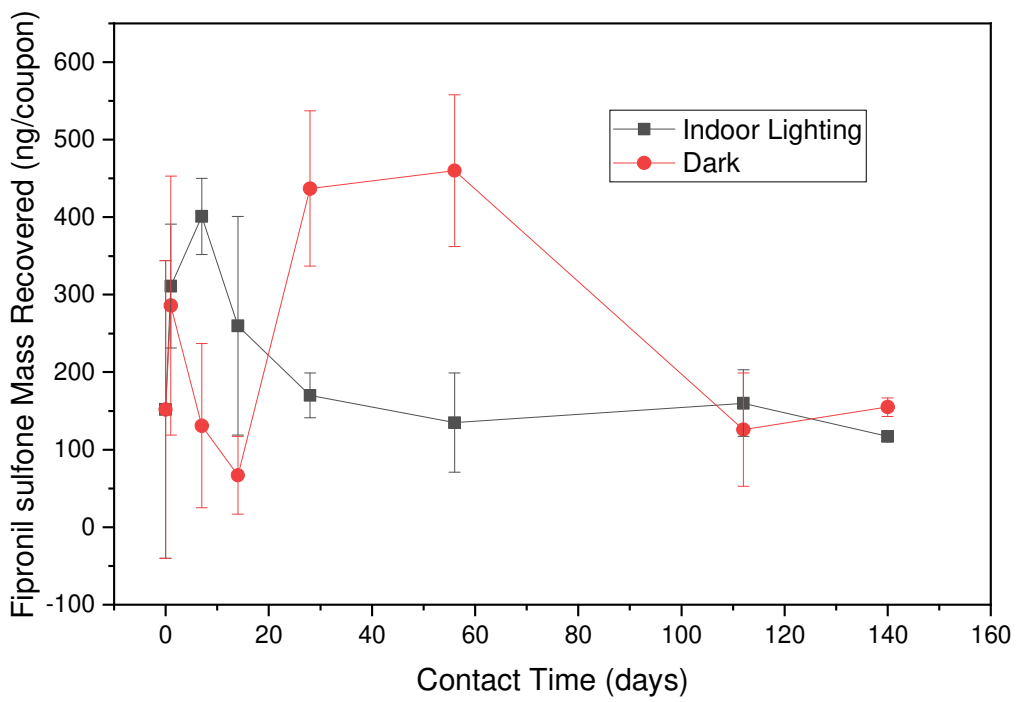
Pesticide	Ionization Mode <sup>1</sup>	Mobile phase (A:B) <sup>1</sup>	T <sub>r</sub> <sup>2</sup> (min.)	Q1 <sup>3</sup>	Q2 <sup>3</sup>	Internal Standard
malathion	M+18	20:80	3.4	348	127	malathion-D <sub>10</sub>
carbaryl	M+18	20:80	2.5	219	145	<sup>13</sup> C <sub>6</sub> carbaryl
<i>cis</i> -permethrin	M+18	2:98	3.2	408	183	<sup>13</sup> C <sub>6</sub> <i>cis</i> - permethrin
<i>trans</i> -permethrin	M+18	2:98	3.5	408	183	
deltamethrin	M+18	2:98	2.7	521	279	deltamethrin D <sub>6</sub>
fipronil	M-1	15:85	2.9	435	330	fipronil des F <sub>3</sub>
fipronil sulfone	M-1	15:85	3.0	451	282	
fipronil amide	M-1	15:85	2.3	453	348	
fipronil desulfinyl	M-1	15:85	2.8	387	351	

- 849 1. A = 5 mM ammonium acetate in water. B = methanol.  
 850 2. Retention time.  
 851 3. Q1 and Q2 are the first and second ion mass filters, respectively.

852  
 853

854 **Persistence of fipronil sulfone**

855  
 856 Figure 1 shows the amount of fipronil-sulfone recovered from stainless steel coupons as function of time  
 857 under dark and indoor light conditions. Fipronil-sulfone is a degradation product of fipronil and was  
 858 present in the spiked fipronil solution on Day 0.  
 859

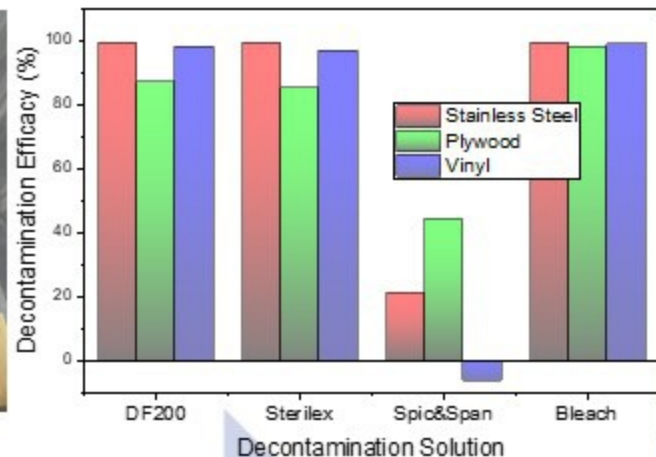
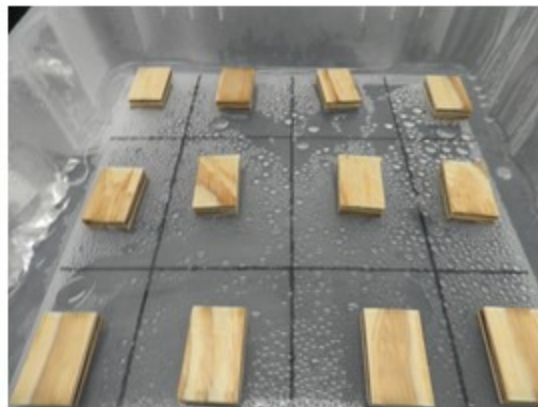


860  
 861 **Figure 1: Recovered fipronil sulfone mass as function of time in dark and indoor light conditions on**  
 862 **stainless-steel. Error bars are one standard deviation from the mean (n=3).**

## Indoor Pesticide Misuse Cases



## Bench-Scale Decontamination Research

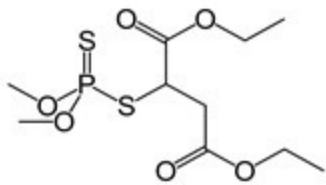


Pesticide Contamination

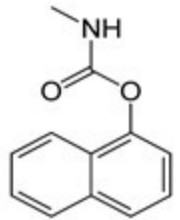
High Persistence Observed

Cleanup Needed

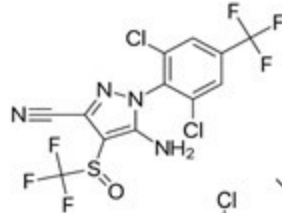
Return to Normal Use



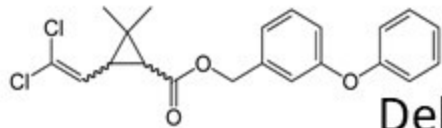
Malathion



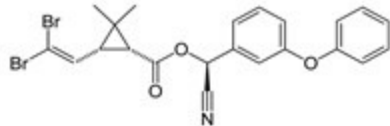
Carbaryl



Fipronil



Permethrin



Deltamethrin

