Dear Chemosphere Editors,

The two corrections born by the paper “From e-waste to living space: Flame retardants contaminating household items add to concern about plastic recycling”, by Liu M, Brandsma SH, Schreder E., *Chemosphere*. 2024 Oct 1;365:143319 still fail to completely correct the math and methodological errors present in the study. The restated median potential exposures in the second corrigendum are still overstated. The errors are sufficient to warrant a restating of the abstract, sections of the paper and conclusions, if not a retraction. The results show that, while there is potential for contamination coming from recycled content, the levels of phased-out flame retardants are low and the chance for significant exposure is similarly low.

The paper states that the reason for the study was “to determine whether black plastic household products sold on the U.S. market contained emerging and phased-out flame retardants (FRs) and whether polymer type was predictive of contamination,” The abstract and math performed in the work tell a different story. The study and subsequent press releases address the likely exposures caused by the presence of flame retardants and compare them to reference dose levels in drawing the conclusion that there is significant contaminations. In spite of the stated goal, the analysis performed addresses whether black plastic presents an unacceptable risk due to presence of flame retardants brought in through recycling. Determination requires two pieces of information, the level of exposure that represents acceptable risk and whether exposure from use of black plastic articles exceeds that level. Math errors and methodological errors persist in the second corrigendum. Sadly, the original study gets both pieces wrong, and not a little wrong. Rather than having exposures to BDE-209 nearly identical to intake from dust and diet, they are at least 800 times lower.

The analyses presented here rely on data extracted from the paper and supplemental materials. Table 1 pulls data from Table 1 of the original paper and merges with data extracted from Table S6. Table 2 also uses data from Table S6. Analysis in Table 2 immediately converts concentration to exposure following the methodology used in the paper and described below. The original study and correction 2 average and incorrectly eliminate concentration data before estimating exposure. This is one source of methodological error still present in the revised study.

The original paper incorrectly identified the level of current exposure, the reference dose, by a factor of 10, identifying the typical exposure as 42 μg/day rather than the correct value, 420 μg/day. This error was noted in the first corrigendum. The finding of 34,700 ng/day was deemed too close to the typical exposure leading to the conclusion stated in the last line of the abstract is “estimation of exposure to BDE-209 from contaminated kitchen utensils indicated users would have a median intake of 34,700 ng/day, exceeding estimates for intake from dust and diet.” The EPA reference dose was used as the intake from dust and diet. Exposure did not exceed the reference dose in the original work. Calculated exposure was only 80% of the reference does. Corrigendum 1 reduced the exposure to only 8%, deemed insufficient to retract the study. The value reported for exposure, 34,700 ng/day has layers of errors. It is reported as the median intake from kitchen utensils. It is, in fact, the mean of all 20 samples subjected to MS analysis. These samples include hair care, toy and serving ware. The second correction lowers this dose to 7.9 μg/day, less than 2% of the expected intake from dust and diet. These errors were again deemed insufficient to retract the study. Analyses presented here shows the value is 527 ng/day or lower, over 65 times lower than the original report and approximately 0.1% of the intake from dust and diet. The measured value is now 800 times lower than the expected intake. This constitutes a major methodological and mathematical

see Chemosphere 385 (2025) 144542 https://doi.org/10.1016/j.chemosphere.2025.144542
Received 17 February 2025; Received in revised form 13 May 2025; Accepted 21 June 2025
Available online 4 July 2025

error. A major restatement of the abstract, conclusions and several sections of the paper is required, if not a full retraction of the study.

The most egregious errors are only partially addressed in the second corrigendum. The error prompting a letter to the editor is application of a correlation developed for exposure to hot oil for all samples, whether or not the use pattern involves exposure to hot oil. The second corrigendum acknowledges this error. Application of the hot oil correlation where no hot oil exposure is expected grossly overstates the exposure for toys, hair products and many kitchen items. Correction requires recalculation of the values presented in the paper and the supplemental materials. The correlation derives from the work of Kuang et al. [Kuang J, Abdallah MA, Harrad S. Brominated flame retardants in black plastic kitchen utensils: Concentrations and human exposure implications. *Science of The Total Environment*. 2018 Jan 1;610:1138-46. doi.org/10.1016/j.scitotenv.2017.08.173.]. In addition to providing the hot oil correlation, it also addresses items that are only handled. From Kuang at the end of Section 3, "results indicate that human update of PBDEs via dermal contact with cooking utensils is much lower than our intake estimates based on cooking and other pathways."…"Therefore, our finding suggest when using BFR-contaminated kitchen utensils, exposure is dominated by the utensil-oil transfer rather than utensil-skin transfer". Kuang addresses contaminated handles and concludes the transfer is negligible.

This mythological error is easily addressed by excluding those items not intended for use in hot oil. The original study applied the hot oil correlation to all 20 samples in the MS cohort. This is clearly in error since toys, serving items and hair care items are not used in hot oil. Some kitchen items, such as peelers are also not used in hot oil. Others are more ambiguous. Clearly the slotted turners, the spatulas, are an example of an item where hot oil exposure is likely. Spoons are unlikely to be used in hot oil and have been excluded from the reanalysis presented here. The second corrigendum reduces the samples used in the analysis to a subset only of the kitchen items. Severe errors remain. Only 4 samples contribute to the calculation of the median exposure. Included in the analysis are the following samples:

S5	slotted turner
S7	basting spoon
S9	pasta server
S10	slotted spoon

Excluded from the revised analysis are:

S4	slotted spoon
S6&8	slotted turner
S11&12	peeler

Three of the excluded items have undetectable FR levels. The peelers have a use pattern that estimates exposure from them as zero. They should not be excluded from the calculation of the median and mean exposures. Their exclusion is mentioned in the corrigendum but never justified. Their inclusion in calculation of the median and mean values is shown in Table 2 as "kitchen only – ng/day". The median value is 0, not 7900. The average exposure drops to 3480 ng/day, less than half of the revised exposure. Continued inclusion of items that do not encounter hot oil during use remains problematic. Analysis using only those items used in oil, S5, S6 and S8, yields a median exposure of 0 and an average exposure of 527 ng/day. This is Tabel 2 the "oil only -ng/day" row. This is well below the value reported in the second corrigendum.

The second corrigendum also provides information that further calls into question the methodology. It indicates the four samples used in the estimate only had handles analyzed. If this is true, the expected

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exposure for all kitchen items based on Kuang is zero. The exposure is well below the typical exposure from dust and diet.

The second corrigendum goes on to question all of the methodology used, arguing the uncertainties are too great to draw conclusions and is only "provided to contextualize potential exposure." These statements alone require restating the conclusions present in the abstract of "exceeding estimates for intake from dust and diet." Similarly, if the error is sufficiently large to only provide context, the statement in the Conclusions that brominated flame retardants "significantly contaminate products" no longer can be supported and must be corrected or retracted following the reasoning presented in the second corrigendum.

Determining what samples should be included remains an issue in the second corrigendum. The calculations do not include those items sampled only with XRF and found to have concentrations that would lead to negligible exposures. The study shows in Figure 2 that items with levels below 50 ppm by XRF will not have detectable FR concentrations. 109 kitchen items and 28 food service items were analyzed yet most were ignored in the original and corrigendum analyses. Inclusion of these items is shown as line starting with "Full Set"in Table 2. Median concentrations, expectedly, are zero. Looking only at food service items, the estimated average exposure drops to 14.5 ng/day.

The abstract must be restated due to the multiple errors present. The statement "FRs were found in 85% of analyzed products" was never correct as it neglected most of the 203 items collected. As Table 1 shows, the actual value is 8%. "Estimation of exposure to BDE-209 from contaminated kitchen utensils indicated users would have a median intake of 34,700 ng/day, exceeding estimates for intake from dust and diet" is incorrect. Values are well below intakes from dust and diet. Further, while the study includes the statement "Overall, screening indicated that about 10% of the products likely contained BFRs at levels to be expected from use of FR-containing recycled content." , this is not mentioned in the abstract or conclusion. The analysis shows the likelihood of significant exposure is low both because most products are not contaminated and even when contamination is present, exposure in excess of diet and environmental exposures is unlikely.

The conclusion section of the paper states: "These results show that when toxic additives are used in plastic, they significantly contaminate products, made with recycled content, that do not require flame retardancy. Products found in this study to contain hazardous flame retardants included items with high exposure potential, including food-contact items as well as toys." The revised results, even without the corrections suggested here, no longer support stating there is significant exposure potential. The predicted exposures are well, well below the expected exposure from dust and diet.

This study caught the public's fancy and is now widely reported. Many spatulas have met an untimely end based on the flawed analysis. The ramifications run much deeper. Recycling requires a level of trust exceeding what is required for virgin material. Testing for all possible materials of concern in a recycle stream is impossible. We must trust that those in the recycling industry act responsibly. It is not unlike the trust we place in the food supply chain where contamination is always a risk. Hazards can clearly be present. What matters is whether the hazards are recognized and steps taken to ensure levels are maintained at levels that don't present an unreasonable risk. The initial study and both corrigendums conclude there is a need for regulation. This is not supported by the analysis and is also not a science-based conclusion. Flame retardants in recyclate are a hazard. Steps to manage the risk to acceptable levels are required. Regulation is one way to manage risk but is not the only way. The study demonstrates the

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risks are being managed. The miscalculation of the reference dose, the misuse of the hot oil correlation of concentration to exposure, the ignoring of most of the collected and analyzed samples, and flawed data analysis invalidates the study's conclusions. The abstract, conclusions and multiple sections of the study need significant revision. The data show that the risks are present well below levels of concern, yet the conclusions state the opposite, that significant concerns exist. The paper must be further corrected or, preferably, retracted.

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Table 1: Summary of total items sampled by both XRF and MS.

Product Category	Number of Items Tested By XRF	% of Products >50 mg/kg Br	MS Tested	# FR Detected In MS Cohort	% of Products with FR Detected By MS	% of Total Products With FR Detected
Food Serviceware	28	7%	2	2	100%	7%
Hair Accessories	30	3%	1	1	100%	3%
Kitchen Utensils	109	8%	9	8	89%	7%
Toys	36	22%	8	5	63%	14%
Total Samples	203		20	16	80.0%	8%

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Table 2: Analysis of food contact items by MS for BDE-209. Measured concentrations are reported on the top line of the table labeled mg/kg. All other numerical values are exposures consistent with the Kuang analysis. Corrigendum 2 shows the analysis done in Corrigendum 2. It ignores samples with an undetectable level of BDE-209. Samples under MS Cohort only correctly include those items with undetectable levels of BDE-209 in the calculation of mean and median. The oil-only calculation includes only those items designed for use in hot oil. Full set includes the samples analyzed by both XRF and MS.

	Sushi Tray	Fast Food Tray	Slotted Spoon	Slotted Turner	Slotted Turner	Basting Spoon	Slotted Turner	Pasta Server	Slotted Spoon	Peeler	Peeler	median	mean
BDE-209	S1	S2	S4	S5	S6	S7	S8	S9	S10	S11	S12		
mg/kg	11900	<2.0	<4.0	50	<30	200	<30	300	440	130	3570		
appropriate to use hot oil correlation of Kuang	N	N	N	Y	Y	N	Y	N	N	N	N		
Corrigendum 2 - ng/day				1580		6320		9480	13900			7900	7820
MS Cohort Only													
all food - ng/day	0	0	0	1580	0	6320	0	9480	13900	0	0	0	2840
kitchen only - ng/day			0	1580	0	6320	0	9480	13900	0	0	0	3480
oil only - ng/day				1580	0		0					0	527
Full Set Kitchen and Service	0	0	0	1580	0	0	0	0	0	0	0	0	11.5
Full Set Kitchen			0	1580	0	0	0	0	0	0	0	0	14.5

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