Charcoal Cloth Pad as a Qualitative Dermal Sampler for Toluene while Spray Painting in an Auto Body Repair Shop

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Abstract:

Background: In small-scale automobile repair shops in Thailand, workers often spray paint without a dedicated booth, which means they are exposed to paint fumes both via the respiratory system and the skin. Although the respiratory system is considered more critical in terms of protection measures, the skin exposure is often neglected.

Objective: To study the feasibility of using charcoal cloth pads as a means to evaluate qualitative skin contact, and specify the body areas that should be sampled.

Materials and Methods: A case study in one automobile repair shop, with 4 spray painters participating in the study. The researcher attached charcoal cloth pads, made from 100% charcoal, to various body areas that were exposed to paint fumes. Samples were collected from 31 pads over 13 days (1 pad per day per person), and participants wore protective masks during their work. Urine samples were also collected from before and after work, the urine samples were analyzed for hippuric acid and toluene levels.

Results: It was found that there was a significant correlation between the level of hippuric acid (milligrams/gram creatinine) and the level of toluene on the charcoal cloth pads in the chest area: hippuric acid (milligrams/gram creatinine) = 227.5 + 0.162 * toluene level (milligrams) in a sample of 258. Results showed that the most suitable area for attaching the charcoal cloth pads was the chest area, which had the highest level of toluene on the pads.

Conclusion and Recommendation: Charcoal cloth pads can be used to evaluate qualitative skin contact. When the level of hippuric acid in the urine is 50% of the indicated value (1.25 grams/gram creatinine), the toluene level on the pads is estimated to be approximately 8,800 milligrams. This indicates a high level of skin contact that may be dangerous to health. It is recommended to provide protective clothing to spray painters when the toluene level on the pads is above 8,800 milligrams.

Keywords: Spray painting / Charcoal cloth pad / Quality skin contact / Toluene
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Abstract:
Small auto body repair (ABR) shops in Thailand mostly do not have spray painting booths, so workers are likely to have inhalation and dermal exposure to paint mist, but only inhalation exposure has been of concern.

Objective: The objectives of this study were to explore the possibility of using a charcoal cloth pad (CCP) as a simple tool to assess dermal exposure qualitatively, and investigate the body site where the CCP should be placed.

Material and Method: A Small ABR was selected as a case study. Four painters in a small ABR were recruited in to the study. The CCPs were made of 100% activated charcoal pad and attached on the potential exposed areas of the participants’ bodies. Thirty-one CCP sample sets were collected in 13 days (1 set/day/participant). Respirators were provided to wear while working. Pre- and post-shift urine samples were collected and analyzed for hippuric acid (HA) using GC. Linear regression was used to analyzed for the relationship of net HA and toluene on the CCP (TolCCP). Result: The net urinary hippuric acid (HA) correlated well with TolCCP attached at painters’ chests of net HA (mg/g creatinine) = 227.5 + 0.162 TolCCP (mg), r² = 0.258, p-value = 0.003. The suitable dermal sampling site on spray painter was the chest, where the average quantity of toluene on the CCPs was the highest.

Conclusion and recommendation: The CCP could be used as a tool for qualitative dermal exposure assessment by attached the pad at the painter’s chest. Substitute 50% BEI of toluene (1.25 g HA/g creatinine) for net HA in the above equation and round up the number to obtain a recommended limit of toluene on CCP of 8,800 mg. The quantity indicates that the worker have high dermal exposure and may permeate through the workers’ skin at significant amount to harm their health. Therefore, it was suggested that protective cloth should be provided to the spray painting workers from 8,800 mg or more toluene was found on CCP.

Keywords:
Spray painting / Charcoal cloth pad / Auto body repair / Qualitative dermal sampler

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Introduction

Several small auto body repair (ABR) shops are located all throughout Thailand. In 2015 the number of registered ABR shop, those have equal to more than 2 horsepower machine or have 7 workers or more, was 6,674 (Department of Industry, 2016). It could be because the cost of new parts is expensive while labor is cheap. Work processes in ABR shops could involve welding, riveting, hammering, painting and polishing; thus, general occupational hazards include noise, welding fumes, paint mist and chemicals, mostly solvents. Hazard exposure control measures, if shops have them, normally include personal protective equipment (PPE) such as masks, gloves, ear plugs and the most popular PPE provided to employees are knitted gloves (Thonglaiat, P., 2015), but no protective clothing to prevent dermal exposure. A few small ABR shops have standard spray booths to control the dispersion of spray mist. According to a study conducted in 2012 in a municipality of a province in the northeastern Thailand (Notesupa, S. and Inmuong, U., 2012) of 23 ABR shops, regardless of size, 37.5% did not have standard paint spray booths. Therefore, for chemical hazards, workers may have both inhalation and dermal exposure, especially during spray painting in which toluene, a chemical with skin notation (Health and Safety Executive, 2011), is a major component of solvent. While air sampling is a widely used method to assess inhalation exposure, no acceptable method to assess dermal exposure, although several tools, e.g., strip tape, wiping, charcoal cloth pad (CCP) etc. were studies (Chen, B., Zheng, L., Wang, D., Liu, F., Huang, Q., 2015; Cohen, B.M., and Popendori, W., 1989; Berna, Van Wendel De Joode., Tielemans, E., Vermeulen, R., Wegh, H., and Kromhout, H., 2005).

CCP was first studied with positive results and was recommended for further study as dermal sampler by Cohen (Cohen, B.M., and Popendori, W., 1989) and later was studied by some other researchers with somewhat positive results for CCP to be used as dermal sampler with additional complicated information needed. (Berna, Van Wendel De Joode, Tielemans, E., Vermeulen, R., Wegh, H., and Kromhout, H., 2005; Vermeulen, R., Qing, L., Guilan, L., Rappaport, S.M., Kim, S.Y., Berna, van Wendel de Joode, et.al., 2006) However, due to their simplicity to make and analyze, CCP drew our attention to study and develop a simple tool for qualitative dermal exposure assessment for the occupational personnel to recommend skin protection for workers.

Hippuric acid (HA) is a major metabolite of toluene in human body and has been used as a biomarker of exposure. Until the American Conference of Governmental Industrial Hygienists (ACGIH) reduced the threshold limit value (TLV) to 20 ppm, the use of HA as a biological exposure index (BEI) was removed. Since urinary HA is a normal constituent of urine, which originating from food, drink, and medications containing benzoic acid or benzoates and at concentrations below 50 ppm is difficult to differentiate occupationally exposed from the background (American Conference of Governmental of Industrial Hygienist, 2010; Foo, S.C., Jeyaratnam, J., Ong, C.N., Khoo, N.Y., Koh, D., and Chia, S.E.; Tardif. R., Truchon, G., and Brodeur, J., 1998). However, the Thai’s occupational exposure limit (OEL), and also the PEL of the US. Occupational Safety and Health Administration, for toluene...
is 200 ppm corresponding to BEI of 2.5 g HA/g creatinine, thus, the use of HA as internal dose indicator should be appropriate. Nevertheless, to cope with the above problem in case of low exposure to toluene, the pre-shift HA should be collected as the background level and the known interferences should be controlled as much as possible, including that the quality assurances process and procedures are strictly followed.

Objective

The aim of this study was to explore the possibility of using CCP as a tool to assess dermal exposure to toluene from spray painting, and the body site where dermal samplers should be placed.

Methods and materials

Study site and participants

A small ABR shop was selected as a study area for this study. The shop was 10 × 80 × 4 m³, with one large space and 2 spray booths. One large area contained 3 spray spaces, 2 chemical and paint storage areas, 3 repair areas, and 2 polishing areas. Most of the dusty and dirty work took place in two thirds of the inner part. The spray painting of large auto parts or a whole car took place in the booths, and the small parts were sprayed in 3 areas. In addition to 2 spray booths, no local exhaust or any control measures were observed even the use of PPEs in the shop. The control units in the spray booths were particulate filters. However, due to the height of the roof and 6 roof fans, the airborne contaminants were diluted to acceptable level after generated with in less than half an hour. The study protocol was approved by the Ethics Review Committee for Human Research, Faculty of Public Health, Mahidol University with the protocol number MUPH 2010-197 before the data would be collected. Four auto body repairers and 5 painters worked in the shop; however, only 4 painters signed the consent form and participated in the study. All four participants performed the same work but not equally including dry and wet sanding, applying filler and putty, mixing paint and spraying primer, lacquer or paint. The preparation works such as sanding and applying filler and putty were mostly done in the morning, and spraying primer, lacquer or paint in the afternoon. Figures 1 and 2 show paint loading and paint spraying outside the booth, respectively. Spray painting on a small part took about 15-30 minutes, while a large part might take an hour. However, the workers usually took short breaks 1-2 times during worked on large part. The workers did not have work clothes; some wore shorts or trousers and T-shirts or long sleeve shirts. The major chemical used in the ABR shop was toluene as the mixture in several chemicals including lacquer, thinner and top coat while primer (putty and primer) was another major material used in the ABR shop composed of cyclohexanone peroxide, xylene and ethylbenzene.
A few days before sample collection started, the researchers conducted a test to familiarize the participants with the study methods. Furthermore, in order to prevent inhalation exposure, the respirator fit testing was conducted for all participants on that day and half-face respirators equipped with dust and organic vapor filters were provided to wear while working throughout the data collection period.

**Dermal sampling and analysis**

The CCPs were made from a 100% activated carbon pad (ACP) with a surface density of 240 g/m², 1 mm. thick. The CCP has 3 x 3 cm² of ACP pad was prepared as shown in Figure 3 according to Cohen’s (6) suggestion, which CCP was sandwiched between gloze and aluminum foil and sewed with stapler. The charcoal cloth pads were tested for absorption capacity to prevent over sampling time which may cause underestimation of exposure. The pads were placed at left and right hands, neck and chest of the participants who worked in the spray booth for 50, 73, 90, 150, 235, and 240 minutes and 7, 10, 13, 20, 23, 29, 30, and 49 minutes for those worked outside the booth, 4 samples for each duration. The highest quantity found was 12.18 mg. The quantity of toluene were plotted against the duration, which a ‘plateau’ was never reach. Thus the maximum of 240 minutes was a safe sampling time. The CCPs were stored in plastic lock zippers to avoid contamination prior to use and after sampler the CCPs were collected in a plastic zip lock held in a temperature control box below 5 degrees Celsius.

Before starting dermal sampling, the participants were interviewed to collect information on work and practice (duration of spraying, amount of paint and toluene loading in a spray can etc.) during the day. Their skin condition, e.g., damage, disease, hair density and exposed skin surface area, were visually inspected and recorded. Furthermore, environmental conditions including temperature, relative humidity and wind velocity were measured and recorded daily.

![Figure 2. Spray painting](image1)

![Figure 3. Dermal sampler](image2)
and T-shirt, the samplers would be placed on the left and right hands (at the dorsal side), left and right forearms (front side), neck (front), chest, left and right lower legs (front side), and left and right foot (dorsal side). The samplers would not be placed on the forearms, lower legs, and feet if trousers and a long sleeve shirt were worn. The samples were collected after finishing each paint spraying work and a new set was attached right before the next spray session started; a total of 31 sets of samples were collected in 13 days (1 set/day/worker). Two unsampled pads were drawn randomly as filed blanks each day. Each set of samples was wrapped with clean aluminum foil, placed in a zip-lock plastic bag and stored in an ice box to transport to the lab where all samples were kept at -20 OC until analysis.

The samples were analyzed using gas chromatography (GC), flame ionization detector (FID) (Hewlett-Packard 6890), equipped with column (HP-Wan Bonded Polyethylene Glycol capillary 30.0 m 250 mm 0.25 mm). The limit of detection (LOD) and limit of quantitation (LOQ) were 6.64 ug/sample and 18 ug/sample, respectively. The standard curve was obtained from 11 known toluene concentrations ranging from 17.25 to 10,229.23 ug/ml.

Air sampling and analysis

Since the participants wore respirators only while mixing paint and spraying, full period air samples were collected daily to estimate inhalation exposure of the participants using coconut shell charcoal tubes (100 mg front/50mg backup) according to NIOSH method 1501. Stationary samples were taken in 6 areas consisting of spray booth 1, spray booth 2, spray area 1, spray area 2, spray area 3, and paint mixing &

Figure 4. Sampling points for shorts/T-shirt and trousers/long sleeves
equipment cleaning area. Area sampling was used in this study due to a posture of participants among spraying that the spray mist may impact to the equipment. Anyways, the inhalation exposure of participants was calculated based on the toluene concentrations via area sampling which located near by a working area and time they spent at each area. A total of 4 to 5 samples were taken daily depending on where the participants worked on those days. The time the participants spent in each area was recorded. Standard industrial hygiene practices were strictly followed for air sampling. A total of 55 samples were obtained over 13 days. Two unsampled charcoal tube were drawn randomly as field blanks each day for quality control of sampling. The samples were analyzed with GC using the same methods and equipment as the dermal samplers. An 8-hr time weight average (TWA) was calculated for the participants daily based on the toluene concentrations in the air and time they spent at each area without the respirator on.

Urine sampling and analysis

The participants were asked to avoid consuming soft drinks, fermented food and certain kinds of medicine. When they did, they were instructed to record on the urine collection form given on each day with a urine collecting bottle. Urine samples were collected before work and at the end of work shift from each participant on the same days with the air and dermal samples taken in cleaned polyethylene bottles. A total of 31 pairs (before and at end of shift) of urine samples were obtained. The samples were analyzed for HA using High Performance Liquid Chromatography (Hewlett-Packard 1100) equipped with column (reverse phase C8, 250 x 4 mm, Stainless-steel column packed with octadecylsilanized silicate). The LOD and LOQ were 0.92 ug/ml and 3.1 ug/ml, respectively. Creatinine in urine was analyzed using an analyzer - Star dust MC15 REF RA116000.

Statistical Analysis

The association between general characteristic of the participant, work and environment factors and the urinary HA was analyzed using Pearson’s correlation. The association between total toluene on the CCP (TolCCPs) and 8-hr TWA from air sampling, and the urinary HA was analyzed using Pearson’s correlation and simple linear regression.

Results: Participants’ characteristics, work and environment factors

Four painters in a small ABR were recruited to participate in this study, all of them are male, age range from 26-48 year old, graduated from Grade 6 to Junior high school, all of them smoker with an average 4 cigarettes/day, 75% of samplers are alcohol drinker which drank at the end of the day or weekend. The participants had mostly intact skin which 3,228 cm2 skin surface areas, only small scratches on hands and legs of participants #1 and #3 in a few day of data collection period. Furthermore, these two participants took pain release medicine for muscle and headache on a few days. All washed hands before lunch. They brought home cook foods to eat in a provided area in the shop. Temperature, humidity, and barometric pressure were normal for Thailand. The work and environment factors are presented in Table1.
Participants, environment and work factors that may contribute to the skin absorption considering by the association with the HA were analyzed using Pearson’s correlation. The factors with the highest and significant correlations were hair density \( r = -0.602 \), toluene loading \( r = 0.460 \) and paint loading \( r = 0.455 \), p-value < 0.01.

**TolCCPs, Toluene in air and HA**

Toluene concentrations in the workplace during 13 days ranged from 1.11 – 31.36 ppm (mean + SD; 7.4 + 6.2). These concentrations were used to calculate the 8-hr TWA exposure level of the participants, with results ranging from 1.2 to 14.6 ppm (mean + SD; 5.5 + 3.6), far below the TLV and less than 10% of Thai’s OEL of 200 ppm. While post-shift HA ranged from 0.06-0.88 g/g creatinine (mean + SD; 0.43+0.21), which was slightly higher than background value (pre-shift HA) ranging from 0.0-0.40 g/g creatinine (mean + SD; 0.14+0.12), and net HA (post-shift HA – pre-shift HA) ranged from 0.03-0.65 g/g creatinine (mean + SD; 0.29+0.18). Table 2 presents mean and standard deviation of the 8-hr TWA, toluene on CCPs and Urinary Hippuric Acid of all participants.

**Table 2.** Mean (SD) of the 8-hr TWA, total TolCCP and pre-, post-shift and net HA of all participants

<table>
<thead>
<tr>
<th>Participant No.</th>
<th>N</th>
<th>8-hr TWA (ppm)</th>
<th>TolCCP (mg)</th>
<th>Urinary Hippuric Acid (g/g creatinine)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Pre-Shift</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Post-shift</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Net</td>
</tr>
<tr>
<td>1</td>
<td>5</td>
<td>4.37 (2.59)</td>
<td>204.05 (118.34)</td>
<td>0.10 (0.13)</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>4.52 (2.59)</td>
<td>157.84 (188.80)</td>
<td>0.08 (0.10)</td>
</tr>
<tr>
<td>3</td>
<td>13</td>
<td>6.66 (4.45)</td>
<td>1071.73 (825.92)</td>
<td>0.17 (0.10)</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
<td>5.67 (4.05)</td>
<td>160.43 (40.64)</td>
<td>0.27 (0.16)</td>
</tr>
</tbody>
</table>
The association between TolCCPs, Toluene in air and HA

The quantity of toluene on CCP samplers varied by the height and size of the auto parts and parts of the participants’ body, ranging from 14.10 - 2849.28 mg (Table 3). The association between independent variables of total TolCCPs and 8-hr TWA, and dependent variables of net-post-shift and net HA were analyzed using Pearson’s correlation and found that the TolCCPs correlated with the net and post-shift HA, with r of 0.508 and 0.445 respectively, and p-value < 0.01. While the 8-hr TWA did not correlate with either net or post-shift HA, indicating that inhalation exposure played trivial role on HA.

Table 3. Toluene on CCPs at different body parts for each subject

<table>
<thead>
<tr>
<th>Participant No.</th>
<th>N</th>
<th>Quantity of toluene on charcoal cloth pads at different body parts (mg)</th>
<th>Sum of toluene (mg)*</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Left hand</td>
<td>Right hand</td>
</tr>
<tr>
<td>1</td>
<td>5</td>
<td>11.72</td>
<td>12.15</td>
</tr>
<tr>
<td>SD</td>
<td></td>
<td>8.80</td>
<td>11.72</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>5.92</td>
<td>7.11</td>
</tr>
<tr>
<td>SD</td>
<td></td>
<td>6.14</td>
<td>7.28</td>
</tr>
<tr>
<td>3</td>
<td>13</td>
<td>93.05</td>
<td>80.43</td>
</tr>
<tr>
<td>SD</td>
<td></td>
<td>70.31</td>
<td>51.28</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
<td>13.67</td>
<td>11.92</td>
</tr>
<tr>
<td>SD</td>
<td></td>
<td>3.83</td>
<td>3.44</td>
</tr>
<tr>
<td>Over all mean</td>
<td></td>
<td>44.14</td>
<td>39.13</td>
</tr>
<tr>
<td>SD</td>
<td></td>
<td>61.58</td>
<td>48.61</td>
</tr>
</tbody>
</table>

Among 31 sets of CCP samples, the highest quantity of toluene was found in 21 samples attached to the chest; the 2nd and 3rd highest were the hands and forearms, respectively. The relation between net HA and TolCCPs attached to parts of the body was analyzed, and the results showed that net HA correlated significantly with only TolCCPs attached on the chest and neck, r = 0.523 and 0.477, respectively and p-value <0.01. Thus, association of TolCCPs (chest) and net HA were further analyzed using linear regression. The association is presented in equation (1), with the coefficient of determinant (r2) = 0.258 and p-value = 0.003. Net HA (mg/g creatinine) = 227.5 + 0.162 TolCCP (mg)…… (1)
Discussion and conclusion

Work and personal factors related significantly to HA were paint and toluene loading and hair density. For negative relation of hair density means that the thicker the hair the less chemical was absorbed, because the solvent mist fell on the hair and evaporated before permeating through skin. Therefore, the correlation was negative for a person with thick hair would have low toluene absorption. For work factors the amount of paint and toluene loading into the spray can suggest workload in each day, which in turn related to toluene exposure and finally the HA.

The most suitable part of the body where the CCPs should be placed was the one with the highest exposure, for the sake of worker protection. The result showed that the CCPs with the highest quantity of toluene were the ones attached on the chest and the second highest was on the neck with significant correlation with the HA. This was no surprise because those positions were most likely to be exposed to the paint mist no matter the size and height of the repaired part. However, to control the cost, only one CCP/person should be used thus, the CPPs sampling points for spray painting should be at the chest.

The 8-hr TWAs were low and no correlation with HA while those of TolCCPs and net HA significantly correlated means that the control of inhalation exposure using respirator during spraying was effective and the major route of exposure to toluene was skin. Furthermore, it can be concluded that the CCP had positive trend to be used as a dermal sampler. However, the association was moderate ($r = 0.508$) due to the shortcomings mentioned above, but could not be easily controlled, i.e. completely control of inhalation exposure, and work schedule. The participants did not cooperate in wearing the respirator at all times of the work shift and internal dose (HA) was quite low, thus inhalation exposure could contribute partly to HA. Although 8-hr TWA was known, the quantity of HA due to inhaled toluene could not be predicted and subtracted from total HA. Furthermore, among the variety of work on one day that the participants had to do, the spray painting which is the target of work to study was scheduled in late afternoon. Thus, excretion peak of HA due to spray painting may not reached yet at the end of the work shift. (Lof, A., Hjelm, E.W., Colmsjo, A., Lundmark, B-O., Norstrom, A., and Sato, A., 1993; Ogata, M., Takatsuka, Y., and Tomokuni, K., 1971)

We may not be able to quantitatively weigh the positive and negative factors influencing HA level, nevertheless, we are confident that the most influencing factor on HA was dermal exposure during spray painting. Therefore, we concluded that CCP is a promising property of the dermal sampler and may be used for qualitative assessment to indicate low/high dermal exposure of the workers by applying the equation 1 as follow.

Substitute the toluene BEI (2,500 mg/gm creatinine) into the equation:

$$2500 \text{ mg/gm of creatinine} = 227.5 + 0.162 \text{ TolCCP (mg)}$$

Then, $\text{TolCCP} = 17,588 \text{ mg}.$

That is 17,588 mg of TolCCP may cause internal dose, HA level, equal to 2.5 g/g creatinine. Therefore to protect the worker, only 50% BEI should be employed in the equation; then critical
value of TolCPP is approximately 8,800 mg. When
the quantity of TolCPP exceeds 8,800 mg, the
dermal exposure is high and toluene may be
absorbed into the body in significant quantity
to cause health effects. Therefore, protective
clothing including cap should be provided for
workers while spray painting.

What is already known on this topic?

Charcoal cloth pad has been developed
and used to assess volatile chemicals on the
skin both in a laboratory setting and field
studies. The correlation between biomarkers
and dermal exposure were not found due to
low dermal exposure. However, it was suggested
that CCP could be a useful tool for quantifying
the probability of dermal exposure to organic
solvents and to provide estimates of the potential
contribution of the dermal pathway to systemic
exposure.

Small ABR shops in Thailand do not mostly
have spray painting booths, so workers are likely
to have inhalation and dermal exposure to paint
mist, but only inhalation exposure has been of
concern. As we know, hazard exposure control
measures in most of the small and medium ABR,
if they have ones, normally include personal
protective equipment (PPE) such as masks, gloves,
ear plugs, but no protective clothing. While some
chemical used in the ABRs, e.g. toluene is capable
of penetrating the skin in significant quantity
to cause health effects.

However, no one demonstrate and quantify
to present that the possibility of using CCP as a
qualitative tool to assess dermal exposure to
toluene from spray painting, and to spot the body
site where dermal samplers should be placed.

What this study adds?

The study showed that the CCP can be
used as a screening tool for occupational health
personnel to use for qualitative assessment to
indicate low/high dermal exposure with the
critical value of TolCPP (8,800 mg). The most
suitable part of the body where the CCPs should
be placed, the chest, is identified in the study
as well. Therefore, protective clothing including
cap should be provided for workers while spray
painting. Nevertheless, due to the limit of the
result from this study which came from one ABR
shop, the critical value may be differential from
other thus the application of this model need to
consideration.
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