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Firefighter exposures to potential endocrine disrupting chemicals measured by military-style silicone dog tags

Carolyn M. Poutasse [a], Christopher K. Haddock [b], Walker S.C. Poston [b], Sara A. Jahnke [b], Lane G. Tidwell [a], Emily M. Bonner [a], Peter D. Hoffman [a], Kim A. Anderson [a,*]

[a] Department of Environmental and Molecular Toxicology, Oregon State University, Corvallis, OR 97331, United States
[b] Center for Fire, Rescue, and EMS Health Research, NDRI-USA, Leawood, KS 66224, United States

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ABSTRACT

Studies suggest that exposure to potential endocrine disrupting chemicals (pEDCs) may contribute to adverse health outcomes, but pEDC exposures among firefighters have not been fully characterized. Previously, we demonstrated the military-style silicone dog tag as a personal passive sampling device for assessing polycyclic aromatic hydrocarbon exposures among structural firefighters. This follow-up analysis examined the pEDC exposures based on department call volume, duty shift, and questionnaire variables. Structural firefighters ($n = 56$) were from one high and one low fire call volume department (Kansas City, MO metropolitan area) and wore separate dog tags while on- and off-duty ($n_{dogtags} = 110$). The targeted 1530 analyte semi-quantitative screening method was conducted using gas chromatography mass spectrometry ($n_{pEDCs} = 433$). A total of 47 pEDCs were detected, and several less-frequently-detected pEDCs (<75%) were more commonly detected in off- compared to on-duty dog tags (conditional logistic regression). Of the 11 phthalates and fragrances detected most frequently (>75%), off-duty pEDC concentrations were strongly correlated ($r = 0.31$–0.82, $p < 0.05$), suggesting co-applications of phthalates and fragrances in consumer products. Questionnaire variables of "regular use of conventional cleaning products" and "fireplace in the home" were associated with select elevated pEDC concentrations by duty shift (paired *t*-test). This suggested researchers should include detailed questions about consumer product use and home environment when examining personal pEDC exposures.

1. Introduction

When engaging with a fire, firefighters experience known exposures to chemicals and other toxicants (Alexander and Baxter, 2014; Fent et al., 2017; Fent et al., 2018; Kolena et al., 2020; Poutasse et al., 2020; Stec et al., 2018). Detected chemicals include polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), volatile organic chemicals (VOCs), and phthalates (Alexander and Baxter, 2014; Dobraca et al., 2015; Fent et al., 2018; Kolena et al., 2020; Oliveira et al., 2017; Poutasse et al., 2020; Stec et al., 2018; Stevenson et al., 2015; Waldman et al., 2016), some documented as possible or probable carcinogens (IARC Working Group, 2010). Previous firefighter exposure studies have typically focused on carcinogens, but there is an increasing need to examine firefighter exposures to endocrine disrupting chemicals (EDCs).

EDCs, defined as "exogenous chemicals that interfere with hormone action," are characterized by mechanism of actions, rather than chemical structure (Gore et al., 2015; La Merrill et al., 2019; Zoeller et al., 2012). Hormones regulate and maintain many developmental and physiological processes in biological organisms, as well as homeostatic functions (Gore et al., 2015; La Merrill et al., 2019). Because hormones generally operate at very low concentrations, trace EDC concentrations may interact with high-affinity receptors in the body (Gore et al., 2015; La Merrill et al., 2019). These interactions can potentially result in adverse health outcomes, such as reproductive and cognitive impairment (La Merrill et al., 2019; Ventrice et al., 2013; Zarean et al., 2016). Because hundreds or more environmental chemicals could act as EDCs, this paper defines a "potential endocrine disrupting chemical" (pEDC) as an exogenous chemical suspected to interfere with hormone action, with

Abbreviations: DBP, Di-n-butyl phthalate; DIBP, Diisobutyl phthalate; DNNP, Di-n-nonyl phthalate; GC, Gas chromatograph; LOQ, Limit of quantitation; MS, Mass spectrometer; PPE, Personal protective equipment; PSD, Passive sampling device.

* Corresponding author at: Oregon State University, Department of Environmental and Molecular Toxicology, 1007 Agricultural and Life Sciences Building, Corvallis, OR 97331, United States.

E-mail address: kim.anderson@oregonstate.edu (K.A. Anderson).

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at least one study demonstrating endocrine-disrupting properties (Gore et al., 2015; La Merrill et al., 2019; Zoeller et al., 2012). A pEDC differs from an EDC in that causation of endocrine disruption has not been determined for a pEDC compared to an EDC.

Known pEDCs include phthalates, personal care products (PCPs), industrial-related chemicals, and pesticides. Although select phthalates and PCPs are confirmed endocrine disruptors (Ventrice et al., 2013), others remain unevaluated and may act as pEDCs. Phthalates are typically used as plasticizers to increase product flexibility, strength, and durability, with applications ranging from the automotive industry to medical products to children's toys to cosmetics (Koniecki et al., 2011; Ventrice et al., 2013; Viñas et al., 2015). PCPs can include fragrances (e.g. synthetic musks), with applications in air fresheners, deodorizers, laundry detergents, dishwashing detergents, cleaning supplies, and cosmetics (Koniecki et al., 2011; Llompart et al., 2013; Viñas et al., 2015). Exposure to pEDCs has been associated with several adverse health outcomes, such as increased incidence of cardiovascular disease, certain cancers, and reproductive effects in offspring (Lamb et al., 1987; Fu et al., 2020). These health outcomes overlap with those previously observed in firefighter epidemiological studies (Stevenson et al., 2015).

To prevent adverse health outcomes, it is useful to investigate what factors contribute to higher pEDC exposures. Previous literature has examined both environment- and participant-based factors associated with higher chemical exposures (Dixon et al., 2019; Fent et al., 2017; Fent et al., 2018; Gibson et al., 2019; Poutasse et al., 2019; Poutasse et al., 2020; Reddam et al., 2020). For instance, urinary flame retardant metabolite concentrations decrease with increased hand washing and house cleaning (Gibson et al., 2019); and concentrations of the flame retardant tris(1,3-dichloro-2-isopropyl) phosphate increase with longer driving commutes (Reddam et al., 2020) and more time spent on upholstered furniture (Poutasse et al., 2019). In the fire service specifically, urinary PAH metabolite concentrations varied by task on the fireground (Fent et al., 2018; Sjöström et al., 2019), but firefighter pEDC exposures have not yet been investigated in the context of participant behaviors and other environmental factors.

Previously, the military-style silicone dog tag was introduced as a new configuration of personalized passive sampling device, which sampled structural firefighters' on- and off-duty exposures to volatile and semi-volatile organic chemicals (VOCs, SVOCs) (Poutasse et al., 2020). The term "structural firefighter" here refers to a firefighter who mainly engaged with building, vehicle, and other urban fires, rather than wildfires. Other personalized passive sampling devices include silicone wristbands (Anderson et al., 2017; Bergmann et al., 2017; De Vecchi et al., 2019; Dixon et al., 2019; Dixon et al., 2018; Donald et al., 2016; Donald et al., 2019; Hammel et al., 2020; Hammel et al., 2016; Hammel et al., 2018; Harley et al., 2019; Hendryx et al., 2019; Kile et al., 2016; Manzano et al., 2019; O'Connell et al., 2014a; Paulik et al., 2018; Reddam et al., 2020). We previously demonstrated that firefighter PAH exposures differed by fire department call volume, duty shift, and number of fire attacks. This paper conducted a companion analysis of firefighter exposure assessments with a focus on pEDCs. The purpose of this paper is to examine the differences in firefighter silicone dog tag concentrations by 1) duty shift, 2) fire department call volume, and 3) questionnaire variables to determine what factors contribute to higher pEDC exposures.

2. Materials and methods

2.1. Methodology summary

All materials and methods were described in a previous publication (Poutasse et al., 2020). Briefly, the silicone dog tags (6.0 cm long by 2.5 cm wide by 0.3 cm thick; ~5.4 g; https://24hourwristbands.com, Houston, TX, USA) were vacuum oven conditioned at 300 °C for 12 h at 0.1 Torr (Vacuum Oven, Blue-M, model no. POM18VC, with Welch Duoseal pump, model no. 1405), and conditioned samplers were stored at 4 °C in sealed metal containers prior to shipment. Dog tags were shipped in sealed polytetrafluoroethylene bags before and after deployment.

All firefighter participants ($n = 56$) provided informed consent before participating, using procedures approved by the National Development and Research Institutes, Inc. (NDRI) Institutional Review Board (IRB00000634; Oregon State University (OSU) IRB Deferral 8313). Participants were provided with new personal protective equipment (GXTREME 3.0, Globe Manufacturing Company, LLC, Pittsfield, NH, USA; Quest Particle Barrier Hoods, Quest Fire Apparel Inc., Saratoga Springs, NY, USA) so the dog tags would not sample SVOCs embedded in used turnout gear. As shown in Fig. S1, firefighters were recruited from one high and one low call volume fire department in Kansas City metropolitan area (Poutasse et al., 2020), and completed a baseline questionnaire on demographics, occupational history, and behaviors prior to wearing the silicone dog tags (November 2018-March 2019). For 30 total on-duty days and 30 total off-duty days, firefighters ($n_{lowvolume} = 29$; $n_{highvolume} = 27$) wore one on-duty tag and one off-duty during all regular activities (e.g. eating, sleeping, showering, etc.). Each dog tag was sealed in separate polytetrafluoroethylene bags while not being worn.

After being returned to lab, the dog tag samples underwent post-deployment cleaning, liquid extraction, solid phase extraction, and instrument analysis as previously described (Anderson et al., 2017; Dixon et al., 2019; O'Connell et al., 2014b; Poutasse et al., 2019). As previously described, quality control samples ensured that data quality objectives were achieved (Poutasse et al., 2020). The samples underwent analysis using a 6890 N gas chromatograph (GC) with a 5975B Mass Selective Detector in full scan mode (see Supplemental Information for instrument parameters), and the semi-quantitative screening method included 1530 target analytes: 124 flame retardants, 185 industrial-related chemicals, 98 polycyclic aromatic hydrocarbons, 773 pesticides, 76 personal care products (PCPs), 14 phthalates, and 260 polychlorinated biphenyls (PCBs), dioxins, and furans (Bergmann et al., 2018; Dixon et al., 2019; Poutasse et al., 2020).

The screening method quantified concentrations within a factor of 2.5 of the true value (Bergmann et al., 2018). Because the GC–MS method was designed to be a broad screen, it had higher limits of detections and less sensitivity than a GC–MS operated in selected ion monitoring (Bergmann et al., 2018). Of the 1530 analytes, 433 are co-classified as pEDCs (http://fses.oregonstate.edu/masv-analyte-list) (Dixon et al., 2019). The full details of the analytical method were previously reported (Bergmann et al., 2018).

2.2. Statistical analysis

Statistical analyses were performed using SAS statistical software (JMP Pro version 14.0.0; SAS Institute Inc., Cary, NC) and R free software (CRAN R Project version 3.6.3).

While a frequency detection threshold of 75% is typically applied for statistical tests, less frequently detected analytes may be relevant when distinguishing occupational-specific chemical exposures and health outcomes (Czarnota et al., 2015). A detection threshold of 20% was set for regression analyses, which includes 22 pEDCs (Table S2). Binary presence-absence data for the 22 pEDCs underwent conditional logistic regression to investigate duty shift-specific relevance between paired dog tags (Woodward, 2013). The regressions were stratified by paired sample ID and included the clogit function from the survival package. Covariates, such as fire department and questionnaire data, were excluded from the regressions because they did not change within the paired samples.

For pEDCs detected in at least one dog tag, concentrations were converted to moles per dog tag (mol/tag). Concentrations below the instrument limits of quantitation (LOQs) were substituted with values equal to $LOQ/\sqrt{2}$ (Bergmann et al., 2018). Non-parametric Spearman's rho coefficients were calculated to explore pEDC concentration correlations ($p < 0.05$) for analytes detected in > 75% of the dog tags. Paired on- and off-duty samples were used to calculate log two-fold change (log2FC) values, which refers to the fold change of the on-duty concentration with respect to the off-duty concentration of a given pEDC.

For target analytes detected in at least 75% of the samples, the concentrations were assessed for normality. If analyte concentrations were approximately log-normally distributed (Kolmogorov's test, $p < 0.05$), then a $\log_{10}$-transformation was conducted and reassessed for normality. Previous results comparing on- versus off-duty paired dog tags and high- versus low call volume departments (paired *t*-test, *t*-test) have been previously reported for select phthalates and industrial-related chemicals (Poutasse et al., 2020). Paired t-tests (false discovery rate-adjusted p-value, Benjamini-Hochberg, alpha = 0.05) between on- and off-duty dog tag concentrations were conducted to examine the influence of select questionnaire variables.

3. Results and discussion

The study population demographics, while reported in a previous publication (Poutasse et al., 2020), were also given in Table S1.

3.1. Dichotomous pEDC detections

Of the 433 pEDCs included in the analytical method, forty-seven were detected in at least one dog tag (Fig. S2). These 47 pEDCs were listed in Table S2 by detection frequency. Phthalates and PCPs were the most commonly detected chemical categories across both fire departments and both duty shifts. pEDCs categorized as pesticides, PCBs/dioxins/furans, PAHs, industrial-related, and flame retardants were less frequently detected (Fig. S2, Table S2). By broadly screening for 433 pEDCs, the semi-quantitative analytical method provided an exploratory glimpse of each firefighters' unique exposure profile to associate with group membership (e.g. fire department).

3.2. Detection frequency and component relevance

When considering health outcomes in association with occupational chemical exposures, exclusively analyzing the most frequently detected analytes (>75%) may inadvertently ignore relevant but uncommon exposures. High detection frequency is necessary for many statistical tests, but a high threshold may exclude pEDCs detected only in on-duty or off-duty samples.

Using binary detection data with paired on- and off-duty firefighter dog tags, conditional logistic regression demonstrated that several pEDCs were significantly influenced by duty shift and low detection frequencies: dimethyl phthalate = 49%, and cinnamal = 38% (fragrance) (Fig. 1). For example, if a dog tag sampler was worn while on-duty, the odds of a positive dimethyl phthalate detection was multiplied by 0.46 (Fig. 1). In other words, dimethyl phthalate and

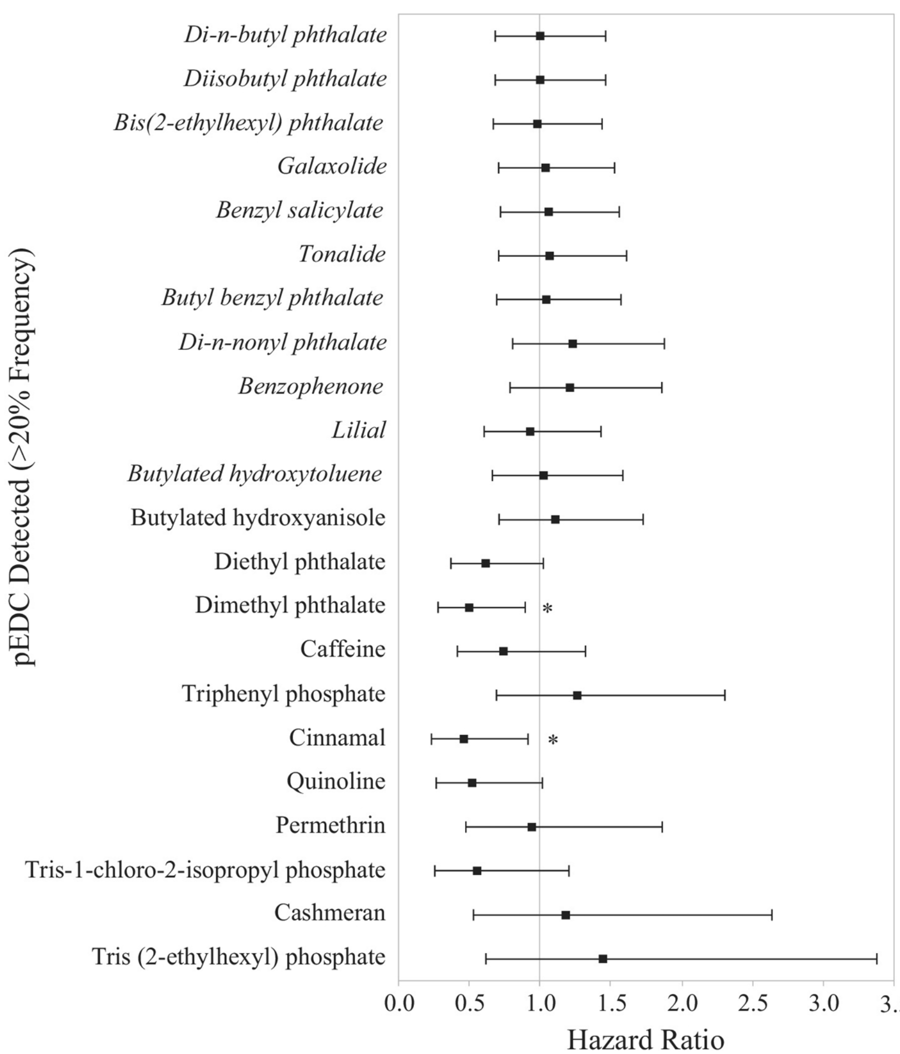

Fig. 1. Conditional logistic regressions were conducted for individual pEDCs by binary presence-absence data using paired on- and off-duty firefighter dog tag samples. pEDCs in *italics* were detected in over 75% of the dog tag samples. *: $p < 0.05$.

cinnamal were more commonly detected in the off- than on-duty dog tag samples. Both pEDCs are commonly detected in personal care products (Koniecki et al., 2011; Llompart et al., 2013; Viñas et al., 2015), which may act as potential off-duty sources. Diethyl phthalate (60%) and quinoline (36%; industrial-related) detections may also be dependent on duty shift, given a suggestively significant coefficient value for duty shift (Fig. 1).

Interestingly, numerous firefighter pEDC exposures overlapped between home and occupational environments when analyzed as binary presence-absence data. This result suggests that less-commonly-detected chemicals may be important to consider in future personal chemical exposure studies via alternative statistical analyses.

3.3. Duty shift differences

For pEDCs detected in > 75% of the dog tags, Spearman's coefficients were more positively correlated when worn off-duty Fig. 2A) compared to on-duty (Fig. 2B). These 11 pEDCs were categorized as either phthalates or PCPs, with the exception of butylated hydroxytoluene (i.e. industrial-related). The on-duty vs. off-duty correlation coefficient trend remained when the fire departments were considered separately (Fig. S3).

The significant off-duty positive correlations between phthalates and PCPs (Fig. 2A) may be due to co-exposures from fragranced consumer products (Llompart et al., 2013). Phthalates are used as a solvent base for fragrances, and have been quantified in a wide variety of consumer products, including deodorant, anti-perspirant, hair gel, aftershave, cologne, and glass cleaner (Al-Saleh and Elkhatib, 2016; Koniecki et al., 2011; Llompart et al., 2013; Viñas et al., 2015). While off-duty, a firefighter may use a few discrete fragrance-containing products. By contrast, while on-duty and at the fire department with other people, a firefighter may interact with many more phthalate- and PCP-containing products. With many potential pEDC sources, this difference may have masked the on-duty pEDC concentration correlations for this study population (Fig. 1B).

3.4. Participant characteristics and behaviors

To examine the influence of different questionnaire variables, we examined the firefighter dog tags as paired duty shift samples. This approach was appropriate because the pEDC exposures were highly individualized by firefighter participant, as shown in previous work (Poutasse et al., 2020) and in the pEDC log2FC profiles (Fig. S4). Paired t-tests (on-duty vs. off-duty; false discovery rate-adjusted p-value, alpha = 0.05) between different questionnaire variables are shown in Fig. 3A and 3B. T-tests (high vs. low; 2-sided p-value) between variables were stratified by duty shift (Fig. S5A-B).

Unlike the previous publication with PAHs (Poutasse et al., 2020),

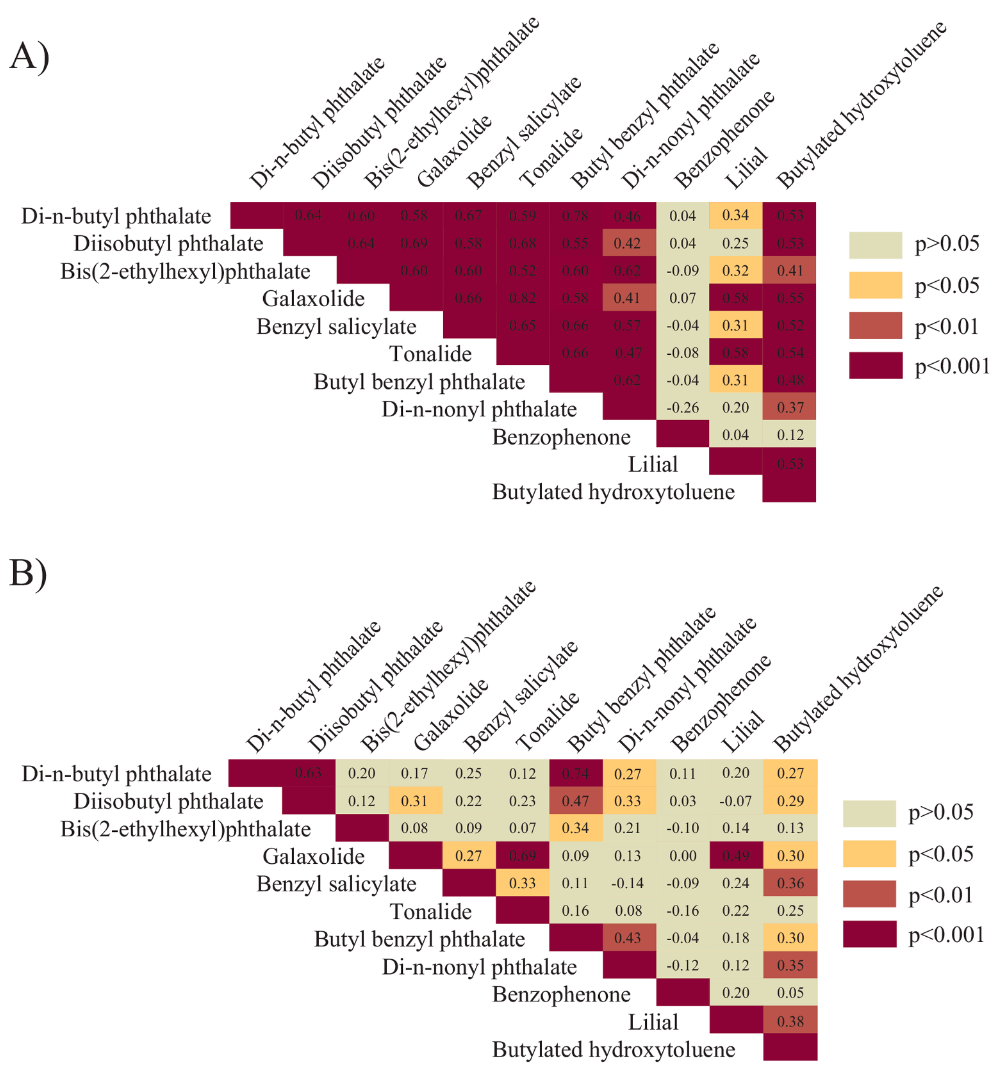

Fig. 2. Spearman's correlation coefficient matrices were calculated for pEDCs detected in >75% of the silicone dog tags. Target analytes are organized by detection frequency (top to bottom, left to right) for A) off-duty and B) on-duty dog tags.

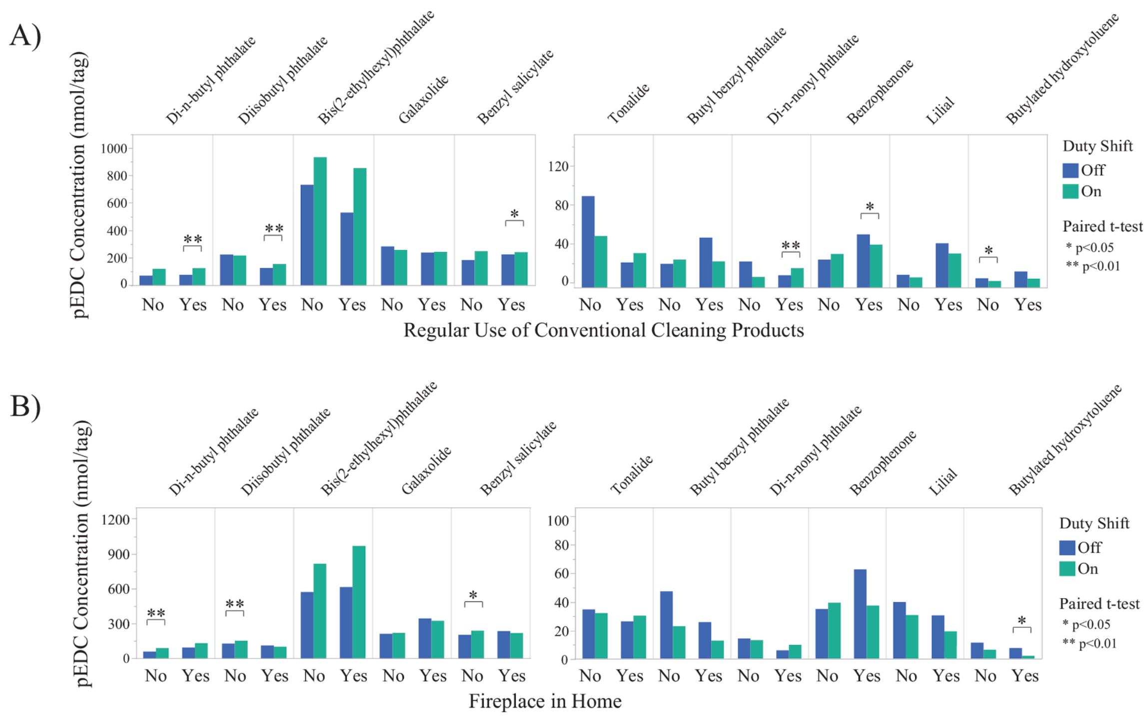

Fig. 3. Paired t-tests (false discovery rate-adjusted p-value, alpha = 0.05) were conducted on firefighter dog tags by duty shift to examine the influence of different questionnaire variables: A) regular use of conventional cleaning products and B) having a fireplace in the home.

the number of fire attacks a firefighter participated in was not correlated with pEDC concentrations (Fig. S6). However, other questionnaire variables did demonstrate subtle trends with the commonly detected pEDCs at the high call volume department: use of conventional cleaning products and having a fireplace in the home.

3.4.1. Conventional cleaning products

Regular use of conventional cleaning products was associated with higher on-duty pEDC concentrations: di-n-butyl phthalate (DBP), diisobutyl phthalate (DIBP), benzyl salicylate, and di-n-nonyl phthalate (DNNP) (Fig. 3A). These associations may have been attributable to co-mixtures of phthalates and fragrance compounds in consumer cleaning products. Cleaning products, such as dish soap, can contain fragrances with a phthalate solvent base (Al-Saleh and Elkhatib, 2016; Koniecki et al., 2011; Llompart et al., 2013; Viñas et al., 2015), and these products may be used more frequently while on-duty for turnout decontamination procedures (Fent et al., 2018). In Fent 2018, decontamination procedures using dish soap were effective at reducing exposures to known carcinogens (e.g. PAHs) (Fent et al., 2018).

If departments use fragranced dish soap for decontamination following a fire incident, then firefighters may experience higher phthalate and fragrance exposures from conventional cleaning chemicals using phthalates as the carrier solvent for various fragrances. Between the two departments in this study, there may be different cleaning procedures for contaminated turnout gear. For instance, firefighters at the low call volume department may have a support team that assisted with decontamination, which may account for the lack of statistical differences with this study population (Fig. 3A). For departments interested in reducing on-duty phthalate exposures, fire chiefs might consider limiting the number of fragrance- and phthalate-containing cleaning products in-house.

3.4.2. Fireplace in home

Not having a fireplace or wood-burning stove in the home was associated with lower off-duty concentrations of DBP, DIBP, and benzyl salicylate compared to on-duty concentrations (Fig. 3B). These associations may have been related to a reduction of indoor particle emissions because phthalate concentrations generally increase with increasing temperature and increasing particle emissions (Lunderberg et al., 2019). Therefore, the absence of a fireplace or wood-burning stove could decrease phthalate concentrations in the home environment (Fig. 3B). Although combustion is a not major a source of fragrances (Balci et al., 2020; Kubwabo et al., 2012), benzyl salicylate also followed this phthalate trend, likely related to decreased particle emissions.

3.5. Limitations

There were several limitations associated with this work. First, the non-random firefighter recruitments were a small sample of convenience, and may not be representative of the wider US firefighter population. Second, this paper's working definition of a pEDC (i.e. suspected to have hormone-disrupting capabilities, mechanism of action not necessarily demonstrated) is very broad and may have categorized some analytes as pEDCs that should not be classified as such. Third, the semi-quantitative analytical method will quantify target analytes within a factor of 2.2 times of the true concentration (Bergmann et al., 2018; Poutasse et al., 2020), which allows for a large confidence interval and these results must be interpreted in that context. Fourth, multiple comparisons were made when exploring chemical detection associations with duty status in the conditional logistic regressions. This should be considered when interpreting significance in this section. Lastly, the firefighter baseline questionnaire was focused on possible carcinogenic exposures and not specifically designed to capture pEDC exposures. As such, potential pEDC sources explored in other studies may not be present in this analysis.

4. Conclusions

This pEDC-focused analysis followed our original study applying military-style silicone dog tags as personal PSDs to investigate firefighter chemical exposures. First, the conditional logistic regression models with binary independent variables indicated that less-frequently-detected pEDCs were important in distinguishing between on- and off-duty dog tags. Second, pEDC concentrations were more strongly correlated among the off-duty than the on-duty dog tags (Spearman's rho). This suggested that a firefighter interacted with a larger number of diverse phthalate- and PCP-containing products while at the fire department. The strong correlations also suggested that phthalate and fragrance source(s) could be co-applications in consumer products. Lastly, between on- and off-duty dog tags, specific pEDC exposure trends were observed between the different questionnaire variables of "regular use of conventional cleaning products" and "fireplace in the home." This is the first study to investigate firefighter pEDC exposures in association with questionnaire variables, and this datum may be used in future studies when examining firefighter chemical exposures in the context of consumer product use and household environments.

CRediT authorship contribution statement

Carolyn M. Poutasse: Methodology, Validation, Formal analysis, Investigation, Resources, Writing – original draft, Writing – review & editing, Visualization. **Christopher K. Haddock:** Conceptualization, Software, Formal analysis, Writing – review & editing, Supervision, Funding acquisition. **Walker S.C. Poston:** Conceptualization, Methodology, Resources, Writing – review & editing, Project administration, Funding acquisition. **Sara A. Jahnke:** Conceptualization, Methodology, Resources, Writing – review & editing, Project administration, Funding acquisition. **Lane G. Tidwell:** Methodology, Validation, Investigation, Writing – review & editing. **Emily M. Bonner:** Software, Formal analysis, Writing – review & editing, Visualization. **Peter D. Hoffman:** Conceptualization, Writing – review & editing, Project administration. **Kim A. Anderson:** Conceptualization, Methodology, Writing – review & editing, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Kim A. Anderson, an author of this research, discloses a financial interest in MyExposome, Inc., which is marketing products related to the research being reported. The terms of this arrangement have been reviewed and approved by OSU in accordance with its policy on research conflicts of interest. The authors have no other disclosures.

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Author Contributions

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Appendix A. Supplementary material

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