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### Journal

ACS Environmental Au, 3(5)

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### Publication Date

2023-09-20

### DOI

10.1021/acsenvironau.3c00011

Peer reviewed

# Fire and Oil Led to Complex Mixtures of PAHs on Burnt and Unburnt Plastic during the M/V X-Press Pearl Disaster

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Cite This: *ACS Environ. Au* 2023, 3, 319–335



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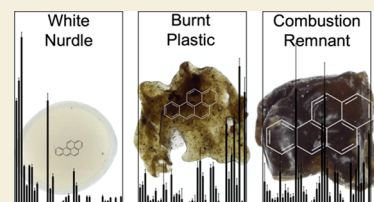
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**ABSTRACT:** In May 2021, the M/V *X-Press Pearl* container ship burned for 2 weeks, leading to the largest maritime spill of resin pellets (nurdles). The disaster was exacerbated by the leakage of other cargo and the ship's underway fuel. This disaster affords the unique opportunity to study a time-stamped, geolocated release of plastic under real-world conditions. Field samples collected from beaches in Sri Lanka nearest to the ship comprised nurdles exposed to heat and combustion, burnt plastic pieces (pyroplastic), and oil-plastic agglomerates (petroplastic). An unresolved question is whether the 1600+ tons of spilled and recovered plastic should be considered hazardous waste. Due to the known formation and toxicity of combustion-derived polycyclic aromatic hydrocarbons (PAHs), we measured 20 parent and 21 alkylated PAHs associated with several types of spilled plastic. The maximum PAH content of the sampled pyroplastic had the greatest amount of PAHs recorded for marine plastic debris (199,000 ng/g). In contrast, the sampled unburnt white nurdles had two orders of magnitude less PAH content. The PAH composition varied between the types of spilled plastic and presented features typical of and conflicting with petrogenic and pyrogenic sources. Nevertheless, specific markers and compositional changes for burning plastics were identified, revealing that the fire was the main source of PAHs. Eight months after the spill, the PAH contents of sampled stray nurdles and pyroplastic were reduced by more than 50%. Due to their PAH content exceeding levels allowable for plastic consumer goods, classifying burnt plastic as hazardous waste may be warranted. Following a largely successful cleanup, we recommend that the Sri Lankans re-evaluate the identification, handling, and disposal of the plastic debris collected from beaches and the potential exposure of responders and the public to PAHs from handling it. The maritime disaster underscores pyroplastic as a type of plastic pollution that has yet to be fully explored, despite the pervasiveness of intentional and unintentional burning of plastic globally.



**KEYWORDS:** microplastic, pollution, open burning, oil, combustion, weathering, maritime accident

## INTRODUCTION

On May 20, 2021, the M/V *X-Press Pearl* container ship caught fire 18 km off the coast of Colombo, Sri Lanka, following an explosion likely resulting from an ongoing, uncontained leak of concentrated nitric acid from one of its containers. At the time, the ship carried 1486 containers, of which 1214 held raw materials, hazardous chemicals, and finished products.<sup>1</sup> Included among the raw materials were 32 containers (~800 metric tons) of high-density polyethylene (HDPE) and 37 containers (~925 metric tons) of low-density polyethylene (LDPE) preproduction resin pellets or “nurdles” along with other plastics. Limited air quality measurements were collected during the fire.<sup>2</sup> Those taken downwind were elevated in CO, CO<sub>2</sub>, NO, NO<sub>2</sub>, SO<sub>2</sub>, and particulate matter relative to upwind locations.<sup>2</sup> Based on the difference in downwind and upwind CO and CO<sub>2</sub> measurements, we estimated the fire had a modified combustion efficiency<sup>3</sup> (MCE) of 0.89, thus, straddling flaming and smoldering conditions. Combustion is considered flaming for MCE ≥ 0.95 and smoldering for MCE 0.65–0.85.<sup>4</sup> Firefighting crews doused the fire by June 1, 2021, but salvage operations were unsuccessful in securing the ship before it sank on June 17, 2021. Recovery of the wreck is

estimated to be completed by mid-2023.<sup>5</sup> Complicating the accident, while the M/V *X-Press Pearl* burned, boat and satellite observations detected an oily sheen emanating from and around the ship heading northeast toward the coast.<sup>2,6</sup> Samples of the surface sheen collected in May and June 2021 determined it to be from the ship's underway fuel (an intermediate fuel oil).<sup>2</sup>

Five days after the fire began, an estimated 70 billion nurdles (~1680 tons) and pieces of burnt plastic began to litter the Sri Lankan coastline, making it the largest maritime plastic spill in history.<sup>1,2</sup> During the event, the plastic was exposed to combustion, heat, chemicals, and petroleum products that led to what was initially described as a “burnt nurdle continuum” of debris.<sup>1</sup> Subsequent analyses by James et al.<sup>7</sup> and others<sup>8–11</sup> revealed that the plastic was more discontinuous in its

Received: April 5, 2023

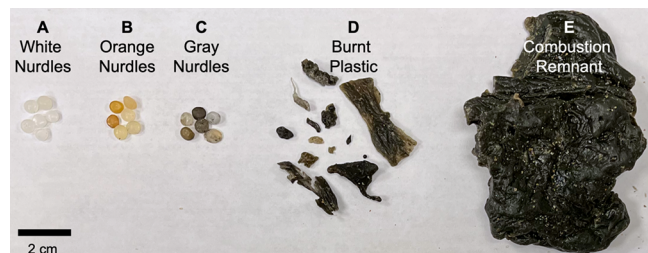
Revised: June 25, 2023

Accepted: June 29, 2023

Published: July 12, 2023



appearance than initially thought, diverging in its properties depending on the extent of its presumed exposure to the conditions of the ship fire.<sup>7</sup> Hence, we sorted the recovered plastic into five discrete, operationally defined visual categories (i) seemingly unburnt white nurdles (Figure 1A), (ii)



**Figure 1.** Representative image of white nurdles (A), orange nurdles (B), gray nurdles (C), burnt plastic pieces (D), and a combustion remnant (E).

discolored orange nurdles resulting from prolonged exposure to elevated temperatures near or below the melting point<sup>7</sup> (Figure 1B), (iii) discolored gray nurdles resulting from exposure to fire (Figure 1C), (iv) burnt plastic pieces (Figure 1D), or (v) large (>6 cm) charred, amalgamations of burnt plastic termed “combustion remnants” (Figure 1E).

The burnt plastic pieces and combustion remnants resembled pyroplastic, seemingly burnt or melted brittle plastic of neutral color and geogenic appearance.<sup>12,13</sup> Pyroplastic is a newly recognized form of plastic in the Anthropocene<sup>13</sup> that, so far, appears to enter the marine environment following large-scale fires (e.g., forest fires) and leakage of openly burned waste.<sup>14–18</sup> Since the first description of pyroplastic in 2019,<sup>19–21</sup> they, along with other forms of charred plastic, have been documented on beaches and waters across five continents (Africa,<sup>22</sup> Asia,<sup>18,23,24</sup> Europe,<sup>19–21</sup> North America,<sup>19,25,26</sup> and South America<sup>27</sup>) and have been found ingested by fish.<sup>22</sup> Little is known about the fate of pyroplastic in the ocean,<sup>13</sup> making the spill of burnt plastic from the M/V *X-Press Pearl* disaster a unique opportunity to monitor the weathering of time-stamped, geolocated pyroplastics and other plastic pieces when exposed to real-world conditions.

An initial assessment of the burnt plastic when it arrived onshore on May 25, 2021 (Figure S1) showed that pieces were either LDPE or HDPE, could be smaller (at least 0.5 mm) and larger (at least 6 cm) than nurdles, were brittle, and were at least threefold more chemically complex than seemingly unburnt white nurdles.<sup>1</sup> Notably, de Vos et al.<sup>1</sup> identified *n*-alkanes, alkenes, alkadienes, polycyclic aromatic hydrocarbons (PAHs), and petroleum-derived biomarkers and tentatively identified a phosphite antioxidant degradation product (2,4-di-*tert*-butylphenol) in the burnt material. Phthalates and benzotriazole UV-stabilizers, commonly found in plastics, were not detected.<sup>1</sup>

The International Pollutants Elimination Network (IPEN), in collaboration with the Sri Lankan Centre for Environmental Justice,<sup>28</sup> analyzed nurdles and “burnt lumps” collected in 2021 from four locations for a wide range of organic and inorganic compounds. The amounts of parent PAHs, a benzotriazole UV-stabilizer, bisphenols, and trace elements were reported. Residues from fluorinated firefighting foams were targeted but not detected. PAHs were expected because of the ship fire and leaking oil.<sup>29</sup> The amount of twelve 3- to 6-ringed parent PAHs (not including dibenzothiophene) varied with sampling

location and sample type ranging from ~20 to ~800 ng/g for nurdles and ~20,000 to ~50,000 ng/g for “burnt lumps” (Table S1).<sup>28</sup> Some variability may be related to differences (e.g., color) among the analyzed spilled nurdles.<sup>7</sup> Visually, the “burnt lumps” closely matched our definition of a combustion remnant (Figure 1E). The PAH content of the more abundant, smaller burnt plastic pieces (shown in Figure 1D) was not reported. To date, no study has characterized the sources of any PAHs associated with the spilled plastic. Apportioning source is critical to post-spill management of the plastic cleaned from beaches, recovery and monitoring efforts, and toxicological risk assessments for this spill and future efforts elsewhere. Establishing the source and character of PAHs associated with the spilled plastic may provide forensic markers of the accident and burnt plastic.

While recovery of the wreck is underway (estimated to be completed by mid-2023<sup>30</sup>) and the cleanup has progressed, the hazardousness of the 1600+ tons of recovered debris has yet to be comprehensively resolved.<sup>5,28</sup> Hazardous waste determination depends on whether a waste material can pose a substantial present or potential risk to human health or the environment when improperly treated, stored, transported or disposed of, or otherwise managed.<sup>31</sup> For a solid waste (spilled plastic) containing toxic constituents (PAHs) to be considered hazardous, requires determining the toxicity, concentration, partitioning, persistence, and degradability of the constituent, the extent to which the constituent bioaccumulates in the environment, quantity, plausibility for improper management, and historical impact of the waste, and the regulatory precedent for managing the waste—many of which are uncertain for burnt plastic (Table S2). This determination is necessary because recovered plastic remains siloed in warehouses, and its disposal method is undetermined. Stray spilled plastic continues to wash up on Sri Lankan beaches, and a fraction is expected to reach foreign coastlines.<sup>32</sup>

Herein, we address the abundance, source, and persistence of PAHs associated with the spilled nurdles and pyroplastic and the plastic’s potential for classification as hazardous waste. Our analyses of parent and alkylated PAHs associated with white, orange, and gray nurdles, burnt plastic pieces, and excised fragments of combustion remnants (Figure 1) reveal the amount, composition, source, and weathering of PAHs associated with the spilled plastic exposed to the marine environment.

## MATERIALS AND METHODS

### Sample Collection and Subsampling

Spilled plastics were collected in liter-sized bags from Pamunugama Beach, Sri Lanka, on May 25, 2021 (Figure S1), one of the closest shorelines to the ship. Based on simulated trajectories for the spilled plastic, the material transited from the ship to Pamunugama Beach in less than 5 days.<sup>33,34</sup> Additional spilled plastic was collected from Pamunugama Beach in May 2021 and on June 11, 2021 (22 days after the spill). Several months after the spill, stray plastic related and unrelated (i.e., different colors and shapes) was collected from Sarakkuwa Beach, Sri Lanka, on January 17, 2022 (242 days after the spill) (Figure S1). The recovered plastic was shipped to the Woods Hole Oceanographic Institution (Woods Hole, MA, USA) and stored at 4 °C as collected. All plastics were manipulated using solvent-rinsed stainless-steel tweezers. The material was visually sorted into five groups according to the categories operationally defined by de Vos et al.<sup>1</sup> and James et al.:<sup>7</sup> (i) white nurdles, (ii) orange nurdles, (iii) gray nurdles, (iv) burnt plastic, and (v) excised pieces of combustion remnant (Figure 1). Each discrete sample was prepared by pooling 10

random nurdles or nurdle-equivalent masses unless otherwise specified (Table S3). Only one to two samples of orange and gray nurdles were included in our study due to the limited availability of these types of spilled plastic. Among the spilled plastic collected from Pamunugama Beach in May 2021, an oil-plastic agglomerate was discovered and treated as a discrete sample (Figure S2). Samples were analyzed for their PAH content and assessed for petroleum biomarker diagnostic ratios.

### Solvent Extraction

Plastic samples were placed in 7 mL glass vials with Teflon-lined caps. For solvent extraction, 5 mL analytical-grade dichloromethane (DCM; Burdick & Jackson 71,739; purity >99.9%) was added to each vial, followed by 100  $\mu$ L of surrogate mixed standard (100 ng each of naphthalene- $d_8$ , acenaphthene- $d_{10}$ , phenanthrene- $d_{10}$ , chrysene- $d_{12}$ , and perylene- $d_{12}$ ; quantitative dilution of Agilent US-108 N-1 ISO 17025 certified compounds; purity > 99.5%). Each sample vial was then sonicated twice for 15 min with a 10 minute resting period between sonication steps. The resting period was to avoid any excess heat generated during sonication that could lead to the loss of any DCM. After sonication, sample vials were centrifuged for 15 min at 3000 rpm. Next, 100  $\mu$ L of an internal standard mixture (100 ng each of fluorene- $d_{10}$ ; Absolute Standards 71,739; purity > 99.5%, and benzo[*a*]pyrene- $d_{12}$ ; Absolute Standards 71,490; purity > 99.5%) was added to each sample vial. The surrogates and internal standard were also added in the same way to the blank, laboratory spike blank (LBS), and laboratory spike blank duplicate (LBSD). The LBS and LBSD were each spiked with 100  $\mu$ L (100 ng) of a U.S. Environmental Protection Agency 16 priority pollutant (EPA16) parent PAH spike solution (quantitative dilution of Absolute Standards 10,017; purity > 99%). EPA16 PAHs include naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, indeno[1,2,3-*c,d*]pyrene, dibenzo[*a,h*]anthracene, and benzo[*g,h,i*]perylene (Supporting Information).

### Gas Chromatography–Mass Spectrometry Quantitative PAH Analysis

The sample extracts were analyzed for PAHs by gas chromatography–mass spectrometry (GC–MS) (Agilent 6890 N GC/5975 inert MSD) by injecting 2  $\mu$ L of a 1 mL aliquot (20 ng/mL of the surrogates, internal standards, and spiked PAH, i.e., 40 pg on column) in both full scan and selected ion monitoring (SIM) mode using a 30 m  $\times$  0.25 mm i.d. DB-5 fused silica capillary column (J&W Scientific). The temperature program, acquisition, and quantification methods are detailed in Bera et al.<sup>35</sup> The 20 targeted parent PAHs were quantitated using calibration standards for each specific PAH. The 21 targeted alkylated PAHs were quantitated by applying the response factors of the parent PAHs (Supporting Information). Surrogate recoveries ranged between 65 and 119% except for naphthalene- $d_8$ , which was not used to calculate final recovery corrected concentrations. Laboratory blanks (all extraction steps without sample) contained no detectable analytes (practical quantitation limit 10 ng/g). LBS and LBSD average recovery of spiked PAHs were 89.4 and 86.3%, respectively, with a relative percentage difference (RPD) of 5.1%. The ratios of select petroleum biomarkers (hopanes and steranes) were obtained by analysis on GC–MS (Agilent 7890B GC/5977 inert MSD) in both full scan and SIM mode using a 30 m  $\times$  0.25 mm i.d. DB-5 MS UI fused silica capillary column (J&W Scientific).<sup>36</sup>

### PAH Source Apportionment Strategies

PAH source apportionment was comprehensively conducted using several strategies. PAH compositions were qualitatively assessed for characteristic features of pyrogenic and petrogenic sources. Percent pyrogenicity was calculated using the empirical source classification of PAHs proposed by Emsbo-Mattingly et al.<sup>37</sup> (Supporting Information). Specifically, percent pyrogenicity was calculated as the sum of the empirically classified pyrogenic PAH content normalized to the total PAH content. In the context of the *X-Press Pearl* disaster, we

used the percent dibenzothiophene content as a proxy for the extent of potential oil contamination of the spilled plastic, defined as the sum of the dibenzothiophenes normalized to the total PAH content (see section Evidence and molecular markers for petrogenic sources of PAHs). Calculated diagnostic ratios were compared to unadjusted and adjusted margins that define the lower or upper bounds for pyrogenic, mixed, and petrogenic sources. Unadjusted margins were those established for pyrogenic and petrogenic sources based on fossil fuel and biomass combustion-derived PAHs and those native to petroleum products. Recognizing that PAHs from burning plastic have not been included in defining these margins, we surveyed the literature and, from the reported PAHs, defined adjusted margins that account for the unique PAH features of burning plastic. Specifically, adjusted margins were defined as either the first or third quartile, where appropriate, of the set of respective diagnostic ratio values calculated from the surveyed literature for burning plastic PAHs (see section Validating conventional diagnostic ratio margins for burning plastic). Lastly, multivariate analyses were used, which compared the PAH compositions of the spilled plastic to those of reference sources (see section Multivariate analysis for source apportionment).

### Statistical Analysis

Statistical analyses were conducted using GraphPad Prism 9.3.1. Data are presented as mean  $\pm$  standard deviation,  $n$  = sample size. Data evaluated by ANOVA satisfied sample size requirements for, at minimum, an  $\alpha$  = 0.05 and  $\beta$  = 0.80, and normality and variance assumptions as determined by the D'Agostino–Pearson omnibus (K2), Anderson–Darling (A2\*), Shapiro–Wilk (W), and Kolmogorov–Smirnov (distance) tests for normality of the residuals and the Brown–Forsythe test for homoscedasticity. Groups were considered significantly different for a  $p$  value < 0.05. Sample sizes and statistical tests are included in the text and figure captions where appropriate.

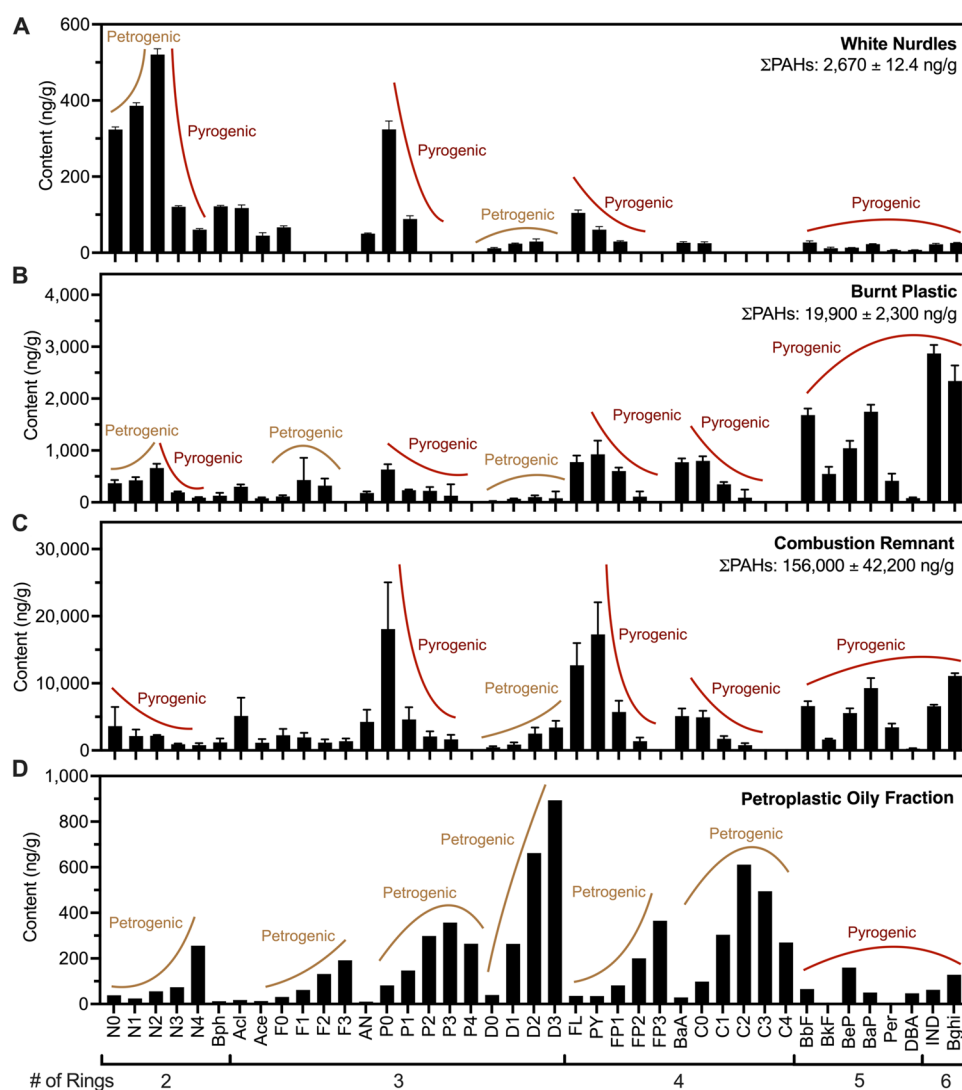
Principal component analysis (PCA) for apportioning source to the PAHs was conducted in GraphPad Prism 9.3.1 following the methods of Burns et al.<sup>38</sup> To remove the effect of PAH amount, the PAH content of each sample was normalized by the sum of the PAHs used in the analysis. The PCA was performed using the standardized method (scaling the data to a mean of 0 and standard deviation of 1), and PCs were selected based on eigenvalues greater than 1 (i.e., “kaiser rule”).

## RESULTS AND DISCUSSION

Samples of plastic collected 5, 22, and 242 days after the *M/V X-Press Pearl* fire were analyzed by GC–MS for 20 parent and 21 alkylated PAHs. First, we compared the PAH content and composition of the white nurdles, burnt plastic, and combustion remnant pieces (Figure 1). From these data, we applied several strategies to apportion the source(s) of PAHs: i) using conventional features and margins of diagnostic ratios for pyrogenic and petrogenic sources (e.g., oils and soot), ii) using features and margins of diagnostic ratios adjusted for burning plastic, and iii) performing a PCA using literature data for oils, various sources of soot, and burning plastic. Next, we assessed changes in PAH content and composition for white nurdles (Figure 1A) and burnt plastic (Figure 1D) collected 3 weeks and 8 months after the spill. Additionally, we compared the PAH content and composition of the different colored nurdles. Last, we discussed whether the spilled plastic should be considered hazardous waste and contextualized the scope of PAH contamination of the spilled plastic compared to levels associated with marine plastic debris globally.

### Trends and Variability of the PAHs Associated with the White Nurdles, Burnt Plastic, and Combustion Remnant

The sufficient availability of white nurdles, burnt plastic, and combustion remnant pieces of the spilled plastic collected on May 25, 2021, enabled us to assess the variability among these different fractions of spilled plastic when it first washed ashore.



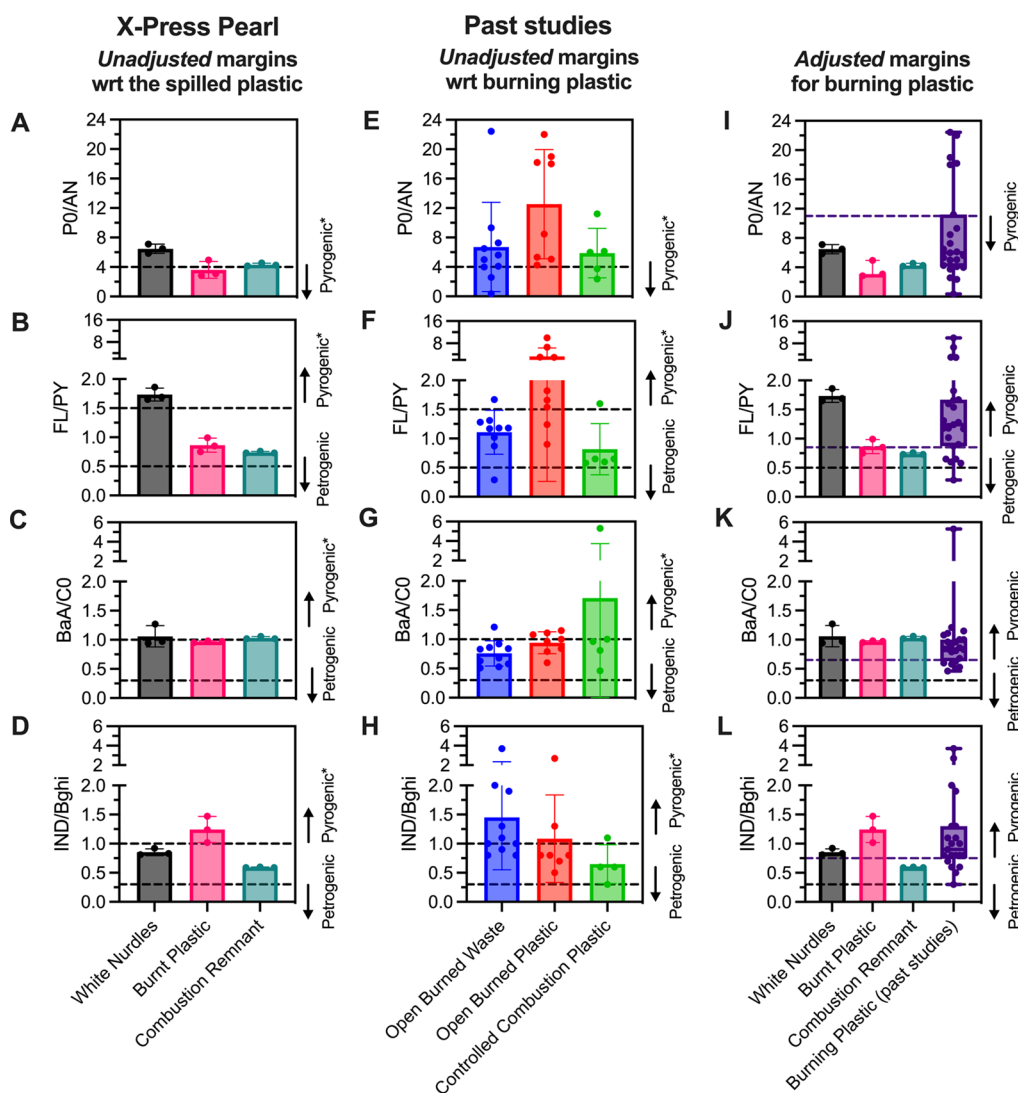
**Figure 2.** Annotated compositions of the 20 parent and 21 alkylated PAHs measured for white nurdles (A), burnt plastic (B), and combustion remnant pieces (C) collected on May 25, 2021, from Pamunugama Beach, Sri Lanka. PAH composition of oil from an oil-plastic agglomerate (petroplastic) collected on Pamunugama Beach, Sri Lanka, in May 2021 (D). The composition shows characteristics of moderately weathered oil.<sup>41,42</sup> In each panel, annotated lines following trends in the PAH composition were assigned based on recognized forensic signatures for pyrogenic and petrogenic PAHs from the combustion of fossil fuels and biomass and those native to petroleum products as described by Douglas et al.<sup>41</sup> Three samples of each plastic type were measured. Data are presented as mean  $\pm$  standard deviation. One sample of the petroplastic oily fraction was analyzed.  $\Sigma$ PAHs is the total amount of the PAHs measured. Data presented and acronym definitions are available in Tables S4–S7.

**PAH Content.** The total amounts of 2- to 6-ringed parent and alkylated PAHs were statistically different (log-transformed, one-way ANOVA, Tukey ad hoc,  $p < 0.0001$ ) among the white nurdles ( $2670 \pm 12.4$  ng/g,  $n = 3$ ), burnt plastic ( $19,900 \pm 2300$  ng/g,  $n = 3$ ), and combustion remnant pieces ( $156,000 \pm 42,200$  ng/g,  $n = 3$ ) (Tables S4–S6). In comparison, the total PAH content of virgin polyethylene nurdles has been reported to be minimal ( $<15$  ng/g).<sup>39</sup> We assumed the nurdles onboard the ship before the disaster had comparably minimal total PAH content. Variability in total PAH content trended with plastic type: white nurdles were largely uniform (%CV = 0.5%), the burnt plastic was more variable (%CV = 11.6%), and the combustion remnant was even more variable (%CV = 27.0%). Notably, the coefficient of variation of the white nurdles was within our analytical RPD (5.1%) for quantifying PAHs. The trend in PAH content variability mirrors the variability in shape, size, and color for these types of spilled plastic, in which white nurdles were

largely uniform in these qualities. In contrast, burnt plastic was much more heterogeneous.<sup>1</sup>

**PAH Composition.** The relative composition of the 41 target PAHs was specific to the type of spilled plastic (Tables S4–S7). The white nurdles were enriched primarily in naphthalene (N0), C<sub>1</sub>- and C<sub>2</sub>-naphthalenes (N1, N2), and phenanthrene (P0) (Figure 2A), which cumulatively accounted for  $\sim$ 58% of the total PAH content. Absent from the white nurdles were C<sub>1</sub>-, C<sub>2</sub>-, and C<sub>3</sub>-fluorenes (F1, F2, F3), C<sub>2</sub>-, C<sub>3</sub>-, and C<sub>4</sub>-phenanthrenes/anthracenes (P2, P3, P4), C<sub>3</sub>-dibenzothiophenes (D3), C<sub>2</sub>- and C<sub>3</sub>-fluoranthenes/pyrenes (FP2, FP3), and C<sub>1</sub>-, C<sub>2</sub>-, C<sub>3</sub>-, and C<sub>4</sub>-chrysenes (C1, C2, C3, C4). Notably, dibenzothiophene (D0) and C<sub>1</sub>- and C<sub>2</sub>-dibenzothiophenes (D1, D2) were detected.

Unlike the white nurdles, the burnt plastic was enriched in 5- to 6-ringed PAHs (Figure 2B). Indeno[1,2,3-*cd*]pyrene (IND), benzo[*ghi*]perylene (Bghi), benzo[*b*]fluoranthene (BbF), and benzo[*a*]pyrene (BaP) were the most prevalent PAHs,



**Figure 3.** Apportionment based on diagnostic ratios. Ratios of AN/P0 (A), FL/PY (B), BaA/C0 (C), and IND/Bghi (D) were calculated for the white nurdles, burnt plastic, and combustion remnant pieces, respectively. Data points are for plastic samples collected from Pamunugama Beach, Sri Lanka, on May 25, 2021 (Tables S4–S6). Ratios of AN/P0 (E), FL/PY (F), BaA/C0 (G), and IND/Bghi (H) were calculated for open-burned municipal waste, open-burned plastic, and the controlled combustion of plastic collated from the literature,<sup>45–49,52,57–59</sup> details of which are available in Table S8. Dashed lines in panels A–H represent phenomenologically defined margins for pyrogenic and petrogenic sources based on fossil fuel and biomass combustion-derived PAHs and those native to petroleum products<sup>42</sup> (i.e., margins *unadjusted* for burning plastic). Ratios of AN/P0 (I), FL/PY (J), BaA/C0 (K), IND/Bghi (L) for the data presented in panels A–H in which data collated from the literature for open-burned municipal waste, open-burned plastic, and the controlled combustion of plastic was collectively termed “burning plastic” and presented as a box (median, first, and third quartile) and whiskers (min and max) plot. Dashed lines in panels I–L represent margins *adjusted* for burning plastic. Adjustments to each margin were defined, where appropriate, as either the first or third quartile of the burning plastic literature data set. Details regarding each diagnostic ratio are included in Table S9. In the figure, “wrt” means “with respect to”.

accounting for 38 to 46% of all targeted PAHs. Compared to the white nurdles, additional alkylated PAHs were detected, including F1, P2, P3, D3, FP2, C1, and C2. Several of the more alkylated PAHs (P3, D3, FP2, and C2) were detected in only some of the samples of burnt plastic.

While the burnt plastic favored 5- to 6-ringed PAHs, the combustion remnant pieces favored 3- to 4-ringed PAHs (Figure 2C). P0, FL, and PY were the three most enriched PAHs in the combustion remnant pieces, constituting ~26% of the measured PAHs. The combustion remnant pieces included F3, and one sample had a relatively minute amount (~4 ng/g) of C4. The distribution of 5- to 6-ringed PAHs for the combustion remnant mirrored that of the burnt plastic with minor deviations only in the relative amounts of IND and BaP.

The distribution of 2- to 4-ringed PAHs was unique to the combustion remnant. No sample collected from Pamunugama Beach, Sri Lanka, on May 25, 2021, had detectable amounts of P4, FP3, and C3. The differences in the amount, composition, and variability of PAHs associated with the spilled plastic provided additional evidence that each material experienced unique conditions during the maritime disaster.

#### Apportioning the Source of the PAHs

Apportioning pyrogenic and petrogenic sources for PAHs is critical to forensic and toxicological analyses. Additionally, the relative bioavailability of PAHs largely separates by source;<sup>40</sup> thus, defining the source of the PAHs informs the risk assessment of past and future cleanup and disposal operations. Confounding what would appear to be a straightforward

apportionment (ship fire, i.e., pyrogenic) was that during the ship fire, some refined petroleum products leaked and created a sheen near and around the ship and presumably acted as a source of petrogenic PAHs for the spilled plastic.<sup>2</sup> Therefore, to apportion the source of the PAHs associated with the plastics, we employed recognized features of PAH compositions from pyrogenic and petrogenic sources and common diagnostic ratios.<sup>41,42</sup> These approaches are well-established for identifying petroleum products and the combustion of fossil fuels and biomass; however, characteristic features and margins of diagnostic ratios for plastic combustion have yet to be determined.<sup>42,43</sup>

**Compositional Features Based on Conventional Pyrogenic and Petrogenic Sources.** Visual inspection of the PAH composition for each type of spilled plastic provided insight into their source.<sup>29,44</sup> Pyrogenic sources are typified by high molecular weight PAHs and greater parent PAH abundance relative to their alkylated homologs (e.g.,  $N0 > N1 > N2 > N3 > N4$ ).<sup>41</sup> In contrast, unweathered petrogenic sources are typified by bell-like distributions of parent and alkylated PAHs with maximums at either the di- or tri-alkylated PAHs (e.g.,  $N0 < N1 < N2 \approx N3 > N4$ ).<sup>41</sup> The  $N0$ – $N4$  content of the white nurdles and burnt plastic did not fit these characteristics (Figure 2A,B). They presented an incomplete bell-like shape— $N0 < N1 < N2 > N3 > N4$ . It is unusual for lightly weathered petroleum products to be more enriched with  $N0$  than  $N3$ .<sup>41,42</sup> The other families of PAHs (parent and alkylated homologs) more readily fit conventional characteristics of pyrogenic and petrogenic sources (Figure 2A–C). Considering features characteristic of conventional pyrogenic and petrogenic sources indicated a mixed source for each type of spilled plastic.

**Empirical Classification Based on Conventional Pyrogenic and Petrogenic Sources.** Emsbo-Mattingly et al.<sup>37</sup> proposed an empirical classification of PAHs as either pyrogenic, petrogenic, mixed, or biogenic (Supporting Information). Results obtained using this approach for plastic collected on May 25, 2021, from Pamunugama Beach, Sri Lanka, suggested that the PAHs associated with the white nurdles, burnt plastic, and combustion remnant pieces were ~16, ~74, and ~60% pyrogenic, respectively (Tables S4–S6). By this method, the reduced pyrogenicity of the white nurdles was due to the predominance of  $N0$ – $N2$ . However, the combustion of plastic, particularly polyethylene, has been shown to be enriched in  $N0$  and  $N1$ , confounding this approach (Table S8).<sup>45–52</sup> The reduced pyrogenicity of the combustion remnant relative to the burnt plastic pieces was attributed to its greater  $P0$  content. The source of  $P0$  is empirically classified as mixed.<sup>37</sup>

**Diagnostic Ratios Based on Conventional Pyrogenic and Petrogenic Sources.** Numerous diagnostic ratios have been correlated to pyrogenic and petrogenic sources.<sup>43,53–55</sup> We calculated 15 diagnostic ratios (Table S9), which indicated a mixed source skewing pyrogenic. The pyrogenic index proposed by Wang et al.<sup>53</sup> supported a pyrogenic source for the PAHs associated with each type of spilled plastic. Values were larger than those of heavy fuel and bunker oils and comparable to soot. Ghetu et al.<sup>55</sup> suggested that the ratios  $A0/(AN + P0)$ ,  $P0/AN$ ,  $P1/(AN + P0)$ , and  $(FL + PY)/(FL + PY + FP1)$  are definitive of source. Values of these ratios indicated a mixed or pyrogenic source for each type of spilled plastic (Table S9). Source determination by other common ratios was less definite (Figure 3A–D). Overall, apportioning

the source of the PAHs by diagnostic ratios yielded a mixed source that skewed pyrogenic but once more, based on margins defined from nonplastic sources.

**Validating Conventional Diagnostic Ratio Margins for Burning Plastic.** There are no known proposed margins of diagnostic ratios for the burning of the most common types of plastic found in the environment.<sup>16,42,43</sup> Conventional margins for petrogenicity and pyrogenicity have been defined phenomenologically from PAHs in petroleum products and the combustion of fossil fuels and biomass rather than from the combustion of plastic and plastic-containing waste.<sup>42,43</sup> Few studies have quantified PAHs produced during the open burning of plastic and mixed waste.<sup>46,56</sup> Those that did, reported limited target lists (primarily the EPA16 PAHs) and did not propose margins of diagnostic ratios in the context of burning plastic and other organic matter (Table S8). Like other combustion-derived PAHs,<sup>29</sup> those from burning plastic and its mixtures depend on flaming conditions, temperature, and fuel composition.<sup>48,49,52</sup> Comparing reports of PAHs from open-burned municipal waste, open-burned plastic, and the controlled combustion of plastic collated from the literature<sup>45–49,52,57–59</sup> (Table S8), burning plastic tends to be abundant in  $N0$  and  $P0$ . The composition of PAHs for burning plastic differs distinctly from those produced during the combustion of other fuel sources (e.g., biomass<sup>60</sup>) and those intrinsic to petroleum products.<sup>41</sup> For these previous studies, we calculated diagnostic ratios for the reported PAHs produced from burning plastic in the absence of petroleum. We found their values could lean toward mixed or petrogenic sources when compared to conventional margins of diagnostic ratios (Figure 3E–H). Sometimes, the margins were too high (e.g.,  $FL/PY$  and  $IND/Bghi$ ). In another instance, the ratio  $AN/(AN + P0)$  did not definitively indicate a pyrogenic source for PAHs from burning plastic (Table S8). This contrasts with the results of Ghetu et al.,<sup>55</sup> who concluded that this ratio was decisive in distinguishing pyrogenic and petrogenic sources. Conversely, margins could be appropriate for other ratios (e.g., the pyrogenic index).

Many conventional margins that distinguish between pyrogenicity and petrogenicity have been defined using standard reference materials for crude oils, shale oils, coal tar, and diesel particulate rather than those for burning plastic.<sup>55</sup> Thus, alternative margins are necessary to rely exclusively on diagnostic ratios for determining source when considering PAHs from the combustion of plastic and plastic-containing waste. From the limited reported data on the PAHs produced from burning plastic, we suggest adjusted margins for several diagnostic ratios that incorporate the trends for burning plastic (Figure 3I–L, Table S9). Although instructive, these proposed adjustments to established margins of diagnostic ratios are preliminary and do not obviate the need for burnt plastic standard reference materials to aid in future source apportionment of PAHs. Nevertheless, it appears that those for  $AN/P0$  and  $BaA/CO$  have promise.

**Evidence and Molecular Markers for Petrogenic Sources of PAHs.** Evidence supports the hypothesis that some of the PAHs associated with the spilled plastic were sourced from oil. Principally, detecting dibenzothiophenes suggested contamination by some refined petroleum products onboard or in the vicinity of the ship. In the limited amount of published work, none report the formation of dibenzothiophenes from burning polyethylene.<sup>61,62</sup> Due to elemental sulfur and other sulfur-containing cargo on the ship,<sup>1</sup> the reaction

Table 1. Apportionment Summary

plastic type	compositional features	pyrogenicity <sup>a</sup> (%)	diagnostic ratios <sup>c</sup> (unadjusted margins <sup>d</sup> )	diagnostic ratios <sup>c</sup> (adjusted margins <sup>e</sup> )	indication of oil contamination (%D0–D4 <sup>f</sup> )	PCA (EPA16)	PCA (EPA16 + alkylated)
white nurdles	mixed	16 <sup>b</sup>	pyro (67%) mixed (33%)	pyro (100%)	2.4	petro > pyro	pyro > petro
burnt plastic	mixed	74	pyro (78%) mixed (22%)	pyro (93%) mixed (7%)	1.3	pyro > petro	pyro > petro
combustion remnant	mixed	60	pyro (54%) mixed (46%)	pyro (54%) mixed (46%)	4.6	pyro > petro	pyro > petro

<sup>a</sup>Calculated as  $\frac{\sum \text{Pyro}}{\text{Total PAH}}$ ; based on empirical classification of PAHs by Emsbo-Mattingly et al.<sup>37</sup> <sup>b</sup>Reduced due to abundance of N0–N4. <sup>c</sup>Presented as source (% of calculable diagnostic ratios specifying the respective source). <sup>d</sup>Using margins based on fossil fuel and biomass combustion-derived PAHs and those native to petroleum products. <sup>e</sup>Using margins adjusted for burning plastic. <sup>f</sup>Calculated as  $\frac{\text{D0} + \text{D1} + \text{D2} + \text{D3} + \text{D4}}{\text{Total PAH}}$ ; this metric was used as a proxy measure for the semi-quantitative extent of oil contamination of the spilled plastic; oil from the petroplastic had %D0–D4 = 26.7%.

and combustion of these compounds to produce thiophenes and their analogs cannot be discounted.<sup>50,51</sup> Despite this possibility, contamination by a petroleum product is more likely the dominant source of dibenzothiophenes associated with the spilled plastic as these compounds are well-known petroleum constituents.<sup>63,64</sup> This is substantiated by (i) the visual confirmation of nurdles and burnt plastic bobbing among the spilled sheen observed near and around the ship,<sup>2</sup> (ii) our finding of an agglomerate of oil and spilled plastic (termed “petroplastic”) (Figure S2), and (iii) the detection of petroleum biomarkers (compounds that are intrinsic to petroleum products and commonly used to identify spilled oil) associated with the spilled plastic (Table S10).<sup>1</sup> Oil from the petroplastic was recovered and analyzed, providing evidence for the composition of some fraction of the petroleum products that the spilled plastic encountered (Table S7). Others determined that the oil observed around the ship was the ship’s underway fuel.<sup>2</sup> Diagnostic ratios and characteristic features of the PAHs from the petroplastic’s oily fraction suggest it was a fuel oil that had been moderately weathered (Figure 2D, Table S7).<sup>41,43,54,55,64</sup> However, we cannot make a definitive match without an original sample of the sheen or ship’s fuel oil. Owing to the spilled plastics’ more oxygenated surfaces,<sup>17</sup> their interactions with petroleum likely changed compared to those with unaltered plastic.<sup>65</sup> The formation of petroplastics and other possible variants of oil-plastic agglomerates (e.g., plastitars<sup>66,67</sup>) during the disaster suggests the need for further study to assess their environmental impact.<sup>68</sup>

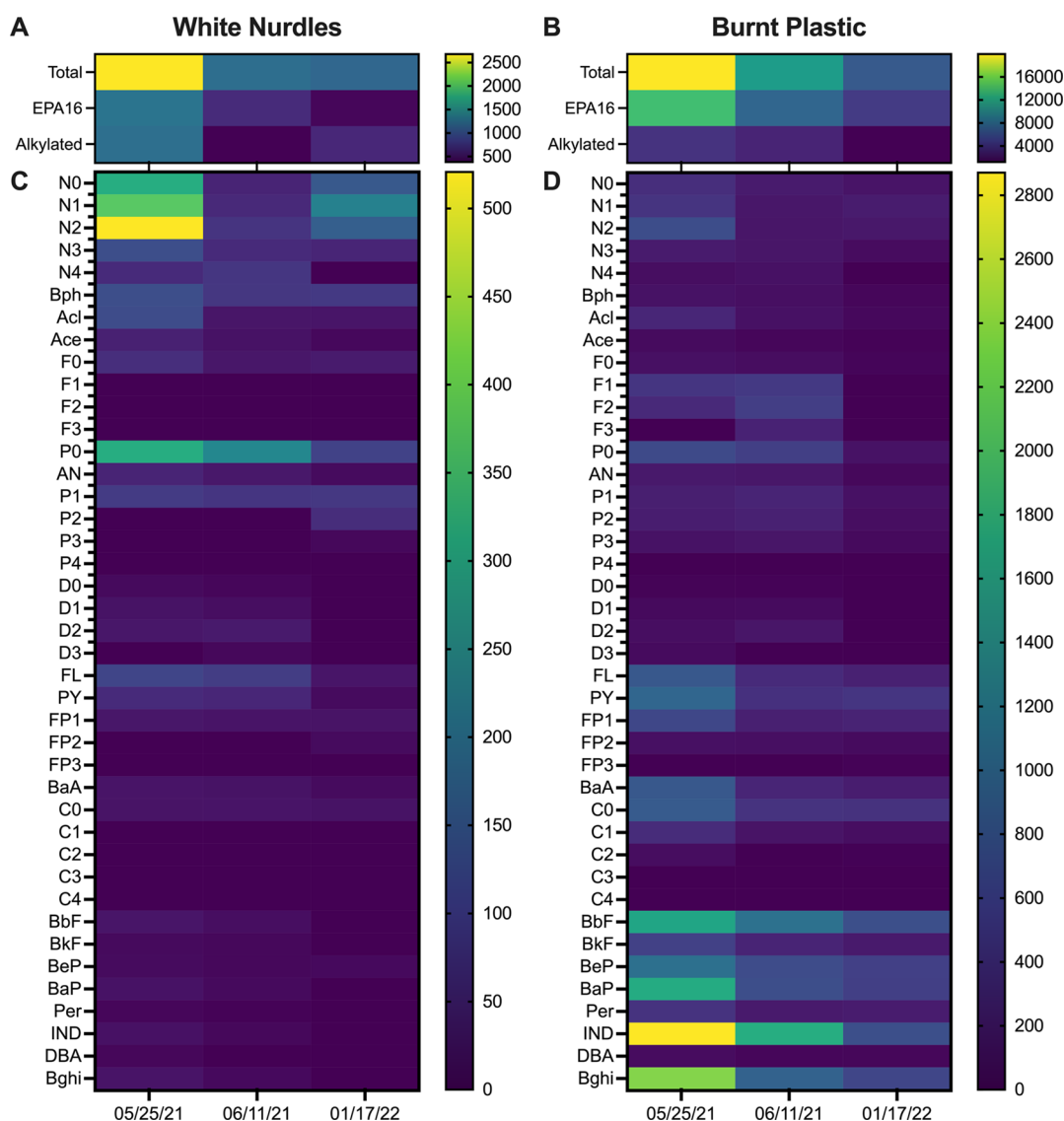
#### Multivariate Analysis for Source Apportionment.

Using PCA, each type of spilled plastic clustered and reinforced the determination that the PAH source skewed pyrogenic. A PCA was performed using the EPA16 PAHs (Section S1) so that PAH compositions reported in the literature for burning plastic<sup>45–49,52,57–59</sup> (Table S8) could be included in addition to common petrogenic and pyrogenic PAH sources (e.g., oils and soot).<sup>38</sup> The first two principal components accounted for 57% of the variability. The first principal component (PC1) was loaded with empirically defined pyrogenic PAHs<sup>37</sup> (FL, PY, BaA, C0, BbF, BkF, BaP, IND, DBA, Bghi), and the second principal component (PC2) was loaded with both empirically defined petrogenic PAHs<sup>37</sup> (N0, Acl, Ace) and pyrogenic PAHs (BbF, BaP, IND, Bghi). Thus, PC1 was considered a pyrogenic source component, and PC2 was considered a mixed source component. The white nurdles clustered with PAH compositions for open-burning plastic and plastic-containing waste (Figure S3). Interestingly,

only the PAH compositions for the controlled combustion of polyethylene at 800 and 850 °C clustered with those for open-burning plastic and the white nurdles. Owing to the details of the ship fire, presumably, many of the PAHs associated with the plastic are from the combustion of polyethylene. The clustering of the white nurdles with PAH compositions for the controlled combustion of polyethylene at 800 and 850 °C hints that the plastic onboard the ship may have burned around this temperature range. The association between these compositions was due to their relative abundance of N0. The white nurdles also clustered near diesel oil because of their mutual relative N0 content. When a PCA including alkylated PAHs was performed (Section S2), the white nurdles were no longer associated with diesel oil, reinforcing the conventional practice that alkylated PAHs would lead to a more robust source apportionment (i.e., the EPA16 PAHs are insufficient for apportionment<sup>69</sup>) (Figure S4). The burnt plastic aligned with PC1 in agreement with pyrogenic sources (soot) but deviated from them by aligning with PC2. The deviation was attributed to the burnt plastic’s greater relative abundance of N0 (Figure S3) relative to common pyrogenic sources. The combustion remnant was primarily aligned with PC1 and not PC2, suggesting a more limited mixed source than the burnt plastic. When a PCA was performed that included alkylated PAHs, the two clustered more together, closer to pyrogenic sources and further away from petrogenic sources (Figure S4).

From this evidence and related arguments, we suggest that the PAHs associated with the white nurdles originated from a myriad of sources (e.g., air and a light sheen of oil) that the nurdles may have encountered during the spill, their transit, and once on shore. For the burnt plastic and combustion remnant pieces, the sources appear to be predominantly from plastic burning in the presence of a petroleum product (Table 1). To date, there has not been a robust study establishing how to apportion the source of PAHs associated with the spilled plastic. In attempting to do so, we recognized the absence of established margins for diagnostic ratios of plastic combustion and relied on studies reporting primarily parent PAHs produced when burning plastic. The diagnostic ratios of the spilled plastic suggested a mixed source substantiated by qualitative characteristics of the PAH compositions and multivariate analyses. This was only possible by including the parent PAHs and their alkylated homologs in our analysis.





**Figure 4.** Weathering of the total, EPA16, and alkylated PAH content associated with the white nurdles (A) and burnt plastic (B). Weathering of the content and composition of the 20 parent and 21 alkylated PAHs associated with the white nurdles (C) and burnt plastic (D). Samples were collected from Pamunugama Beach, Sri Lanka, on May 25, 2021 (5 days since the fire) and June 11, 2021 (22 days since the fire) and Sarakkuwa Beach, Sri Lanka, January 17, 2022 (242 days since the fire). Data are presented as the mean of three samples and are available in [Tables S4 and S5](#).

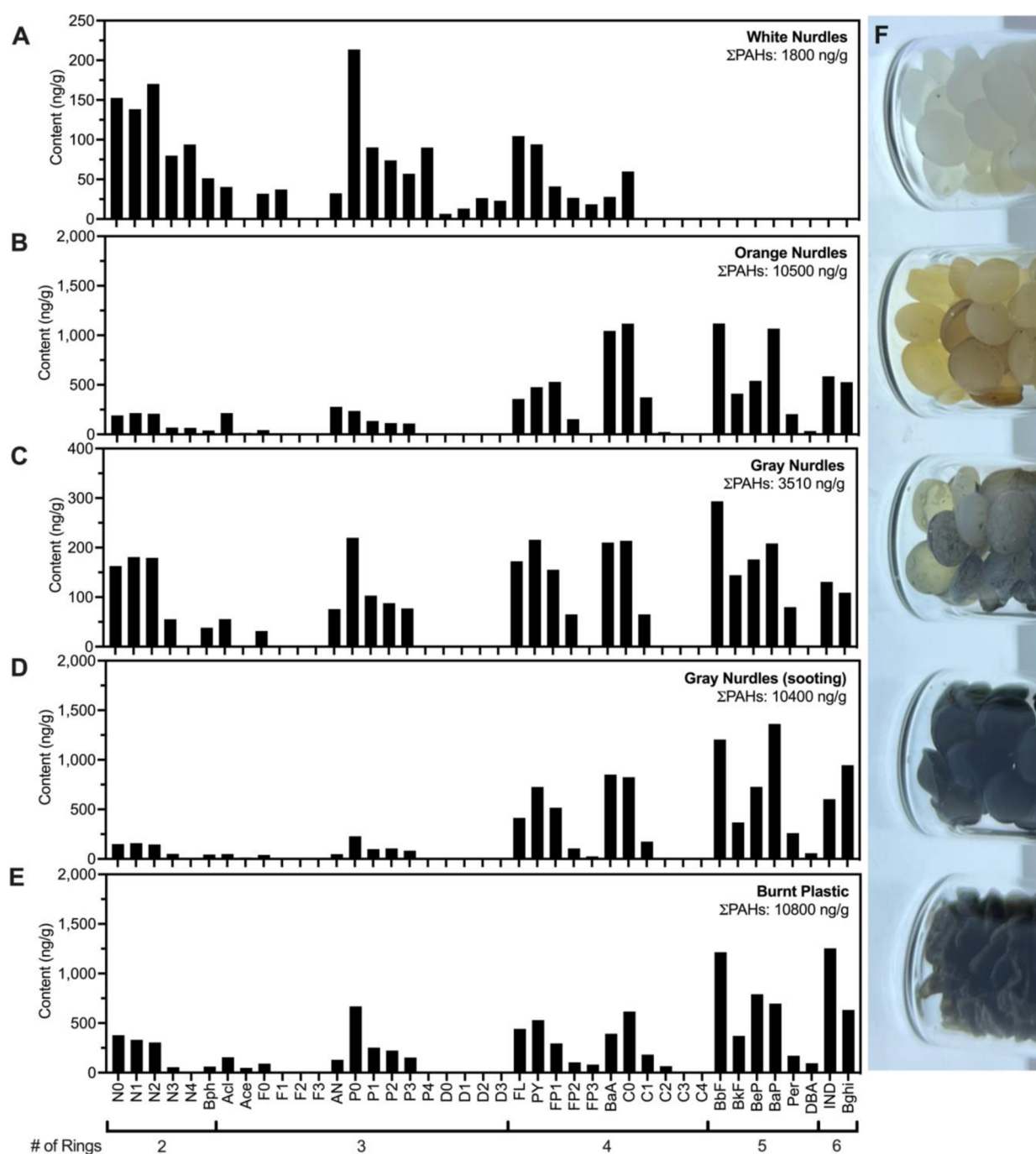
### Weathering of the PAHs Associated with the Spilled Plastic

Although the cleanup has removed much of the spilled plastic, stray nurdles and burnt plastic pieces have continued to wash up and be found on Sri Lankan beaches months after the spill. Analyses of these remaining pieces provided an opportunity to understand the fate of PAHs associated with nurdles and burnt plastic that are time-stamped and geolocated for their release into the marine environment.

The effects of weathering (e.g., evaporation, dissolution, emulsification, sedimentation, photooxidation, and microbial degradation) on the PAH compositions in the marine environment from oil spills are well studied.<sup>70</sup> The PAHs in petroleum weather nonuniformly and their lifetimes vary dramatically; some PAHs are quickly diluted by the environment and weathered with half-lives of days, while others may persist for decades.<sup>71,72</sup> Weathering processes would have commenced during the transport of the plastic to the shore from the ship. At the same time, the plastic could have

acquired additional PAHs from the seawater background and small oil slicks due to other boat and ship operations in the nearshore environment. By analyzing nurdles and burnt plastic collected weeks to months after the spill, we addressed how these processes affected the sorption and persistence of PAHs to the spilled plastic and the resultant content and composition associated with the spilled plastic exposed to real-world conditions.

The total PAH content of the spilled plastic decreased by nearly half within a few weeks. Due to sample limitations, weathering was only assessed for the white nurdles and burnt plastic. Twenty-two and 242 days after the fire, the PAH content of the white nurdles had decreased to  $1190 \pm 20.7$  ng/g ( $n = 3$ ) and  $1130 \pm 43.3$  ng/g ( $n = 3$ ), a reduction of 55 and 58%, respectively, from those collected on May 25, 2021 ([Table S4](#)). For the white nurdles, the total PAH content reduced quickly within the first few weeks and then plateaued. There was no statistical difference in total PAH content between white nurdles collected at 22 and 242 days (one-way



**Figure 5.** Compositions of the 20 parent and 21 alkylated PAHs measured for white nurdles (A), orange nurdles (B), gray nurdles without sooting (C), gray nurdles with sooting (D), and burnt plastic (E). Plastic was collected from Pamunugama Beach on the same day in May 2021 and was measured for one sample of each type of spilled plastic.  $\Sigma$ PAHs is the total amount of the PAHs measured. Representative image of the different sample types (F). Data presented and acronym definitions are available in Table S12.

ANOVA, Tukey ad hoc,  $p = 0.08$ ). While most of the EPA16 and alkylated PAH content decreased, the N0, N1, and N2 content decreased after 22 days and then increased at 242 days, resulting in the perceived stasis in total PAH content from 22 to 242 days after the spill (Figure 4A,B). The increase in N0, N1, and N2 content from 22 to 242 days after the spill could be due to nearshore sources. Analysis of plastic collected on January 17, 2022, that was presumed to not be from the spill because of its color and appearance (Table S3), corroborated this possibility by having amounts of naphthalenes comparable to those of the white nurdles and a PAH composition

characteristic of a petrogenic source (Table S11, Figure S5). Notably, this plastic had a comparable or greater amount of PAHs than the white nurdles, suggesting that PAH contamination of the white nurdles was within the range of plastic pollution endemic to the region when it was first released.

Nonetheless, the fact that naphthalenes, especially N0 and N1, remained associated with the nurdles long after the spill conflicts with the current view that naphthalenes are rapidly weathered in the environment (e.g., during oil spills).<sup>73–76</sup> Unlike their usefulness as indicators of petroleum weathering,

naphthalenes may be poor indicators of weathering processes for plastic-associated PAHs, potentially because typical processes (evaporation, dissolution, biodegradation, etc.) are hindered due to N0–N2 sorption to plastic. N0 and N1 are made when burning plastic. Thus, because of the ship fire N0 and N1 cannot be assumed to be petrogenic in their source. Consequently, naphthalenes as forensic indicators of petroleum contamination and weathering should be scrutinized; their source determination in the context of burning plastic and weathering characteristics (i.e., they do not appear to weather) defy convention.

Several PAHs associated with the white nurdles decreased exponentially with time in the marine environment. Decreases in the amount of N3, Ace, Acl, D0, D1, P0, AN, FL, PY, BaA, BbF, BkF, BaP, IND, and Bghi followed a one-phase exponential decay model (Figure S6), with half-lives and rate constants comparable to those previously reported for polyethylene pellets.<sup>39</sup> All other PAHs did not follow an exponential decay model after 242 days in the beach environment (Figure S6). The sorption of organic compounds to plastic depends on the material's surface properties.<sup>77,78</sup> James et al.<sup>7</sup> found that the nurdles that first washed ashore were more hydrophilic, more oxygenated, more crystalline, and could be dirtied with soot on their surface. However, it appears these initial transformations had a muted effect on the sorption kinetics of PAHs to the spilled plastic, given the similar rate constants of the white nurdles to those of unaltered polyethylene.<sup>39</sup>

As for the burnt plastic, 22 and 242 days after the spill, the PAH content of the burnt plastic had decreased to  $11,400 \pm 660$  ng/g ( $n = 3$ ) and  $6390 \pm 5280$  ng/g ( $n = 3$ ), a reduction of 43 and 68%, respectively, from those of burnt plastic collected on May 25, 2021 (Table S5). Unlike the white nurdles, the total, EPA16, and alkylated PAH content of the burnt plastic, on average, continued to decrease from 22 to 242 days after the spill rather than plateau (Figure 4C,D). The extent to which this loss was due to weathering and not variability of the plastic remains to be determined. Nonetheless, it reinforces the piece-to-piece heterogeneity of the burnt plastic, complicating the treatment of this type of contaminant. Additionally, the pyroplastic's black carbon (or soot) content presumably affects the sorption properties of any associated PAHs. Whether the partitioning of PAHs between the plastic and black carbon fractions of pyroplastic behaves like that of sediments<sup>79–82</sup> requires further study.

Most diagnostic ratios were largely the same after 242 days in the marine environment as when the plastic first spilled (Tables S4 and S5). This included P0/AN and FL/PY (Figure S7A,B), suggesting their use as robust indicators for determining PAH sources for plastics found in the marine environment. In contrast, BaA/C0 and, to an extent, the pyrogenic index decreased after 242 days toward a more petrogenic source determination (Figure S7C,D). Continued monitoring of stray plastic from the spill provides a unique opportunity to study the fate of PAHs associated with plastic in the ocean.

#### PAH Content and Composition Associated with the Different Colored Nurdles

So far, our analyses have focused on the white nurdles and pyroplastic; however, James et al.<sup>7</sup> estimated that ~5.4% or 91 tons of the spilled plastic was orange and gray nurdles (partial pyroplastics<sup>7</sup>) (Figure 1B,C). Despite sample limitations for

this fraction of spilled material, we confirmed that partial pyroplastics could be enriched in PAHs like that of pyroplastics. The PAH content of the different colored nurdles spanned an order of magnitude (1800 to 10,500 ng/g), with the lowest amounts for the white nurdles and much greater amounts for the partial pyroplastics. The orange and gray nurdles had different PAH compositions than the otherwise similarly shaped and sized white nurdles (Figure 5A–D). Sorting the gray nurdles according to the taxonomic groups defined by James et al.<sup>7</sup> revealed that gray nurdles categorized as “sooting” (those entirely black) were more enriched in PAHs than gray nurdles from the other taxonomic groups (Figure 5C,D). We hypothesized that the gray nurdles classified as sooting would have greater PAH content than the other taxonomic groups because their presumed greater amount of combustion-derived particulate matter is the source of their black color. The orange and gray nurdles had compositions similar to those of burnt plastic (Figure 5B–E); thus, their source apportionment trended like that of the burnt plastic (Table S12). The reasoning for the comparable PAH content of the orange nurdles to the gray nurdles and burnt plastic remains unclear. Because of their apparent greater enrichment for PAHs than the white nurdles and potentially still sizable spillage, the orange and gray nurdles should not be discounted in any hazardous waste determination of the spilled material.

#### Implications for the M/V X-Press Pearl Disaster

The plastic that spilled during the M/V X-Press Pearl disaster was enriched in PAHs, with some amounts being orders of magnitude greater than levels of concern for sediments<sup>83</sup> and allowable in consumer products.<sup>84</sup> Thus, the responders to this crisis were rightly justified in preemptively categorizing the material recovered from beaches as hazardous waste.<sup>2,85</sup> Sri Lankan hazardous waste regulations do not specifically identify PAHs as a cause for hazardous waste classification, though material and debris from a spill can qualify as hazardous waste.<sup>86,87</sup> For perspective, according to U.S. regulations, solid waste containing PAHs is not automatically classified as hazardous waste, either.<sup>88</sup>

By July 2021, 1610 tons of plastic, debris, and contaminated sand had been collected and siloed in warehouses.<sup>2</sup> This mass equates to slightly more than one-third of the total organic solid hazardous waste generated annually by Sri Lanka heavy industry.<sup>89</sup> If the spilled plastic is considered hazardous waste (Table S2), its significant quantity will likely pose continued challenges for Sri Lanka. As a signatory of the Basel Convention,<sup>90</sup> Sri Lanka must manage the spilled plastic within its borders. Under the Convention, plastic waste is restricted from transboundary movement except for plastic “destined for recycling in an environmentally sound manner and almost free from contamination and other types of waste.”<sup>90</sup> It is unlikely that the spilled plastic meets these criteria. Storage of the recovered material at depots and its transport have already led to secondary pollution,<sup>2</sup> necessitating countermeasures to mitigate these occurrences during the disposal process.<sup>2</sup> Using robust, tear-resistant storage containers or bags should be prioritized as one measure to prevent secondary spillage.<sup>2,91</sup> Currently, only one authorized facility (a cement plant) in the country can dispose of hazardous waste; it does so by incineration.<sup>92</sup> Sri Lanka's Central Environmental Authority has published guidelines for waste incineration to manage effluent gases from incineration facilities.<sup>93</sup> Other than

incineration, recycling has been proposed as another option. Within Sri Lanka, two plastic recycling facilities are authorized to handle plastic contaminated by chemicals and hazardous waste.<sup>92</sup> However, in light of the amount of PAHs associated with the spilled plastic, other possible contaminants not measured in this study, and the thermal degradation of the plastic, this option may pose challenges to meeting feedstock quality and could pose health risks in products made from the recycled material.<sup>94</sup> The difficulty of separating the white nurdles from the unusable burnt plastic in an efficient manner may also stymie this option.<sup>8</sup> Despite the good intentions to reuse the material, recycling should be considered with caution as a disposal solution. Another possibility is solvent rinsing or washing the plastic en masse (or after sorting) to remove contaminants. Similar logistical challenges, such as cleaning efficiency and throughput, will likely arise.<sup>95</sup> Conventional solid municipal waste methods may be options for fractions of the spilled plastic deemed non-hazardous waste. Recycling or upcycling reclaimed nurdles and other oceanic plastic has been suggested as a useful alternative to landfill disposal.<sup>94</sup> However, separating nonhazardous material from hazardous material may prove too difficult to justify as this requires multiple disposal options. Ultimately, the method for disposing of the recovered material must consider risk, expense, and resource availability.

Nurdles from spills are endemic.<sup>96</sup> Collecting all the spilled material is an unrealistic objective, posing the oft question, "How clean is clean?"<sup>2,97</sup> Following previous nurdle spills, cleanup efforts predominantly relied on community-based activities to recover spilled material.<sup>96,98–102</sup> After a spill in the North Sea, efforts focused on removing any piece of plastic from beaches.<sup>96</sup> However, this is a false equivalency because different plastics behave differently in the environment. Such an approach should be avoided, particularly as burnt plastic poses an uncertain risk due to its elevated amounts of PAHs and other contaminants.

The burnt plastic is also brittle, making it more likely to fracture and fragment into smaller nanoplastic and microplastic pieces.<sup>1,19</sup> Because of this process, beaches may artificially appear cleaner over time as pieces of the burnt plastic fragment into pieces undetectable by the human eye. Fragmentation of the burnt material may cause it to degrade faster as it becomes more accessible to microbial activity and sunlight because of increased surface area. However, these smaller pieces may prove more toxic and act as a "Trojan horse" for PAHs and other contaminants associated with the plastic.<sup>103–106</sup> This scenario necessitates various forms of toxicological assessments of the material.

The additional complexity of the *X-Press Pearl* disaster is that the hazardousness of the spilled plastic may change after weathering in the environment. Our findings suggest that the PAH burden of the nurdles and burnt plastic was reduced on average by 58 and 68%, respectively, 242 days after the ship fire. From the perspective of contamination, weathering appears to reduce the PAH content of the nurdles and pyroplastics. Nonetheless, plastics sorb contaminants (e.g., DDT and PCBs) while in the environment,<sup>107</sup> necessitating a study of the extent to which other contaminants adsorbed to the spilled nurdles, partial pyroplastics, and pyroplastics. We hypothesize that pyroplastics will adsorb hydrophobic organic contaminants more readily because of their presumed black carbon (soot) content, much like black carbon-containing sediments.<sup>79–82</sup>

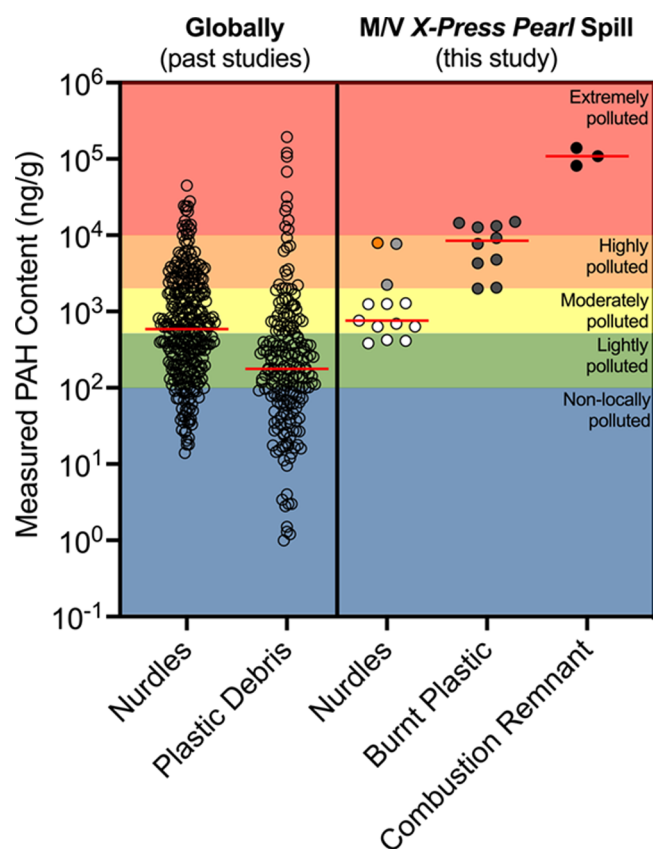
The bioavailability of pollutants associated with the spilled plastic is relevant to the plastic's riskiness. For instance, more environmentally weathered nurdles have been shown to have reduced in vitro bioaccessibility of PAHs compared to lightly environmentally weathered nurdles.<sup>108</sup> Black carbon can also reduce the bioavailability of pyrogenic PAHs while doing less so for petrogenic PAHs.<sup>40</sup> Additionally, in migration studies of PAHs from polyethylene using dermal simulants, after 24 h, no PAHs were detected to migrate from polyethylene into the simulant medium.<sup>109</sup> These findings suggest that PAHs associated with the spilled nurdles and pyroplastics may have limited bioavailability, despite the PAHs being at elevated amounts. Future work should address this critical uncertainty.

Toxicity from other exposure routes, such as ingestion, is of particular concern. For instance, the bioavailability of associated pollutants (e.g., PAHs) can be greater for ingested nurdles.<sup>107</sup> Moreover, nurdles are commonly found in the stomachs of birds and other marine life, which can cause a false sense of satiation, leading to starvation.<sup>110</sup> Comprehensive analyses have concluded that plastics, across their lifecycle, negatively impact human and ocean health.<sup>111</sup> These points contribute to the open question of whether to consider plastic itself as hazardous<sup>112</sup> regardless of the toxic constituents (e.g., PAHs, trace heavy metals, or other contaminants) that it may contain. For instance, following a minor container-related nurdle spill in New Orleans, LA, USA, in August 2020, to our knowledge, there has been no formal response (local, state, federal) to clean up the plastic in part because, at present, nurdles are not considered a "hazardous material" under the U.S. Clean Water Act.<sup>99,113</sup> Yet, plastic can cause harm.<sup>111</sup> As the impact of and response to the *M/V X-Press Pearl* disaster continue, it will be necessary to determine the spilled plastic's long-term fate, persistence, and toxicity to instruct future efforts.

### Implications Globally

Our findings showed that pyroplastics were enriched in PAHs relative to other marine plastic. To contextualize the PAH content of the spilled plastic, we compared our results to the PAH content measured and reported in the literature from 19 studies of plastic pellets (317 data points) and 15 studies of plastic debris (198 data points) found globally in the marine environment (Figure 6).<sup>39,108,114–143</sup> When they arrived onshore on May 25, 2021, the white nurdles had an EPA16 PAH content (~1260 ng/g) double the median for nurdles found globally in the marine environment (~589 ng/g). It should be noted that many of these other studies examined nurdles with a yellowness index of 30 to 50, assuming that the yellowing indicated that these nurdles had been in the environment long enough to accumulate significant amounts of PAHs.<sup>133</sup> In contrast, our measurements were made for white nurdles, i.e., those with minimal color development, that had only been in the environment for at most 5 days. When they arrived onshore, pieces of burnt plastic and combustion remnants had EPA16 PAH content greater than the majority of plastic debris sampled to date from the marine environment (Figure 6). In fact, compared to the values reported in our comprehensive literature survey of PAHs associated with nurdles and marine debris, one combustion remnant piece had the greatest measured PAH content (199,000 ng/g) to date of any plastic found in the marine environment.

Unlike previous reports for PAHs associated with oceanic plastic, the PAHs of the pyroplastics investigated in this study



**Figure 6.** Comparison of PAH content for the plastic spilled during the M/V X-Press Pearl disaster analyzed in this study and plastic found in the ocean globally. Ranges of pollution level are those defined according to Yeo et al.<sup>133</sup> Data presented for the nurdles and plastic debris were collated from literature-reported absolute, mean, or median values,<sup>39,108,114–143</sup> details of which are available in Table S13. A comparison of each study is included in Figure S8. Because the EPA16 PAHs were predominantly reported for the nurdles and plastic debris found globally, the PAH contents for the plastic investigated in this study are presented as the EPA16 PAHs. These 16 PAHs do not include some parent PAHs (e.g., benzo[*e*]pyrene), alkylated PAHs, or heteroatom-containing PAHs, which are known to contribute significantly to the total PAH content.<sup>144</sup> Relying on this abbreviated list for the spilled plastic misses 15–20% and 45–60% of the total measured PAHs for the pyroplastics and nurdles, respectively. The red lines indicate the median of the data. The color of each point corresponds to the color of the nurdle analyzed in this study (Figure 1).

were more attributed to combustion than petroleum sources.<sup>118,120,128–131,133</sup> Owing to their camouflaged appearance, only recently have pyroplastics been reported on beaches.<sup>19–21,23,27,145</sup> This fact prompted us to review previous reports for PAHs associated with oceanic plastic to determine whether descriptions, images, PAH compositions, and diagnostic ratios for the most highly contaminated samples reflected features of pyroplastics. From our assessment, samples from Mai et al.<sup>131</sup> and Chen et al.<sup>132</sup> had attributes suggestive of pyroplastics, and their PAHs were characteristic of pyrogenic sources (Section S3); nonetheless, weathering could skew their PAH composition.<sup>42</sup> Although circumstantial, this evidence highlights the potential pervasiveness, enrichment, and stability for PAHs in this form of plastic in the environment and the need for additional chemical markers to identify pyroplastics in environmental samples.

Pyroplastics enter the marine environment not only from a maritime disaster like that of the M/V X-Press Pearl and presumably other ship fires (e.g., M/V Golden Ray<sup>146</sup> and Felicity Ace<sup>147</sup>) but also from more frequent open burning of waste and large-scale fires.<sup>14–16</sup> Such material arises from the open burning of an estimated 970 million tons of plastic-containing waste each year.<sup>14,56</sup> Natural disasters—especially wildfires—can be another source of pyroplastic entering the environment.<sup>15,16,148,149</sup> Municipal water lines have already been shown to be contaminated by polyvinyl chloride and polyethylene pipes that melted and burned during wildfires.<sup>149,150</sup> Forms of plastic like those from the X-Press Pearl are likely produced by these actions, and remnants of this material presumably enter the environment along the same paths as other mismanaged waste.<sup>151</sup> Defining comprehensive chemical features for openly burned plastic is needed. It should prove helpful in monitoring efforts for air quality, waste management, pyroplastic pollution, and fires at the forest-urban interface.<sup>14,16,56</sup> Atmospheric transport of pyroplastic is also conceivable, given recent evidence for microplastic transport in this way.<sup>152–154</sup> Moreover, given their elevated amounts of PAHs, pyroplastics should be considered a potentially significant environmental pollutant with uncertain toxicity.

## ■ ASSOCIATED CONTENT

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsenvironau.3c00011>.

Abbreviations for PAHs; map of sample locations; image of oil-nurdle agglomerate; PCA for source apportionment using EPA16 PAH; PCA for source apportionment using EPA16 and alkylated PAH; PAH composition of background plastic; model fits for weathered PAHs; change in diagnostic ratios with time; PAH content measured by IPEN; table for hazardous waste determination of spilled plastic; table of sample details; PAH diagnostic ratios; details of PCA for source apportionment using EPA16 PAH; details of PCA for source apportionment using EPA16 and alkylated PAH; attributes of marine plastic debris in the literature suggestive of pyroplastic (PDF)

Spreadsheet of PAH data for all samples; PAH data from a literature survey of burning plastic; petroleum biomarker data; literature survey of PAH content for nurdles and marine debris (XLSX)

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## Notes

The authors declare no competing financial interest.

## ACKNOWLEDGMENTS

We would like to thank David Campion and Conor Bolas (ITOPF) for their helpful discussions and assistance in collecting samples, and Thomas Ryerson (Scientific Aviation), Philip Gschwend (MIT), Anna Michel (WHOI), Collin Ward (WHOI), Sarah Youngs (WHOI), and Jagoš Radović (University of Calgary) for their valued discussion. This work was supported by the Postdoctoral Scholar Program at the Woods Hole Oceanographic Institution (WHOI), with funding provided by the Weston Howland Jr. Postdoctoral Scholarship. Additional support was provided by The March Marine Initiative, a program of March Limited, Bermuda, through WHOI's Marine Microplastics Innovation Accelerator program, the Seaver Institute, and the Texas A&M University Research Foundation.

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