Measurement of endocrine disrupting and asthma-associated chemicals in hair products used by Black women

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A R T I C L E   I N F O

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Consumer product
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A B S T R A C T

Background: Personal care products are a source of exposure to endocrine disrupting and asthma-associated chemicals. Because use of hair products differs by race/ethnicity, these products may contribute to exposure and disease disparities.

Objective: This preliminary study investigates the endocrine disrupting and asthma-associated chemical content of hair products used by U.S. Black women.

Methods: We used gas chromatography/mass spectrometry (GC/MS) to test 18 hair products in 6 categories used by Black women: hot oil treatment, anti-frizz/polish, leave-in conditioner, root stimulator, hair lotion, and relaxer. We tested for 66 chemicals belonging to 10 chemical classes: ultraviolet (UV) filters, cyclosiloxanes, glycol ethers, fragrances, alkylphenols, ethanolamines, antimicrobials, bisphenol A, phthalates, and parabens.

Results: The hair products tested contained 45 endocrine disrupting or asthma-associated chemicals, including every targeted chemical class. We found cyclosiloxanes, parabens, and the fragrance marker diethyl phthalate (DEP) at the highest levels, and DEP most frequently. Root stimulators, hair lotions, and relaxers frequently contained nonylphenols, parabens, and fragrances; anti-frizz products contained cyclosiloxanes. Hair relaxers for children contained five chemicals regulated by California’s Proposition 65 or prohibited by EU cosmetics regulation. Targeted chemicals were generally not listed on the product label.

Conclusions: Hair products used by Black women and children contained multiple chemicals associated with endocrine disruption and asthma. The prevalence of parabens and DEP is consistent with higher levels of these compounds in biomonitoring samples from Black women compared with White women. These results indicate the need for more information about the contribution of consumer products to exposure disparities. A precautionary approach would reduce the use of endocrine disrupting chemicals in personal care products and improve labeling so women can select products consistent with their values.

1. Introduction

People are exposed to a wide range of endocrine disrupting chemicals (EDCs) and asthma-related chemicals from consumer products (Dodson et al., 2012; Rudel et al., 2010). Personal care products are a significant source of exposure to phthalates, phenols, and parabens (Berger et al., 2018; Branch et al., 2015; Braun et al., 2014; Fisher et al., 2017; Guo and Kannan, 2013; Harley et al., 2016; Meeker et al., 2013; Parlett et al., 2013; Philippat et al., 2015a). Exposure to chemicals from personal care products is thought to occur primarily via absorption through skin and inhalation (Ernstoff et al., 2016). Urinary levels of these chemicals vary by race/ethnicity, with higher urinary levels of some phthalates and parabens in U.S. Black women compared to U.S. White women (Branch et al., 2015; Calafat et al., 2010; James-Todd et al., 2017; Varshavsky et al., 2016). Racial/ethnic differences in the use of personal care products may contribute to observed exposure disparities.

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Assessing exposures to EDCs and asthma-associated chemicals for Black women is important given the higher incidence of hormone-mediated diseases (James-Todd et al., 2016a) and higher prevalence of asthma (CDC, 2015) in Black women and children. When considering conditions potentially impacted by EDCs, U. S. Black women have higher rates of obesity (May et al., 2013), diabetes (Beckles and Chou, 2013), and pre-term birth (Martin and Osterman, 2013) than U.S. White women. In addition, Black women experience earlier menarche (Cabrera et al., 2014), and more prevalent fibroids (Jacoby et al., 2010) and infertility (Chandra et al., 2013), including poor in vitro fertilization (IVF) outcomes (McQueen et al., 2015). The incidence of breast and endometrial cancers in Black women is increasing, and Black women experience more aggressive forms of these cancers (Cote et al., 2015; DeSantis et al., 2016).

Recent research suggests that EDCs in common consumer products, including hair products, contribute to these hormone-mediated diseases. For example, use of hair oil and hair relaxers is associated with earlier menarche (James-Todd et al., 2011; McDonald et al., 2018), higher incidence of fibroids (Wise et al., 2012), and increased risk of breast cancer (Brinton et al., 2018; Heikkinen et al., 2015; Llanos et al., 2017). Higher urinary concentrations of phthalate metabolites have been associated with fibroids (Huang et al., 2014; Kim et al., 2017, 2016), earlier indicators of puberty (Watkins et al., 2014), less successful IVF outcomes (Hauser et al., 2016) and preterm birth (Ferguson et al., 2017, 2014). However, other studies have not found some of these associations (Mendelsohn et al., 2009; Pollack et al., 2015; Weuve et al., 2010).

Market and exposure research suggest that differences in personal care product use may contribute to higher observed levels of associated consumer product chemicals in Black women (James-Todd et al., 2017). African-American women spend more money on fragrances, feminine hygiene, and deodorizing products than other US women (Nielson, 2013), potentially resulting in greater product use and exposures. Use of fragrances was positively associated with urinary metabolite levels of diethyl phthalate (DEP) in a study of pregnant African-American and Dominican women in New York City (Just et al., 2010), and data from the National Health and Nutrition Examination Survey (NHANES) showed that reported use of vaginal douches, which are heavily marketed to Black women as aesthetic products (Ferranti, 2011), was associated with higher urinary metabolite levels of DEP in Black women (Branch et al., 2015).

Hair products are of particular interest as a potential source of exposure and health disparities (Stiel et al., 2016). Certain hair products are more commonly used by Black women, including straightening and moisturizing hair products (James-Todd et al., 2012; Nielson, 2013; Taylor et al., 2017) that are often used to meet social beauty norms (Zota and Shamsanuder, 2017). Black women and children use hormone-containing hair and skin products more often than White women and children (Donovan et al., 2007; Li et al., 2002; Tiwary and Ward, 2003) and some professional hair straightening products contain and release substantial amounts of formaldehyde (Pierce et al., 2011). Testing of Black women’s hair products has been limited to one study that tested four products for estradiol, estritol, and estrone (Tiwary and Ward, 2003), and another that measured estrogenic and anti-estrogenic activity (Myers et al., 2015). Consumer product chemicals have not previously been measured in Black women’s hair products.

To learn whether hair products used by Black women differ from products tested previously (Dodson et al., 2012; Guo and Kannan, 2013; Horii and Kannan, 2008; Liao and Kannan, 2014), we characterized the concentrations of 66 EDCs and asthma-associated chemicals in six types of hair products commonly used by Black women. We present results by chemical class, for comparison to previous work, and by product category. To determine whether product ingredient labels provide sufficient information to avoid chemicals of concern in hair products, we evaluated the concordance between the labeled ingredients and the measured concentrations. We also compare detected chemicals with regulated restrictions on ingredients. This study provides some of the first data on measured concentrations of chemicals in products used by Black women.

2. Methods

2.1. Product selection

We tested 18 products in six hair product categories: hot oil treatment, anti-frizz/polish, leave-in conditioner, root stimulator, hair lotion, and relaxer. Hot oil treatments are used periodically before shampooing to make hair smoother and stronger, whereas anti-frizz products are used anytime to smooth hair and give it shine. Leave-in conditioner and hair lotions are used after showering or as needed to condition and moisturize hair, and root stimulators advertise increased hair growth and strength. The root stimulator products we tested range from a “treatment” serum to products that are used like hair lotions. These product types are most often used one to seven times a week, while hair relaxers are used several times a year to permanently straighten hair (James-Todd et al., 2012). We purchased products in 2008 as part of a larger product study and focus in this analysis on one leave-in conditioner, one hot oil treatment, and multiple products for anti-frizz, root stimulators, hair lotions, and hair relaxers. A higher percentage of Black women report using these six types of hair products compared with White and Hispanic women (James-Todd, 2008).

We selected specific products for testing based on surveys of product use. We relied primarily on results from a 2004–2005 convenience sample of 301 women in New York City, 54% of whom were Black (non-Hispanic African-American or African-Caribbean) (James-Todd et al., 2012). We selected the 12 most frequently used products (at least five percent of survey respondents reported using these products) and 6 products that were frequently used by Black women, but not others, in this survey (James-Todd, 2008). A 2007 survey of pregnant women in the National Collaborative Perinatal Project (James-Todd, 2008) included nine of the tested products and confirmed that each was predominantly and/or commonly used by Black women (James-Todd, 2008). We obtained product names through personal communication (James-Todd T). Product names and manufacturers are in Supplemental Material Table 1.

2.2. Chemical analysis

We targeted 66 chemicals for analysis based on evidence of their association with endocrine disruption or asthma, their expected presence in consumer products, and analytical capacity. Chemical analysis details were described in (Dodson et al., 2012), and health evidence is provided in Supplemental Table 2. In brief, samples were extracted with 3:1 dichloromethane/methanol and analyzed using gas chromatography/mass spectrometry (GC/MS) with the internal standard method of quantification. A separate aliquot of the extract was derivatized and analyzed for phenolic compounds via GC/MS. We aimed to detect chemicals present at >0.0001% (1 μg/g) in the products. The 6-point calibration curve (0.15–25 μg/mL) spanned an equivalent product concentration range of 0.003–0.5% (0.03–5 mg/g); extract concentrations that exceeded this range were diluted and reanalyzed. Samples were extracted and detected as part of the second analytical round of this prior analysis (Dodson et al., 2012).

To control against potential false positives, we only reported samples over the Method Reporting Limit (MRL). We defined the MRL as the maximum of the analytical level of detection (LOD) and the 90th percentile of the method blank (n = 5) concentrations. When there was
no detectable level of an analyte in the blank, the LOD and MRL were 1.0 μg/g (0.0001%). Fourteen of 66 chemicals were detected in blanks. For two chemicals (4-t-nonylphenol and octylphenol diethoxylate), levels in the blanks were at or below the 1 μg/g LOD. For the remaining chemicals, the 90th percentile of the blank levels (and resulting MRL) was less than 2.5 μg/g, except for decamethylcyclopentasiloxane (D5, 17.2 μg/g), octamethylcyclotetrasiloxane (D4, 5.2 μg/g), and benzophenone-3 (BP-3, 4.8 μg/g). For each analyte, we corrected results by subtracting the median blank value when at least 75% of blanks had detectable values. Only D5 values were corrected, resulting in a maximum −100% change to the reported concentration of D5. All reported detects of D5 were detected at concentrations higher than the maximum found in the blanks.

To assess precision, we analyzed five samples in duplicate. The relative percent difference between duplicates was generally <50%. To assess accuracy on a sample-by-sample basis, we fortified each sample with 5 labeled surrogate compounds containing functional groups found in the target analytes, and measured their recoveries. We also spiked six representative products with the full panel of chemical analytes. Median percent recoveries of all surrogates were within the 50–150% acceptance range. Median spike recoveries across products were within 50–150% except for benzophenone-2, monoethanolamine, and diethanolamine, which had recoveries in the range of 9–22%. Therefore, the results presented for these three compounds may underestimate the true concentrations. Quality control results are summarized in the supplemental material in (Dodson et al., 2012).
Most of the products comprised one material; however, hair relaxer kits comprised several component materials to be used in sequence. We tested kit components from three relaxer kits (#1, 2, and 3). Kits #1 and #3 were no-lye formulations marketed to children. Each hair relaxer kit contained relaxer cream, activator, shampoo, and conditioner. Relaxer kit #1 also contained a protective gel, pre-relaxer treatment, and hair lotion; relaxer kit #3 contained a second conditioning product. To cost-effectively evaluate typical exposures from hair relaxer kits we composites the kit shampoo and conditioners into a single sample for each kit, but tested and reported other kit components separately. Compositing gives us a representative concentration of analytes in product components that are intended to be used together but limits our ability to detect an unusually high or low concentration in a single component.

2.3. Data analysis

Data analysis was exploratory and descriptive, because the number of products tested was too small for statistical testing. We visualized results using a “heat map” approach in which values > MRL are represented in shades of purple. We also report summary statistics (median, maxima and detection frequencies). Six of the 66 targeted chemicals were never detected in these or any products in (Dodson et al., 2012) and were dropped from the data visualization and reporting, including summary statistics.

Since hair relaxer kits contain several components, we summed the detected concentrations in all the component products within each relaxer kit, including detects < MRL. For each chemical, we used the highest MRL from the component products as the maximum MRL, and only reported summed values greater than this maximum MRL.

To check whether chemicals were reliably listed on the product label (and thus whether the product label can be used to avoid certain chemicals), we compared the 45 chemicals found in the products with the chemicals listed on ingredient labels of all 15 non-relaxer hair products and 13 relaxer kit components tested (each component in each relaxer kit was labeled with ingredients, so each component or shampoo/conditioner composite was evaluated as a separate product).

3. Results

We detected 45 of the 66 target chemicals, including chemicals from every chemical class tested (Fig. 1 and Supplement Table 3). Products contained more cyclosiloxane, fragrance chemicals, diethyl phthalate (DEP), and parabens than chemicals from other chemical classes (Fig. 1), and we detected methyl paraben, all three cyclosiloxane chemicals, and DEP most frequently and at the highest levels (Fig. 1, Table 1). The fragrance chemicals linalool, limonene, and 1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta-2-benzopyran (HHCB, or Galaxolide) were also found at higher detection frequencies and concentrations compared to other target chemicals. The majority of products contained an ultraviolet (UV) filter. We found nonylphenols in 30% of products, and antimicrobials, BPA, ethanolamines, glycol ethers, and higher molecular weight phthalates in fewer than 20% of products. Root stimulators, hair lotions, and relaxers frequently contained nonylphenols, parabens, and fragrance chemicals whereas anti-frizz products contained cyclosiloxanes. We did not find strong agreement between detected and labeled chemicals, although chemicals detected at higher concentrations were labeled more often.

3.1. Test results by chemical class

3.1.1. Parabens

Most (78%) of the products contained parabens, which are used as preservatives. We found methyl paraben most frequently (72% of products) and typically at concentrations greater than 100 μg/g (median 800 μg/g). The highest paraben levels were in hair lotions: two (of six) hair lotions had methyl paraben concentrations over 2000 μg/g (0.2% by weight). Ethyl and butyl paraben were always detected together or with methyl paraben, and all detected concentrations were below 100 μg/g.

3.1.2. Phthalates

We found the lower molecular weight phthalate DEP, commonly used as a solvent in fragrances (ATSDR, 1995) and fragranced products (Dodson et al., 2012), in 78% of hair products, and at higher concentrations (median = 47 μg/g, maximum = 2450 μg/g) than other phthalates and most other targeted chemicals. Six products contained DEP concentrations > 100 μg/g including one hair lotion with 0.25% by weight DEP. Four products contained bis(2-ethylhexyl) phthalate (DEHP), di-n-octyl phthalate (DOP), di-n-propyl phthalate (DPP), or benzylbutyl phthalate (BBP), higher molecular weight phthalates used in products as solvents and plastic additives, and DEHP was detected in three of these. One no-lye relaxer kit for children (kit #1) contained 90 μg/g DEHP, which was primarily from the activator component. Other higher molecular weight phthalate concentrations were between 4 and 20 μg/g. We detected the phthalate substitute bis(2-ethylhexyl) adipate (DEHA) in only one product: the no-lye relaxer kit #3 marketed to children.

3.1.3. Cyclosiloxanes

The majority (67%) of products contained at least one of three targeted cyclosiloxanes, which are added to products for conditioning and spreadability. Eight products contained a mixture of all three cyclosiloxanes and two products had just one cyclosiloxane. We found decamethylcyclopentasiloxane (D5) at the highest concentration of any chemical in our study: three anti-frizz/polish products contained D5 at 380,000–460,000 μg/g, or 38–46% by weight. The same products also contained D4 and D6 at higher levels: D4 in the range of 480–2600 μg/g, and D6 at 640–17,000 μg/g. One hair relaxer kit also had combined D5 concentrations over 1000 μg/g.

3.1.4. Fragrances

All products contained at least one of the 19 targeted fragrance chemicals, and most had multiple fragrances, with several root stimulators containing over half of the fragrances targeted. Linalool, limonene, and HHCB (or Galaxolide) were the most common (found in about 40% of products). We found limonene at the highest concentration among fragrance chemicals (1900 μg/g, or 0.19% by weight), and limonene, linalool, phenethyl alcohol, and terpineol were frequently detected at > 100 μg/g in multiple products. We did not detect the synthetic nitro-musks (musk xylene and musk ketone), synthetic fragrance DPMI, and the natural fragrance chemicals methyl eugenol and pinene.

3.1.5. UV filters

Most (72%) products contained at least one UV filter, which are used as active ingredients in sunscreens and to protect products from degradation. Although one hair relaxer contained three UV filters, nine products had only one of the seven targeted UV filters, suggesting these targeted UV filters are not typically used in combination. We found octinoxate (octyl methoxycinnamate) at the highest concentration among UV filters (650 μg/g in an anti-frizz product), and octyldimethyl PABA at the next highest level (210 μg/g in a hair lotion). We found benzophenone in three products (hair oil and two root stimulators) between 8 and 20 μg/g. The other UV filters were detected at concentrations between 2 and 38 μg/g, except for 4-methylbenzylidene camphor which was never detected.

3.1.6. Glycol ethers

Glycol ethers, which are used as solvents, were not widely found in these hair products. We detected three of the eight targeted glycol ethers, and only found these in two root stimulators and one relaxer.
insulators (2 out of 4), and the hair oil. However, we did not.

out of 3; one hair lotion, one relaxing cream component), root stimu-

phenols were relatively common in hair lotions (2 out of 6), relaxers (2

39%, typically at concentrations above 10

surfactants in consumer products (EPA, 2010), in seven of 18 products

3.1.7. Alkylphenols

'yethanol, and 2-butoxyethanol.

kit’s composite shampoo/conditioner. We found the highest con-

tended in the presence of certain chemical

ants (which are responsible for breaking chemical bonds to relax hair)

had very few of the chemicals that we targeted.

Table 1

Median and maximum concentration and percent of products with concentrations > MRL for selected chemicals by product category.

<table>
<thead>
<tr>
<th>Chemical Class</th>
<th>Select Chemical</th>
<th>Anti-Friz/Polish (n = 3)</th>
<th>Root Stimulator (n = 4)</th>
<th>Hair Lotion (n = 6)</th>
<th>Hair Relaxer (n = 3)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>&gt; MRL</td>
<td>Med</td>
<td>Max</td>
<td>&gt; MRL</td>
</tr>
<tr>
<td>UV filter</td>
<td>octinoxate</td>
<td>67%</td>
<td>34</td>
<td>646</td>
<td>25%</td>
</tr>
<tr>
<td>UV filter</td>
<td>benzoic acid</td>
<td>0%</td>
<td>-</td>
<td>-</td>
<td>25%</td>
</tr>
<tr>
<td>cyclosiloxane</td>
<td>decamethylocyclopentasiloxane</td>
<td>100%</td>
<td>391,394</td>
<td>456,794</td>
<td>75%</td>
</tr>
<tr>
<td>cyclosiloxane</td>
<td>octamethylocyclopentasiloxane</td>
<td>100%</td>
<td>1,282</td>
<td>2,591</td>
<td>75%</td>
</tr>
<tr>
<td>glycol ether</td>
<td>2-phenoxethanol</td>
<td>0%</td>
<td>-</td>
<td>-</td>
<td>50%</td>
</tr>
<tr>
<td>fragrance</td>
<td>HHCB</td>
<td>67%</td>
<td>16</td>
<td>34</td>
<td>75%</td>
</tr>
<tr>
<td>fragrance</td>
<td>linalool</td>
<td>33%</td>
<td>-</td>
<td>112</td>
<td>100%</td>
</tr>
<tr>
<td>fragrance</td>
<td>limonene</td>
<td>100%</td>
<td>316</td>
<td>376</td>
<td>75%</td>
</tr>
<tr>
<td>alkylphenol</td>
<td>nonylphenol</td>
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<td>-</td>
<td>-</td>
<td>50%</td>
</tr>
<tr>
<td>ethanolamine</td>
<td>diethanolamine</td>
<td>0%</td>
<td>-</td>
<td>-</td>
<td>0%</td>
</tr>
<tr>
<td>antimicrobial</td>
<td>tricosan</td>
<td>0%</td>
<td>-</td>
<td>-</td>
<td>0%</td>
</tr>
<tr>
<td>bisphenol A</td>
<td>bisphenol A</td>
<td>0%</td>
<td>-</td>
<td>-</td>
<td>0%</td>
</tr>
<tr>
<td>phthalate</td>
<td>diethyl phthalate</td>
<td>0%</td>
<td>-</td>
<td>-</td>
<td>100%</td>
</tr>
<tr>
<td>phthalate</td>
<td>bis(2-ethylhexyl) phthalate</td>
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<td>-</td>
<td>8</td>
<td>25%</td>
</tr>
<tr>
<td>paraben</td>
<td>methyl paraben</td>
<td>0%</td>
<td>-</td>
<td>-</td>
<td>100%</td>
</tr>
</tbody>
</table>

Med = median; Max = maximum; MRL = method reporting limit; n = number of products tested per product category.

The MRL is 1 µg/g (0.001%) unless elevated due to blank detects.

– Insufficient number of data available above the MRL to calculate the summary statistic.

* Selected chemicals are the one to three chemicals most frequently detected and/or found at the highest concentrations in each class.

# Elevated MRL due to blank detect.

3.1.7. Alkylphenols

We detected alkylphenols, which are used in ethoxylate form as surfactants in consumer products (EPA, 2010), in seven of 18 products (39%), typically at concentrations above 10 µg/g. The targeted alkylphenols were relatively common in hair lotions (2 out of 6), relaxers (2 out of 3; one hair lotion, one relaxing cream component), root stimulators (2 out of 4), and the hair oil. However, we did not find any of the targeted alkylphenols in anti-frizz products or the leave-in moisturizer. We mostly detected nonylphenol and its ethoxylates, and always detected in any of the hair products: ethylene glycol monopropyl ether, ethylene glycol isopropyl ether, 2,2-butoxyethoxyethanol, 2,2-methoxyethoxyethanol, and 2-butoxyethanol.

3.2. Test results by product category

Product categories differed in the presence of certain chemical classes and concentrations. Hair lotions, root stimulators, and hair relaxers contained more frequent detects and higher concentrations of alkylphenols, DEP, and parabens than anti-frizz products (Fig. 1, Table 1). Although all products contained at least one of the 19 targeted fragrance chemicals, hair lotions, root stimulators, and hair relaxers contained multiple fragrance chemicals per product, with an average of five to eight targeted fragrance chemicals detected per product versus an average of two in the anti-frizz products. The anti-frizz products contained frequent detects and higher concentrations of cyclosiloxanes (up to 46% by weight) compared with other products, while the hair lotions did not contain the targeted cyclosiloxanes. Since cyclosiloxanes are widely considered to be conditioning chemicals, it is interesting that their use differs widely between two products that could be considered conditioning products. Looking at individual components of the relaxer kits (Fig. 2), composite shampoo/conditioner frequently contained methyl paraben, cyclosiloxanes and DEP, while the activator components (which are responsible for breaking chemical bonds to relax hair) had very few of the chemicals that we targeted.

3.3. Label accuracy of ingredients grouped by product

Ingredient labels on products did not generally list the chemicals we found in the products (Table 2), although the ingredient labels more frequently listed chemicals that we found at higher concentrations. Chemicals detected at concentrations over 1000 µg/g were labeled 54% of the time, while chemicals detected at concentrations less than 100 µg/g were labeled 8% of the time. Overall, only 16% of chemicals found above the MRL were listed on the ingredient label.

Concordance between product ingredient lists and testing results varied by chemical class. Parabens were the most accurately labeled, similar to our previous study (Dodson et al., 2012). Although butyl and ethyl paraben were never labeled, 88% of the products with detectable concentrations of methyl paraben listed it on the label. In contrast with our previous testing, most UV filters were not listed as a product ingredient. Among cyclosiloxanes, only products containing the highest concentrations of D5 (in the 40% range, or 370,000–460,000 µg/g) listed D5 on the label. Cyclosiloxanes were otherwise not listed on labels, even when present at higher levels (above 1000 µg/g). Consistent with our previous analysis, we did not find alkylphenols, ethanolamines, antimicrobials, BPA, or phthalates in the product ingredient lists.

Fragrance chemicals were generally not included in ingredient lists - the generic term “fragrance” was used instead. Six of the 19 targeted fragrance chemicals were sometimes listed as product ingredients (the natural fragrances eugenol, hexyl cinnamal, linalool, and limonene and the synthetic fragrances bucinal and methyl ionone). The US Food and Drug Administration (FDA) requires cosmetics to have ingredient lists that contain intentionally added ingredients. However, the FDA exempts trade secrets, incidental ingredients including ingredients added for processing or fragrance masking purposes, and fragrance chemicals, which can be summarized using the term “fragrance” (21 C.F.R. § 701.3 2016). Only HHCB and isobornyl acetate, both synthetic fragrances detected in the gel component of hair relaxer kit #1, were detected without either the chemical name or “fragrance” being listed.
3.4. Presence of chemicals targeted by existing policies due to health hazards

Eleven products contained seven chemicals prohibited under European Union (EU) Cosmetics Directive and/or regulated under California’s Proposition 65 (Prop 65): benzophenone, BBP, BPA, DEHP, diethanolamine, nonylphenol, and o-phenylphenol (Supplemental Table 4). The EU Cosmetics Directive prohibits chemicals on the basis of cancer, birth defects, or reproductive harm. Detected concentrations ranged between 2 and 310 μg/g. The two no-lye hair relaxers for children (kits #1 and #3) contained the highest levels of four EDCs prohibited in the EU (DEHP, nonylphenol, BPA, and diethanolamine), as well as Prop 65 listed o-phenylphenol, with kit #1 containing all five of these EDCs. In addition to the seven chemicals prohibited in the EU, we detected another 16 chemicals regulated but not prohibited in the EU. These 16 chemicals were in compliance with EU restrictions, but methyl paraben came closest to the maximum concentration permitted, with one hair lotion containing 0.21% by weight compared to the 0.4% limit.

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**Fig. 2.** Concentrations of 60 targeted chemicals (left) in hair relaxer kit components (bottom). Chemicals are grouped by chemical class, with natural and synthetic fragrances separated by a dashed horizontal line. Hair relaxer kit components are grouped by kit (Relaxer #1, Relaxer #2, Relaxer #3). Shampoo and conditioner components represent a composite sample. Numbers at the top of the figure indicate the number of chemicals detected in each product; numbers on the right indicate the number of products containing each chemical. Detects below the method reporting limit (MRL) are not shown. The MRL is 1 μg/g (0.0001%) unless elevated due to blank detects.
We frequently detected EDCs and asthma-associated chemicals in hair products commonly used by Black women. Root stimulators, hair lotions, and relaxers frequently contained fragrance chemicals, nonylphenols, DEP, and parabens. Anti-frizz products contained the highest levels of cyclosiloxanes. A majority of products contained UV filters. We found poor concordance between detected and labeled ingredients, especially at lower concentrations. Eleven products contained seven EDCs prohibited in the EU or regulated under Prop 65, including two relaxer kits marketed for use by children containing the highest levels of five of these chemicals.

We found several chemicals at higher concentrations in these products compared with most previously tested hair products (Supplemental Figure 1), although previous studies have been limited to shampoos, conditioners, and hair styling products. We measured nonylphenols, cyclosiloxanes, and the UV filter BP-3 at higher maximum concentrations than in previous studies (Capela et al., 2016; Dodson et al., 2012; Dudzina et al., 2014; Horii and Kannan, 2008; Liao and Kannan, 2014; Lu et al., 2011; Wang et al., 2009), and DEP and parabens at concentrations comparable to the high end of the range reported previously (Dodson et al., 2012; Guo and Kannan, 2013; Hubinger, 2010; Koniecki et al., 2011). Our previous study of 213 products representing 50 types of consumer products tested composites of different brands to create “average” profiles for shampoo, conditioner, and hair styling products. Average concentrations of parabens in hair products used by Black women were similar to concentrations in composited commercial products in our previous study, but average DEP concentrations were somewhat higher in products used by Black women, and nonylphenol ethoxylates were only detected in these products. Previous studies have reported higher concentrations in other types of personal care products (e.g. DEP in perfume, cyclosiloxanes in antiperspirant, and BP-3 in lip care) than we found in this study (Dodson et al., 2012; Manova et al., 2013; Wang et al., 2009).

Many of the frequently detected chemicals are estrogenic (Ajj et al., 2013; Habauzt et al., 2017; Kim et al., 2011; Kunz and Fent, 2006; Pan et al., 2016; Quinn et al., 2007; Schlumpf et al., 2004; Thomas and Dong, 2006; Wielogorska et al., 2015) or associated with developmental or reproductive disruption. While methyl paraben (the paraben present at the highest concentration and frequency in this study) is the least hormonally active of the parabens tested, it is associated with altered hormone levels in pregnant women in NHANES (Aker et al., 2016) and has estrogenic and adipogenic activity in vivo (Hu et al., 2016) and in vitro (Hu et al., 2013, 2017; Morohoshi et al., 2005; Pereira-Fernandes et al., 2013; Routledge et al., 1998). Methyl paraben and total parabens were also associated with increased size in male babies and toddlers in China (Guo et al., 2017; Wu et al., 2017). Although a small study in babies in New York found an inverse effect with longer chain parabens and no effect of methyl paraben (Geer et al., 2017). DEP (the most frequently detected chemical in this study), which does not seem to exhibit the same effects as other phthalates on fertility (Hauser et al., 2016), pre-term birth (Ferguson et al., 2014), or male development (Gray et al., 2000), has been associated with lower progesterone levels, impaired glucose tolerance, and gestational weight gain in pregnant women (James-Todd et al., 2016b; Johns et al., 2015). In addition, DEP exhibits hormone disruptive effects in vivo (Fujii et al., 2005; Kumar et al., 2014, 2015; Sohn et al., 2016) and in vitro (Mankidy et al., 2013; Sohn et al., 2016). Its mechanism doesn’t appear to be the same as the well-known anti-androgenic phthalates (Gray et al., 2000; Howdeshell et al., 2008), but there are indications DEP affects hormone synthesis, possibly via effects on aromatase (Karmaus et al., 2016; Kumar et al., 2014, 2015; Mankidy et al., 2013; Sohn et al., 2016). Exposure to DEP in dust is associated with additional neurological impairments in children with autism or developmental delay (Philippat et al., 2015b) and DEP affects migration of neural stem cells in vitro (Ishido and Suzuki, 2014). DEP is a common solvent in fragrance formulations and so may be a proxy for fragrance chemical exposure in epidemiological studies. Some UV filters and nonylphenols affect development and reproduction in animals (Baich and Metcalfe, 2006; Chang et al., 2012), and some fragrances also disrupt hormone signaling (Li et al., 2013; Schreurs et al., 2005; van der Burg et al., 2008).

In addition to their endocrine disrupting effects, benzophenone, DEHP, diethanolamine, and D5 are carcinogenic in animals (EU-SCCS, 2016; IARC (International Agency for Research on Cancer), 2012a, 2012b, 2012c). While in principle most carcinogens including DEHP and diethanolamine are banned in cosmetics in the EU, the EU has not banned D5 – which caused uterine tumors in rats - based on a risk assessment that assumed a threshold for the cancer effect (EU-SCCS, 2016). The EU has also not banned benzophenone, an International Agency for Research on Cancer (IARC) 2B carcinogen (IARC (International Agency for Research on Cancer), 2012a), and the EU Scientific Committee on Consumer Safety has not reviewed that chemical’s status.

While use of personal care products has been shown to increase exposure to product ingredients (Berger et al., 2018; Branch et al.,
2015; Braun et al., 2014; Fisher et al., 2017; Guo and Kannan, 2013; Harley et al., 2016; Meeker et al., 2013; Parlett et al., 2013; Philippat et al., 2015a) and many of the ingredients pose hazards, health risks from these exposures are difficult to predict because of remaining uncertainties. We did not find authoritative assessments of the health risk associated with these chemicals in personal care products or from dermal exposure in general. Assessments for hair products used by Black women would require additional information or assumptions about product use and exposure. In addition, these types of risk estimates still have a lot of uncertainty. For example, epidemiological studies have shown effects of DEHP in humans at doses below those estimated to be safe by these risk-based methods (NAS, 2017). In addition to risk estimates for individual chemicals in specific products, it is also important to consider the cumulative risk resulting from multiple chemicals in the full range of frequently used products.

Hair products we tested contained many chemicals associated with similar health endpoints. Mixtures of chemicals may act additively through a common biological pathway or affect multiple carcinogenic mechanisms, resulting in a greater effect than each chemical in isolation (SCHER, SCCS, SCENIHR, 2012; Miller et al., 2017; NRC, 2008). Low-dose mixtures of phthalates, parabens, UV filters, fragrances and other common chemicals exhibit additive anti-androgenic activity (Howdeshell et al., 2008; Ma et al., 2014; Orton et al., 2014; Pop et al., 2016) and additive estrogenic activity (Evans et al., 2012; Kunz and Fent, 2006; Silva et al., 2002). Similar additive effects might be expected from the combinations of chemicals frequently detected here. Thus the effect of chemical mixtures in product formulations and cumulative exposures from multiple products needs to be considered, and new approaches are being developed to do this (Bois et al., 2017; NRC, 2008).

This study supports our previous observation that product labels are an incomplete guide to exposure for many EDCs (Dodson et al., 2012). In our analysis of ingredient labels, we found that most (84%) of the detected chemicals in these hair products were not listed on the label. Chemicals detected at the lowest concentrations (under 0.01% by weight) were least often labeled, while chemicals detected at higher concentrations (over 0.1% by weight) were more often labeled. Parabens were found on the product ingredient label more often than other chemical classes. Incomplete labeling is worrisome because product ingredient labels are the often the only source of information for individuals seeking to reduce their exposure to a chemical of concern.

4.1. Limitations

This study provides preliminary data to identify exposures that Black women experience from hair products, an under-studied research area. We tested six commonly used hair product categories that include frequently used brands for the presence of a large number of chemicals associated with endocrine disruption and asthma. However, the number of products tested was limited by project budget, so we were not able to make statistical comparisons between categories. Furthermore, we had only one leave-in conditioner and one hair oil, limiting our ability to draw larger conclusions about these two product categories.

We selected products based on a 2004–5 survey of New York City women including African-Caribbean and African-American women and purchased products in 2008, so this study best represents the large variety of hair products used by Black women from the US and Caribbean living in New York at that time. However, these products still have relevance today. A 2013 market survey found that older “heritage” brands (some of which were included here) were still being used by a large percentage of Black respondents (Mintel Group, 2013), and seventeen of the 18 products that we tested are still available in major walk-in stores in the same or similar packaging. The eighteenth product (hair relaxer #2) is available from secondary retailers online, but we did not find it in stores. We confirmed that for the products still in stores, 78–100% of the ingredients listed on the original, tested product are still found on the labels of products sold today, and a majority (11 out of 17) of products still list all of the original ingredients on the newer product label. A majority (10 out of 17) also list 1–12 additional ingredients not listed on the older products. Of the chemicals we detected, only D4 was added to or removed from ingredient labels: two anti-frizz products with detectable concentrations of D4 now list D4 in the current product ingredient lists, so the label concordance of this chemical may have improved. Although product formulations appear temporally consistent, it is possible that concentrations in products have changed since the products were purchased. Population level exposures to some chemicals including methyl paraben and DEP have declined over time (CDC, 2017), likely reflecting a shift in manufacturing practices.

While our product testing represents one of the first studies to characterize chemical concentrations in hair products used by Black women, the products tested here do not represent all hair products used by Black women in the US in 2005 or at present. Women who identify as Black may use very different products depending on geography, culture, country of origin, and socioeconomic status, so these results cannot necessarily be generalized to all Black women. Furthermore, the market for Black women’s hair products continues to expand, in part driven by the increase in natural (un-straightened) hairstyles: between 2008 and 2012 the market for relaxers declined by twenty percent (Mintel Group, 2013). Products such as curl puddings, conditioner for washing, edge control products, and blow-out straightening products are not assessed here. We included four root stimulator products and one hot oil treatment; however other hair growth and hair oil products for Black women (and off-label uses of other products) could be included in future studies. Hair glues, sometimes used to attach extensions, may contain chemicals of concern but were not tested in this study.

We targeted a large number of endocrine disrupting and asthma-associated chemicals. However, our products listed other ingredients including propyl paraben and formaldehyde releasers that would contribute to the cumulative health effects of the product. Furthermore, many chemicals used in hair products have not been extensively tested for endocrine disrupting activity and were not targeted here. One study found evidence of endocrine activity for petrolatum, or petroleum jelly (Myers et al., 2015), suggesting that other common chemicals may be found to have endocrine disrupting properties.

Based on differences in hair product use by Black women (James-Todd et al., 2012; Wu et al., 2010), Black women’s chemical exposures from hair products may differ from hair product exposures among other women. The frequent detection of DEP and parabens in the products is consistent with elevated biological levels of DEP and parabens among Black women (Calafat et al., 2010; James-Todd et al., 2017). Many of the other chemicals that we frequently detected (fragrance chemicals, nonylphenols, and most UV filters) are not included in biomonitoring programs, making it difficult to discern whether disparities exist. Future work will need to determine whether use of these types of products contributes to measurable exposures and to exposure disparities in these chemicals.

5. Conclusion

We identified a wide range of endocrine disrupting and asthma-associated compounds in hair products used by Black women, including products marketed towards children. We found cyclosiloxanes, parabens, and the fragrance marker DEP at the highest levels, and DEP most frequently. Root stimulators, hair lotions, and hair relaxers frequently contained fragrances, nonylphenols, DEP, and parabens, while anti-frizz products had higher concentrations of cyclosiloxanes. We also found UV filters in a majority of products and at higher maximum concentrations than previously detected in hair products. Hair relaxers for children contained multiple chemicals prohibited in cosmetics in the EU and regulated under California’s Proposition 65 because of their
links to reproductive toxicity and cancer. A further implication of our study is that consumers cannot reliably use product labels to reduce certain exposures, as indicated by poor concordance between measured ingredients and product labels.

Given the exposure and endocrine-mediated health disparities experienced by Black women, new research and regulatory activities should consider the effects of ethnic differences in product use on exposures and health. Additional research is needed on the chemical composition of products marketed to and used by Black women and on product use and exposure in this population. Risk assessments should include dermal routes of exposure and co-exposures in susceptible populations including Black women. In the future, emerging approaches to estimate mixture effects could be applied to evaluate each multi-ingredient product by itself and in combination with other commonly used products (Bois et al., 2017; NRC, 2008; Schug et al., 2013). Considering the uncertainties in estimating risks, a precautionary approach would reduce the use of endocrine disrupting chemicals in personal care products and improve labeling so women can select products consistent with their values.

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Conflicts of Interest

The authors declare they have no actual or potential competing financial interests.

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.envres.2018.03.030.

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