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# Screening the release of chemicals and microplastic particles from diverse plastic consumer products into water under accelerated UV weathering conditions

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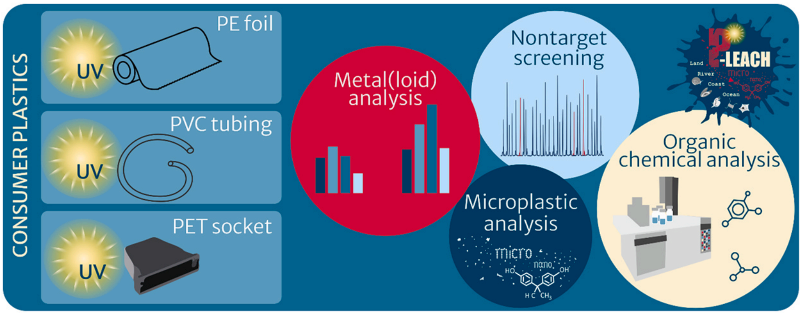
## HIGHLIGHTS

- Plastic products leach complex mixtures of particles, chemicals and metals into water.
- Leaching was more explicit under artificial UV irradiation compared to dark controls.
- Complementary analytical tools give a unique, comprehensive assessment.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

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## ABSTRACT

Photodegradation of plastic consumer products is known to accelerate weathering and facilitate the release of chemicals and plastic particles into the aquatic environment. However, these processes are complex. In our presented pilot study, eight plastic consumer products were leached in distilled water under strong ultraviolet (UV) light simulating eight months of Central European climate and compared to their respective dark controls

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Leaching
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Non-targeted screening
Elemental fingerprints
Particle analysis
Microplastics
Mass spectrometry

(DCs). The leachates and formed plastic particles were exploratorily characterized using a range of chemical analytical tools to describe degradation and leaching processes. These techniques covered (a) microplastic analysis, showing substantial liberation of plastic particles further increased under UV exposure, (b) non-targeted mass spectrometric characterization of the leachates, revealing several hundreds of chemical features with typically only minor agreement between the UV exposure and the corresponding DCs, (c) target analysis of 71 organic analytes, of which 15 could be detected in at least one sample, and (d) metal(loid) analysis, which revealed substantial release of toxic metal(loid)s further enhanced under UV exposure. A data comparison with the US-EPA's ToxVal and ToxCast databases showed that the detected metals and organic additives might pose substantial health and environmental concerns, requiring further study and comprehensive impact assessments.

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## 1. Introduction

Since 1950, about 9 billion tons of plastics have been produced globally [1-4]. About half of the plastics have ended up in landfills or have been dumped in the natural environment [5], and only less than 10 % have been adequately recycled [6].

Plastic products consist of various kinds of polymers in combination with versatile and processable mixtures of a variety of chemicals (> 13,000) [7]. Overall, more than 3200 monomers, additives, processing aids (e.g., lubricants), and non-intentionally added substances used in plastic manufacturing potentially exhibit hazardous properties [8,9]. Due to the known toxicity of certain components and their high release potential, specific plastic-associated chemicals are of particular concern, e.g., flame retardants, plasticizers, or heavy metals. However, only some consumer products are well studied [10], e.g., those intended for food [11] or medical applications [12]. Additive chemicals improve the performance, functionality, stability and durability of polymers and plastic products. However, if plastic products are exposed to water, the chemicals can leach from the products and become bioavailable [10], since these compounds are mostly not covalently bound to the polymer backbone. Hence, plastic debris that enters the (aquatic) environment may be of concern, as release of chemicals into the environment [13], marine organisms [14], seabirds [15] and algae [16,17] has been described in the literature. Experimental studies have also revealed possible toxic effects on both plants [18] and animals [19].

In the aquatic environment, plastic waste is transported, weathered, degraded, and altered chemically and physically. Several weathering factors including UV-light, fluctuating temperatures, biofouling, hydrolysis, and mechanical stress have been discussed [20-23]. Photodegradation is one of the major driving forces inducing the disintegration of plastics in the environment, including the reduction of chain length, surface oxidation, formation of microplastics and loss of mechanical properties [24-29].

The polymer backbone of floating plastics breaks down and can release low molecular-weight dissolved organic matter like carboxylic acids, monomers [30] and more complex hydrocarbons or halogenated compounds under simulated marine conditions, and photodegradation can be a key mechanism for fragmentation of plastic debris floating in the oceans [24]. Additionally, photodegradation of plastics induces the formation of micro- and nanoplastic particles [31,32].

From an analytical chemistry perspective, leaching studies focusing on plastic-associated chemicals and their transformation products are challenging, since many consumer products contain (and potentially release) highly complex and mostly unknown chemical mixtures, with varying characteristics across polymer types and products [30,18,33]. As far as inorganic chemicals are concerned, Turner and Filella [34] underpinned that 1) hazardous metals occur widely in plastics in societal circulation and lost in nature, and 2) metal diffusion from specific plastics can be an important health and environmental concern ([34] p. 20). Especially legacy heavy metal additives can pose an ecotoxicological risk.

Detailed chemical characterizations of the complex plastic leachates including target analysis and non-targeted characterization of organic compounds, metal(loid)s and plastic particles are urgently needed for a sound assessment of the environmental implications of plastic debris as well as future decision making. Such comprehensive assessments require a toolbox of different analytical instruments, the respective know-how and often the formation of research consortia covering scientists from different analytical disciplines, as in the P-LEACH project (www.ufz.de/p-leach). The complementary analytical toolbox of P-LEACH covers Attenuated Total Reflectance Fourier Transform Infrared (ATR-FTIR) spectroscopy (qualitative indication of oxidation) and Laser Direct Infrared (LDIR) imaging (microplastic number concentration, size and shape), liquid chromatography coupled to (high resolution) mass spectrometry (LC-(HR)MS (target analysis and non-targeted chemical characterization of organic compounds)) and Inductively Coupled Plasma (ICP)-MS/MS (metal(loid) fingerprinting).

The objectives of this screening study were to 1) investigate the influence of UV-radiation on the leaching behavior of chemicals and particles from selected model consumer plastics into water compared to dark controls under well-defined laboratory conditions and 2) extensively characterize the released particles, organic analytes, and metal(loids). This comprehensive array of approaches yields a unique, comprehensive picture of the chemicals and particles liberated into water from commercial products under simulated environmental weathering conditions. Overall, our pilot study provides a first sound data basis for future assessment of the environmental impact of chemicals leaching from plastics and decision making.

## 2. Materials and methods

### 2.1. Selection of plastic consumer products

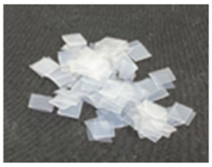
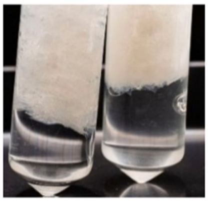
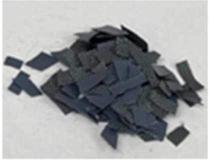
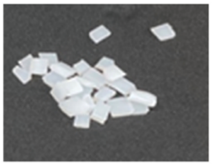
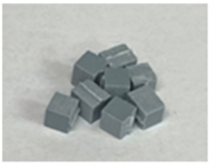
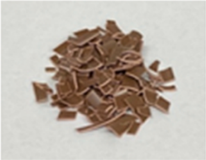
Different plastic materials were selected to cover a broad range of applications and chemical properties. The materials included the most used polyolefin (Polyethylene (PE), 105.6 million tons produced globally in 2021), polyester (polyethylene terephthalate (PET), 24.2 million tons), and halogenated polymer (polyvinylchloride (PVC), 50.8 million tons) [35]. These polymers also covered different densities with associated floating (PE) and sinking (PET, PVC) properties in water. Importantly, consumer products that promoted specific properties such as "flame retardant" or "UV-stable" (provided by the distributors) were selected whenever possible, as these appeared likely to contain functional chemicals from important additive groups.

Four PE, two PET and two PVC consumer products were selected (Table 1 and Text S1 in supporting information A (SI_A)).

### 2.2. Experimental setup

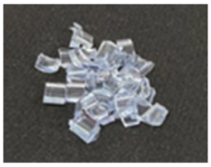
The plastic materials were cut into pieces of < 5 mm length using scissors, scalpels, or water jet cutting, depending on the mechanical properties of the product. For each sample, 30 g of cut and cleaned material were added to 200 mL of double-distilled water in boron silicate cuvettes (for exact weights see Table_S1 in SI_A). Two samples were prepared from each material to compare the UV-exposure treatment (UV) with a dark control (DC; covered in aluminum foil and submitted to the same movement and temperature regime as the UV-treated material).

**Table 1**
Information on selected consumer products, including sample ID, short description, information whether the polymer type could be confirmed using ATR-FTIR, its classification including hit quality index (HQI) value, the additive groups expected to be contained based on manufacturer information, a photo of the cut product, and a visual comparison of dark control (DC) (left) with ultraviolet (UV) treated samples (right) after the UV weathering experiment (high-resolution images in Fig. S2 in SI_A).

| Sample ID | Short description | Polymer type confirmed? | Classification by IR-spectroscopy (HQI value) | Expected additive groups | The cut product | Visual comparison after the experiment |
|---|---|---|---|---|---|---|
| PE_1 | greenhouse cover foil | Yes | PE (906/1000) | UV stabilizers |  |  |
| PE_2 | Sun protection cover | Two materials | PE (899/1000)<br>PET (788/1000) | UV stabilizers |  |  |
| PE_3 | jerrycan | Yes | PE (892/1000) | UV stabilizers |  |  |
| PE_4 | agricultural foil | Yes | PE (914/1000) | UV stabilizers |  |  |
| PET_1 | D-Sub plug housing | No | Styrene Acrylonitrile (SAN) copolymer (745/1000) | flame retardants |  |  |
| PET_2 | Multipurpose housing | No | Unidentifiable | flame retardants |  |  |
| PVC_1 | cable housing | No | Unidentifiable | flame retardants |  |  |

(continued on next page)

Table 1 (*continued*)

| Sample ID | Short description | Polymer type confirmed? | Classification by IR-spectroscopy (HQI value) | Expected additive groups | The cut product | Visual comparison after the experiment |
|---|---|---|---|---|---|---|
| PVC_2 | transparent PVC-tubing | (Yes) | Vinyl chloride vinyl acetate hydroxypropyl terpolymer (581/1000) | no phthalates according to supplier; non-phthalate plasticizers expected |  |  |

A weathering chamber, as described in detail by Gewert et al. [36] (G [36]), was used for artificial UV-exposure in a custom-made UV-chamber (Figure_S1 in SI_A). Samples were exposed for 10 days, corresponding to natural light exposure of roughly eight months in Central European climate (Text S2 in SI_A). Batches of up to three materials were exposed in parallel with one blank control containing double-distilled water only. During weathering, samples were continuously rotated around a strong UV-light source (HTC 400–241 SUPRATEC HTC/HTT, OSRAM GmbH) to ensure even exposure of all samples irrespective of their densities. The UV source had a radiation flux maximum mainly in the UVA range of 490 W (~350 to 390 nm), which could pass through the 2 mm thick boron silicate glass with more than 70 % transmission efficiency (https://www.pgo-online.com/de/borofloat.html). After exposure, samples were allowed to cool down to room temperature for 30 min from peak temperatures between 30 °C and 50 °C (typical for DC and UV samples, respectively). Different subsamples were then collected for distribution among project partners to make use of the broad analytical toolbox of P-LEACH (Fig. 1). For microplastic analysis, 15 mL of unfiltered leachate were transferred to a glass vial after re-suspension of the plastic particles in the leachate by careful shaking. For metal(loid) analysis, another 50 mL of the unfiltered leachate were poured into acid-leached DigiTUBEs (SCP Science, Quebec, Canada). For (non-)targeted analysis, the remaining leachate was filtered through a 20 µm stainless steel sieve (Retsch, Haan, Germany) and filled into solvent rinsed and baked-out Duran bottles. Residual plastic pieces were collected in DigiTUBEs.

### 2.3. *Particle characterization using LDIR spectroscopy*

100 - 200 µL of the unfiltered leachates (4 samples per slide) were pipetted onto MirrIR (low-e microscope) slides (Kevley Technologies, Chesterland, USA). After evaporation of the aqueous medium, the residues were analyzed using the Agilent 8700 LDIR Chemical Imaging system (Agilent Technologies) in transflection mode. Corresponding applications of LDIR imaging to detect microplastics in marine filter samples have been described in previous publications [37,38]. The Agilent Clarity software (version 1.1.2) and the corresponding database were used to run the analysis. More details are provided in Text S3 in SI_A.

### 2.4. *Characterization of the chemical composition using LC-HRMS*

**Sample preparation**: Leachates were filtered through 0.2 µm PTFE syringe filters (Whatman, Th.Geyer). To 900 µL of leachate 100 µL of methanol and 40 µL of internal standard mix ($c = 1$ µg/mL, $n = 47$, compounds are listed in Table S1 in SI_B) were added. Samples were stored frozen at −20 °C until analysis via direct injection.

**Analytical method:** A Thermo Fisher VanquishTM HPLC system coupled to an Orbitrap Exploris 480 mass spectrometer equipped with an electrospray ionization (ESI) source was used for the non-target screening (instrumental details in Text S4 in SI_A).

**Data evaluation**: Peak-picking was performed in mzMine 3.4 using batch mode (settings are given in Table S2 in SI_A) [39]. The main procedure steps were (i) mass detection, (ii) Automated Data Analysis Pipeline (ADAP) Chromatogram Builder, (iii) smoothing, (iv) chromatogram deconvolution, and (v) feature alignment. Subsequently, stable isotope-labeled standards were matched with an in-house database as quality control. Aligned features with chromatographic peak heights > 1.0E5 were exported and used for characterization of the chemical composition of the leachate samples.

### 2.5. *Target analysis of organic leaching chemicals using LC-MS/MS*

**Sample preparation:** Filtered leachate samples were extracted and concentrated 20-fold by solid-phase extraction (SPE) for targeted analysis using LC-MS/MS following an SPE protocol adapted from Neale et al. [40] (details are given in Text S5 in SI_A).

**Instrumental analysis:** Samples were analyzed using an in-house method for plastic-associated chemicals ($n = 71$) covering plasticizers ($n = 13$), UV stabilizers ($n = 21$), antioxidants ($n = 11$), flame retardants ($n = 17$), precursor monomers ($n = 8$) and others ($n = 1$) (details in Table S2 in SI_B). This method has not been published previously. Instrumental analysis was performed using an UHPLC-MS/MS system (Agilent 1290 Infinity coupled to an Agilent 6490 triple quadrupole LC/MS, Agilent Technologies, Santa Clara, USA) with an ESI source. For the chromatographic separation a Zorbax Eclipse Plus C18 column (95 Å, 2.1 mm × 150 mm, 1.8 µm, Agilent Technologies, Santa Clara, USA) (for details see Text S6 in SI_A) was used. Multiple reaction monitoring (MRM) with mass spectrometric parameters optimized for each analyte was employed (details in Table S2 in SI_B). For quantification, a 14-point external calibration curve (0 – 250 ng mL$^{-1}$) was used.

### 2.6. *Metal(loid) fingerprinting by ICP-MS/MS*

**Sample preparation leachates:** Unfiltered leachates from UV-treated and DC plastic consumer products were subjected to vacuum-filtration (0.45 µm polytetrafluoroethylene (PTFE) filters; SCP Science, Quebec, Canada) using a manifold to remove any remaining (undigested) particles and prevent blockage of the nebulizer [41].

**Sample preparation plastics:** Plastic pieces were freeze-dried (Gamma 1–16 LSC plus, Christ, Osterode, Germany). Afterwards, three replicates of 100 mg ± 5 mg (1 *SD*; $n = 3$)) per plastic consumer product were weighed into precleaned 35 mL quartz pressure vessels (CEM GmbH, Kamp-Lintfort, Germany) for digestion, followed by vacuum-filtration (for details see Text S7 in SI_A and Hildebrandt et al. [42]).

**Data evaluation**: Multi-elemental analysis covering 54 elements in four different modes (no gas, He, $N_2O$ and $H_2$) was performed using an ESI SC-4 DX FAST autosampler (Elemental Scientific, Omaha, USA) coupled to an ICP-MS/MS instrument (Agilent 8800, Agilent Technologies, Tokyo, Japan) (for details see Text S8 in SI_A).

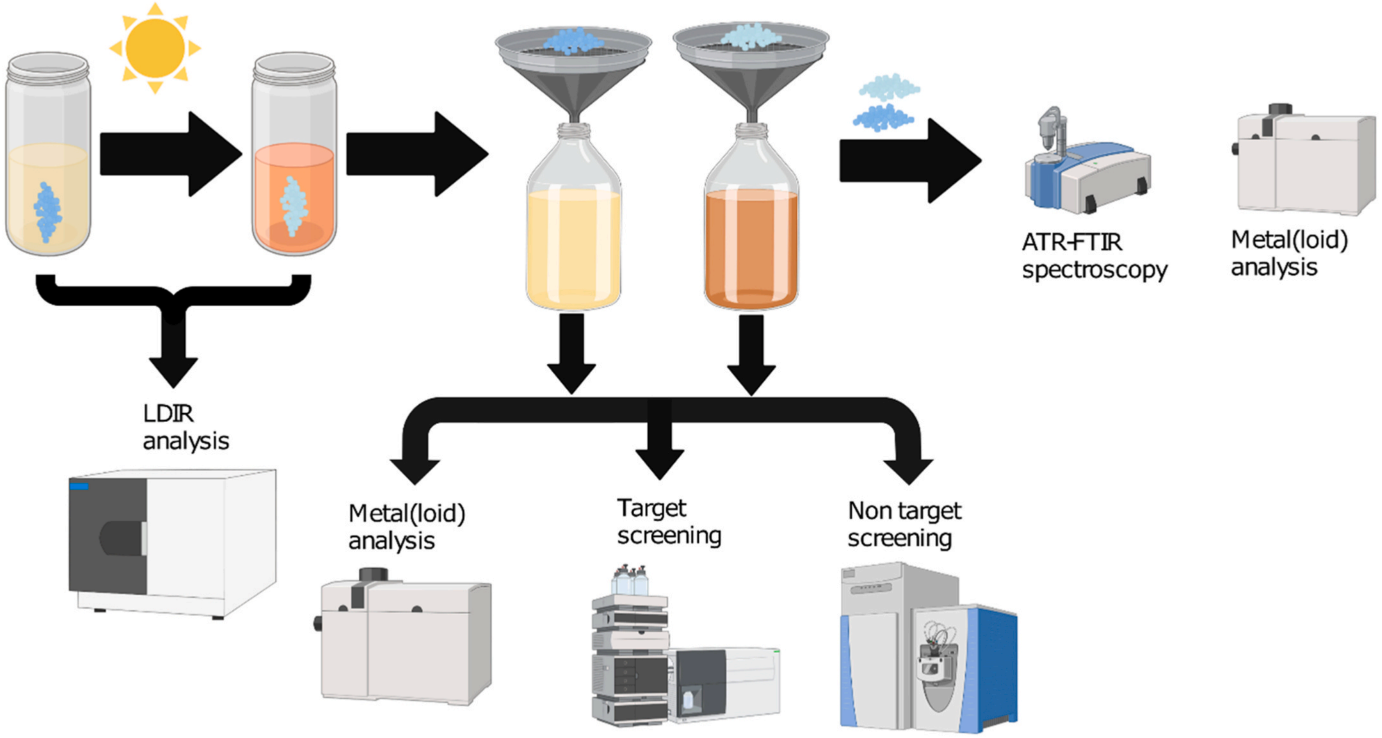


**Fig. 1.** Schematic of the experimental setup of UV-treatment and the chemical-analytical characterization pipeline. Laser Direct Infrared (LDIR) analysis: Analysis of microplastics. Figure created with BioRender.

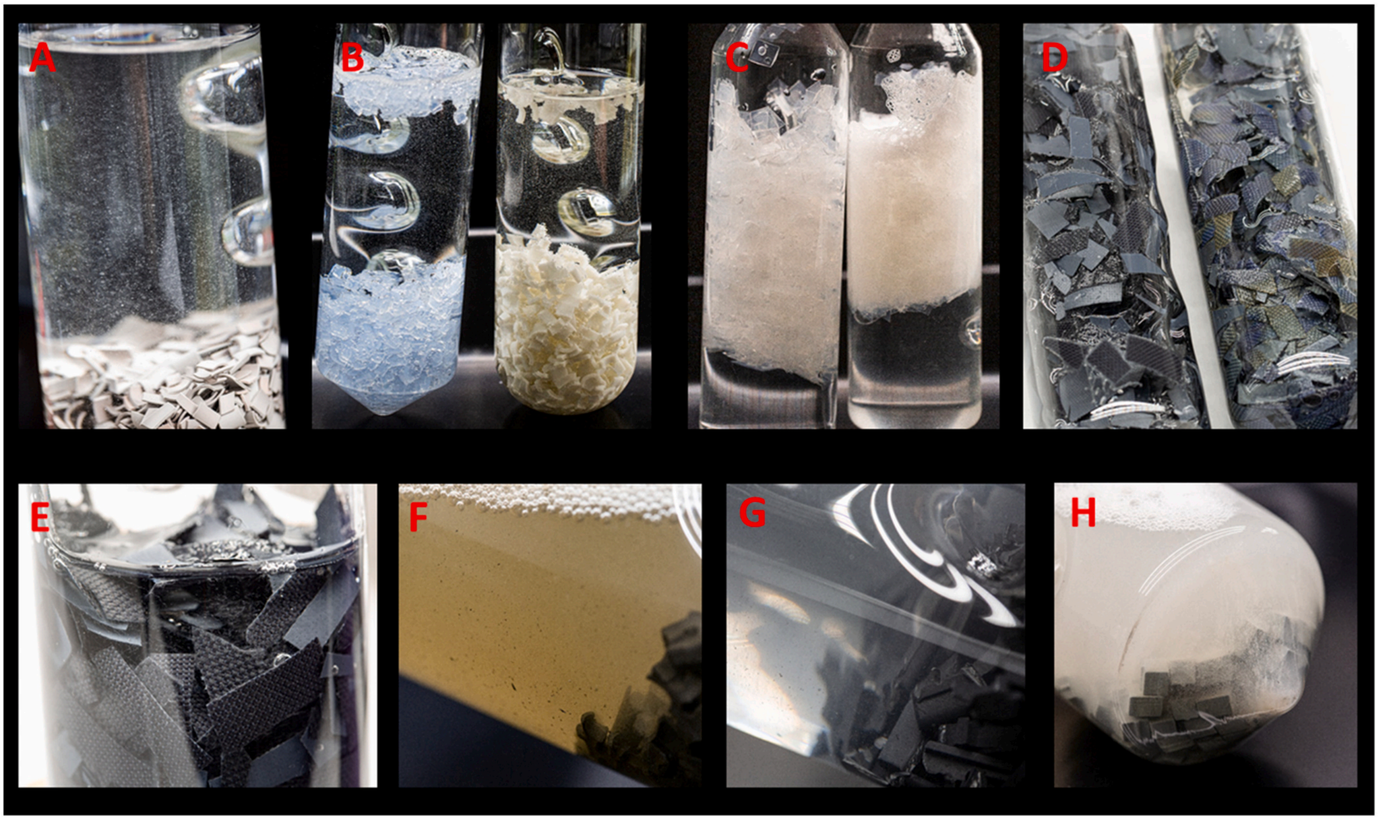


**Fig. 2.** Effects of UV treatment on different consumer plastics. A: Particle formation from PVC_1. B: Bleaching of PVC_2 (left = dark control (DC), right = UV treatment). C: Foam formation for PE_1. D: Yellowing of PE_2. E: Microfibre formation from PE_2. F: Microplastic formation and yellowing of leachate of PET_1 (UV). G: Microplastic formation from PET_1 (DC). H: Turbidity of PET_2 leachate.

## 3. Results and discussion

### 3.1. *Visual changes*

A range of visual changes could be observed after the experiment (Table 1, and Fig. S2 in SI_A). All observed changes were summarized in Table S4 SI_A, and pictures of the following examples are provided in Fig. 2: Formation of particles from PVC_1, PE_2 and PET_1 (panels A, E and F and G, respectively) (see also 3.3 Microplastic analysis), bleaching of PVC_2 (panel B), foam formation for PE_1 (panel C), yellowing of PE_2 and PET_1 (panels D and F, respectively), and turbidity of the PET_2 leachate (panel H).

The overall strongest visual changes induced by the UV treatment were observed for the PET and PVC materials. In the case of PET, yellowing of the leachates as well as particle formation and decolouring of the plastic pieces were noticeable.

Yellowing of polymers may be associated with formation of chromophore systems, which increase polymer degradation [43]. For PVC materials, strong bleaching was observed, while the leachates remained clear.

Interestingly, substantial foam formation was observed for three materials, namely PE_1, PET_1, and PET_2. Here, long-chain molecules with surfactant functionalities might have been formed or released. Another possible explanation could have been the presence of a coating originating from the production, e.g., in the case of PE_1 (greenhouse foil), with anti-sticking properties to facilitate the separation of the thin layers.

### 3.2. *Microplastic analysis*

Microplastic release from larger plastic items is known to be induced by UV radiation [44,45], and we therefore examined the generation of particles and fibers under accelerated UV weathering conditions for all consumer plastics using LDIR (SI_C).

Interferences arose from the precipitation of leached chemicals in the surrounding of the particles (Figure in SI_C), which occurred because aliquots of the leachates had to be pipetted directly onto the Ag-coated slides to allow LDIR analysis, as stated in the methods section.

In the cases of PVC_1 and PET_2, the IR spectra showed strong agreement with database spectra of stearates and rubber (both indicators of long chain aliphatic compounds), and chlorinated PE. The water blanks contained no microplastics. The results for PE_2 and PET_1 are shown in Fig. 3 and discussed below as examples. The detailed findings for the other consumer plastics can be found in SI_C.

The DC leachate of PE_2 contained 400 PET fibers mL$^{-1}$ (20 fibers (g plastic)$^{-1}$), whereas 3280 PET fibers mL$^{-1}$ (163 fibers (g plastic)$^{-1}$) were found in the corresponding UV leachate ($>$ 8-times higher number concentration). Interestingly, neither for the UV treatment nor the DC leachate of PE_2 could a release of any particles attributable to the material's PE side be observed. In the DC leachate of PET_1, 2460 SAN particles mL$^{-1}$ ($\sim$ 81 particles (g plastic)$^{-1}$) were detected. The UV leachate did not contain a considerably higher SAN particle number concentration (2810 particles mL$^{-1}$, $\sim$ 91 particles (g plastic)$^{-1}$).

These observations indicate that the rotation of the cuvettes and collisions between the plastic pieces led to surface erosion of materials that are prone to mechanical abrasion, probably facilitated by the large number of plastic particles immersed in a comparably small volume of water. PET_1 was particularly brittle.

The size distributions of the SAN particles followed an exponential distribution ($R^2 > 0.997$) (Fig. S3 in SI_A). These findings match laboratory studies of photodegradation [46] and marine and riverine monitoring studies [47-49].

Interestingly, the size distribution of the PET fibers in the DC leachate of PE_2 (SI_C) was comparable to the ones reported in studies that focused on fiber release by laundry [50-52] (tendency towards normal distribution), whereas the size distribution of PET fibers in the corresponding UV leachate exhibited a linear distribution ($R^2 > 0.994$) (Fig. S2 in SI_A). As most environmental studies focusing on microplastics of $<$ 300 μm report combined size distributions for microplastic particles and fibers, comparison of the results with literature data is not possible.

The results of the current study showed that both, mechanical stress and UV exposure (and possibly elevated temperatures) enhanced fragmentation and increased the formation of microplastics/-fibers from larger plastic pieces, corroborating previous findings [53].

### 3.3. *Characterization of the chemical composition using LC-HRMS*

The effects of UV treatment on the leaching of organic substances were characterized through investigation of the HRMS data. This data

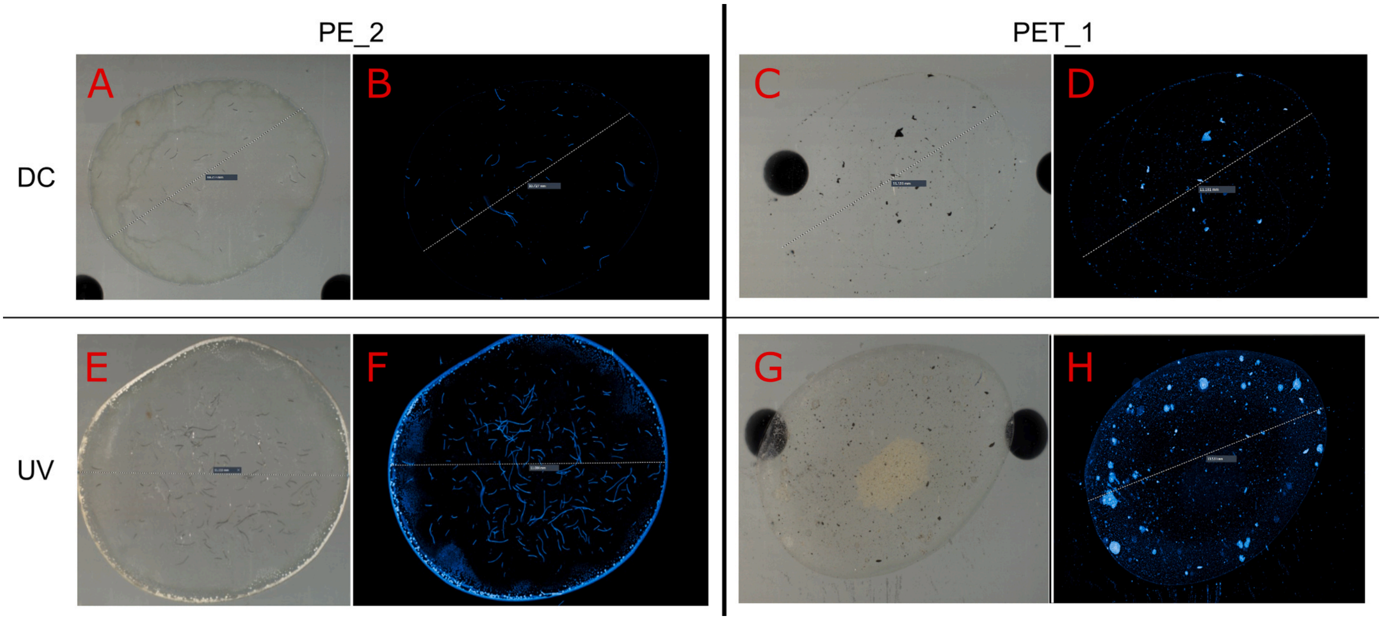


**Fig. 3.** Vis and IR images of the LDIR analysis for PE_2 (left) and PET_1 (right) of the DC (A and B, and C and D, respectively) and UV-weathered (E and F, and G and H, respectively). Circles exhibit diameters of approximately 11 mm.

contained information on the chemical composition of the leachates including functional additives as well as other plastic-related chemicals like non-intentionally added substances, monomers, as well as chain scission- and transformation products.

Only features unique to a sample or with peak heights > 10-fold the respective blanks were considered. The number of features in each DC leachate was lower compared to the respective UV leachate for all consumer plastics except for two products advertised as “UV stable” (PE_3 (jerrycan) and PE_4 (agricultural foil)). A median of 239 (min. 164 to max. 337) features were detected across all DC samples, and a median of 343 (min. 208 to max. 559) features across the UV leachates (details in Fig. S4 in SI_A). This overall tendency for increased numbers of features in the UV-treated samples can likely be attributed to 1) increased leaching rates due to the UV irradiation [43] and 2) formation of intermediates and chain scission products [54]. The results of the individual consumer products are discussed in Text S9 in SI_A.

Most features were unique to either the DC or the UV sample of a given consumer product (Fig. 4, panels A and B, and Figs. S5-S12 in SI_A). This observation further indicated the formation of intermediates or chain scission products [36,54]. Features that were detected in both DC and the corresponding UV samples generally had peak heights within a factor of 10 of each other, which indicates a limited difference in leaching behavior for these compounds between UV and DC samples. In few cases, clearly elevated peak heights (>10-fold difference) were observed in the UV samples (e.g., PET_2, PVC_1, PE_2; see Fig. S13 in SI_A), suggesting UV-related enhancement of leaching rates for some compounds, as has been described, e.g., for certain phthalates [55,56].

Chemical profiles of different plastic products are known to be complex and mostly product-specific [57], and the current results showed that UV-treatment further increased this complexity.

The mean rt of all unique features (i.e., features occurring in either DC or UV, but not both) was in all cases lower in UV than in DC and differences ranged from 0.64 min to 4.67 min (Fig. 4, panels C and D, and Figs. S14-S21 in SI_A).

This differene indicates a shift towards more hydrophilic compounds and suggests the formation of transformation products that are

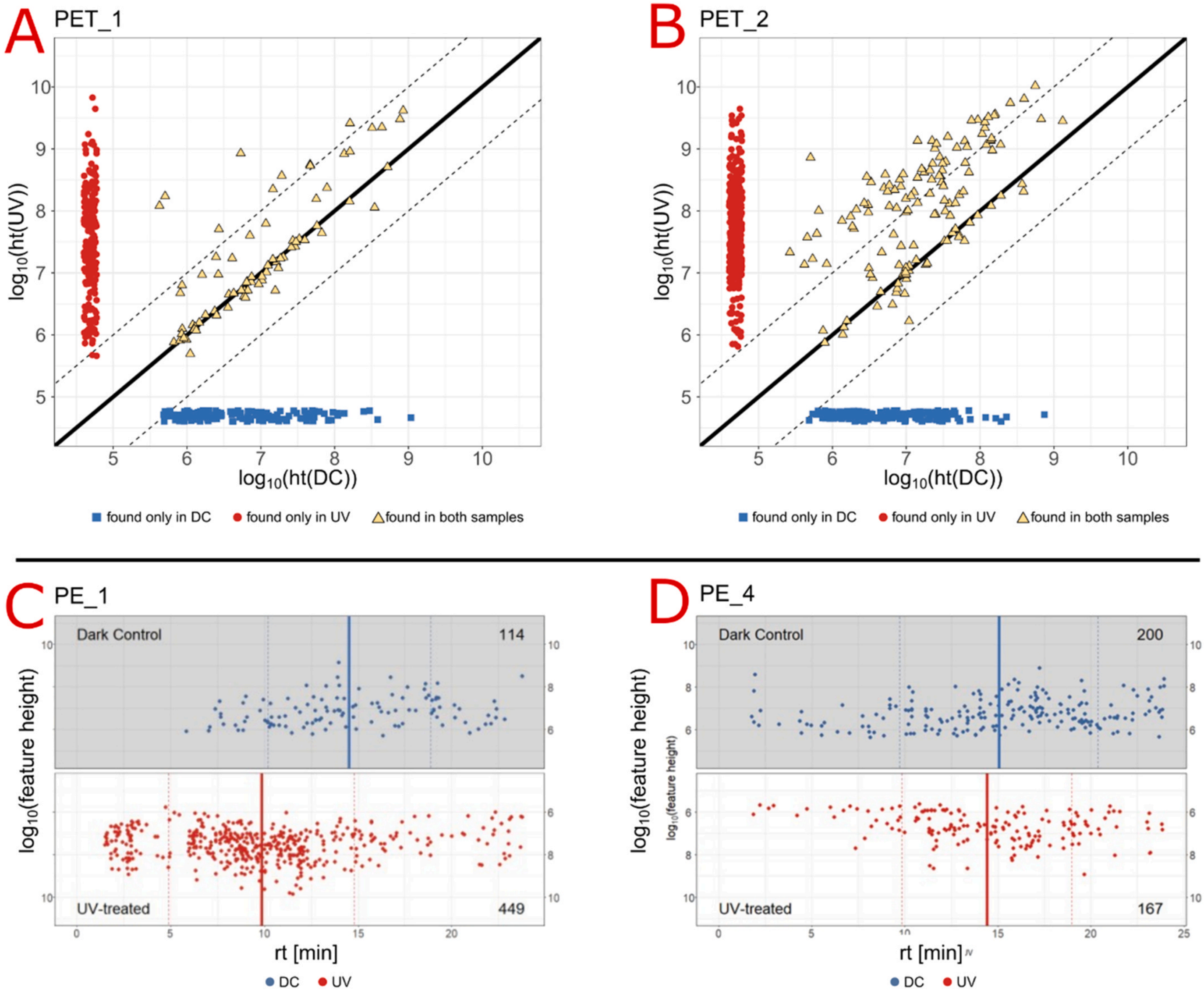


**Fig. 4.** TOP - Plot of feature height in dark control (DC) against UV-treated samples for PET-1 (A) and PET-2 (B). Most features were only present in one of the sample types (DC-only blue, UV-only red). If features were present in both samples, feature height was mostly not affected by the irradiation, as exemplified for PET_1 (A). For PET_2, on the other hand, the irradiation increased the leaching of some compounds, which can be seen from the points above the dashed 10:1 line (B). BOTTOM - Peak heights of unique features in DC (grey background) and UV-treated samples (white background) plotted against retention time (rt) for the samples with the largest (PE_1, panel C) and smallest (PE_4, panel D) observed differences in rt. The bold vertical lines mark the mean rt. Dashed lines mark the standard deviation of the mean rt. Only features with a rt > 1.5 min and height > $10^6$ were considered.

potentially more bioavailable than their parent compounds [58]. Increased bioavailability raises concerns for environmental and health risk assessments.

### 3.4. *Target analysis of organic leaching chemicals using LC-MS/MS*

The data from the spiked mass-labelled standards (Fig. S22 in SI_A) indicated matrix-related challenges after SPE in the accurate quantification of the targeted analytes (discussed in detail in Text S10 in SI_A). These challenges did not affect the LC-HRMS measurements, which were analyzed via direct-injection. Consequently, we decided to calculate only absolute concentrations without internal standard correction. Since the analyte loss and/or the matrix effects induced by SPE were not corrected for, the values reported here are probably underestimated. Substantial differences induced by UV treatment, defined as those exceeding one order of magnitude, were specifically highlighted to underscore substantial variations that overshadowed common uncertainties typically adjusted for by internal standards.

Fifteen of the 71 target analytes were detected (13 of which were quantifiable) in at least one sample. These covered four groups of leaching additives including plasticizers (diisononylcyclohexane-1,2-dicarboxylate (DINCH), diisononyl phthalate (DINP)), antioxidants (Cyanox 2246, Irgafos 126, Irganox 1010), flame retardants (diphenylphosphate (DPhP), bis(2-ethylhexyl)phosphate (HDEHP), tertrabromobisphenol-A (TBBPA), tri(butoxyethyl)phosphate (TBEP), tris-(2-chloroethyl) phosphate (TCEP), trimethyl phosphate (TMP), triphenyl phosphate (TPhP)) and UV stabilizers (Tinuvin 327, Tinuvin 329 and Tinuvin P) (Fig. 5 and Table S3 in SI_B). The concentrations spanned an extensive range from below the lowest calibration point (0.01 ng mL$^{-1}$) to levels far exceeding the highest one (250 ng mL$^{-1}$). This wide span showed the analytical challenges that can be expected in such leachates and indicated vastly different amounts of additives used in the investigated consumer products combined with differences in the leaching susceptibility across chemical compounds. Additive amounts in the bulk plastic can vary substantially depending on the polymer type, the additive, and the intended function of the product, as described in the ECHA Mapping exercise and other literature [59,60]. Additionally, polymer type, size, shape and other factors (e.g. leaching medium) can influence leaching behavior [61,62,55,56].

Differences in composition and concentration were observed for most investigated consumer product leachates depending on whether UV radiation was used or not. The results of the individual consumer products are discussed in Text S11 in SI_A.

UV radiation, possibly combined with mechanical stress and/or the associated temperature increase resulted in higher concentrations for some UV samples compared with the DC samples. For example, the leaching of DPhP from PET_1 was strongly enhanced by UV radiation. In some cases, compounds were only detected after UV exposure but not in the respective DC sample, e.g. Tinuvin 329, Tinuvin P and TMP. In contrast, some chemicals were only observed in the DC leachates but not in the UV samples. This pattern might indicate transformation of photolabile compounds in the UV-treated leachates to products not detected with the applied target method. Examples are all three detected antioxidants Cyanox 2246 (PE_1), Irganox 1010 (PE_2) and Irgafos 126

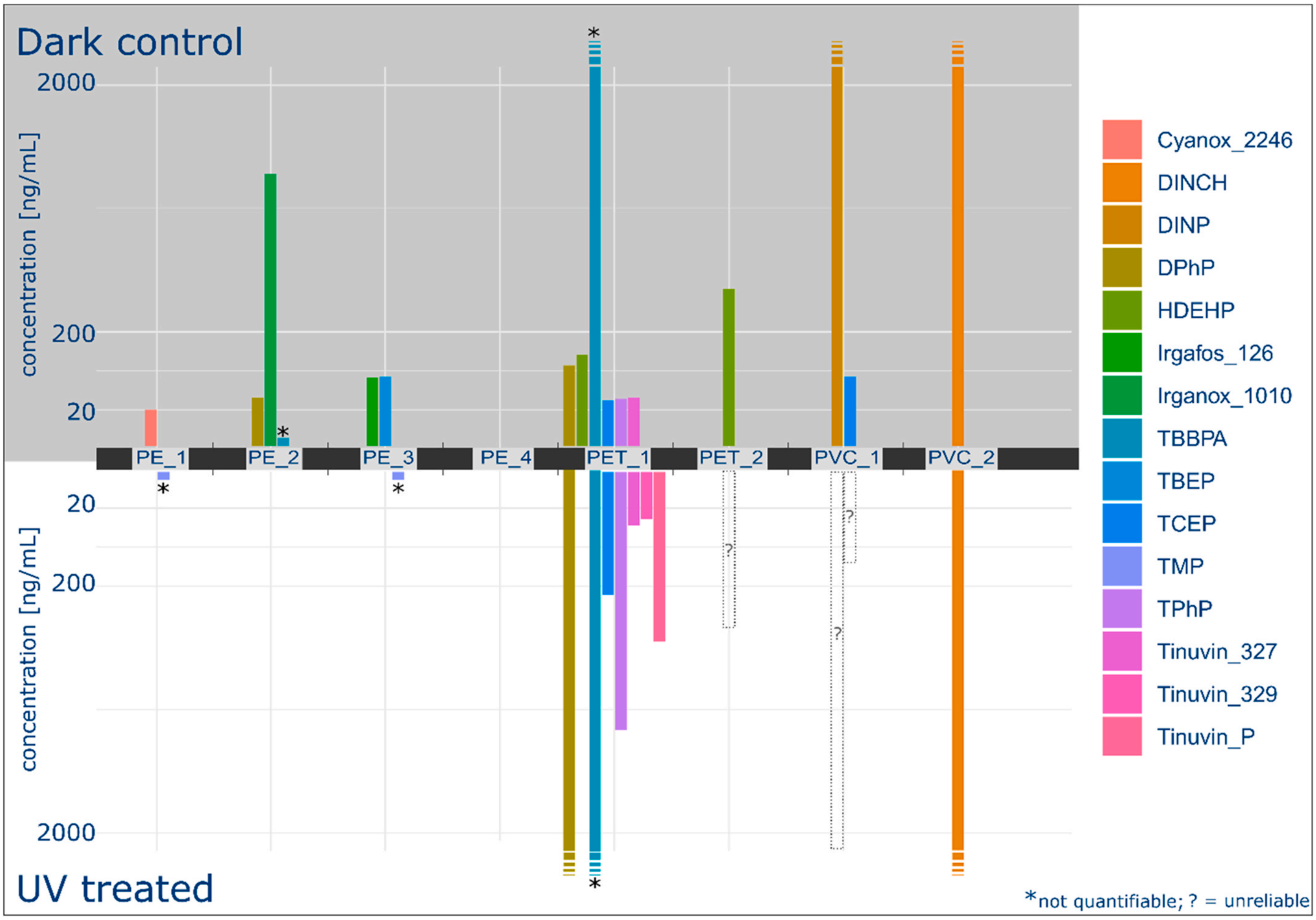


**Fig. 5.** Results of the target analysis comparing leaching in the DC (top half with grey background) with UV exposure (bottom half) samples. Concentrations were calculated without internal standard correction and are likely underestimated. Details are given in the text and in Tables S2 and S3 in SI_B.

(PE_3), and the flame retardant HDEHP (PET_1).

One interesting case was the leaching behavior observed for DPhP. Leaching of DPhP from PET_1 (a styrene acrylonitrile copolymer, according to our ATR-FTIR results) was strongly enhanced by UV radiation, whereas it was detected in the DC but not the UV leachate from PE_2 (containing one PE and one PET side). This pattern could indicate a dependency of the leaching behavior of DPhP on the polymer structure or product-specific properties. Similar observations have been reported for certain phthalates [63].

### 3.5. Metal(loid) fingerprinting by ICP-MS/MS

According to the recoveries for metals and metalloids in the chosen certified reference material (ERM-EC680m, low-density PE, LDPE), the sample preparation workflow was suitable for the quantification of metal(loid)s in plastics namely arsenic (As): 90 %; cadmium (Cd): 100 %; chromium (Cr): 96 %; mercury (Hg): 91 %; lead (Pb): 104 %; antimony (Sb): 102 %; tin (Sn): 93 %; and zinc (Zn): 96 %. In total, mass fractions (bulk plastics) and concentrations (leachates) were analyzed for 54 elements. The entire dataset is given in Table S4 in SI_B.

#### 3.5.1. Metal(loid)s in bulk plastics

For reasons of elevated relevance regarding toxic plastic additives [10,42,34] and available information on potential (eco)toxicity [64], only results for selected elements are discussed in depth within this study namely As, barium (Ba), Cd, Cr, copper (Cu), Hg, nickel (Ni), Sb, Sn, titanium (Ti), uranium (U), vanadium (V) and Zn.

The four analyzed PE materials and PVC_2 exhibited variable but low elemental mass fractions. More information on elemental mass fractions of each plastic material can be found in Table S4 in SI_B. High mass fractions of Cu were found in PE_2, PE_4, PET_1 and PET_2 (up to 730 ± 160 mg $kg^{-1}$ for PE_4 (U ($k = 2$))).

Furthermore, a Sb mass fraction of 17,000 ± 3000 mg $kg^{-1}$ (1.7 % (*w*/*w*)) accompanied by a Sn mass fraction of 270 ± 60 mg $kg^{-1}$ was found for PET_1. The Sb mass fraction of PE_2 was 59 ± 3 mg $kg^{-1}$.

Ni-containing molecules are frequently used as catalysts in PE polymerization [65]. Zinc oxide is used both as a pigment and as a stabilizer in various plastic products made from different polymers, providing increased heat resistance and tensile strength [66]. As-containing compounds are typically used as antimicrobial agents but not for food contact products [65] which fits well with their absence in PE_3 and PVC_2. Sb is also frequently used as a catalyst in PET polymerization and beyond this mainly added to plastic products as a flame-retardant synergist (in combination with halogenated compounds) [67,68,10]. Since PE_2 is used as sun protection cover (for outdoor furniture, which includes fire risk from e.g., smoking and barbecueing) and PET_1 serves as a D-Sub plug housing (cable fire), flame retardant properties seem reasonable.

The toxicity of antimony salts for living organisms including humans is well documented (summarized in [69] and [70]). In various aquatic organisms, antimony salts can cause embryonic malformation, DNA damage, neurotoxicity, oxidative stress, and carcinogenicity [69,71]. Thus, Sb is categorized as a priority contaminant by the United States Environmental Protection Agency (US EPA) and the Council of the European Union (EU) [72]. Despite known toxicity and suspected carcinogenic potential for humans (International Agency for Research on Cancer (IARC): Group 2B), the EU Restriction of Hazardous Substances in Electrical and Electronic Equipment Directive (RoHS) does not provide a limit for Sb or antimony trioxide [60].

It has been demonstrated that the leaching of Sb from plastics increases at high temperatures [73]. As our leaching conditions included elevated temperatures, these could explain substantial release of Sb in our study.

Among the other elements covered by the EU RoHS directive (i.e., Cd (0.01 %), hexavalent Cr (0.1 %), Hg (0.1 %) and Pb (0.1 %)), the mass fraction of Cd (PET_1: 0.0004 %) was closest to (but 25-times lower than) the RoHS limit value (0.01 %).

Plastics used with electric or electronic equipment are known to contain high mass fractions of Sb and different other heavy metals [74-76]. Overall, the bulk material analysis revealed low levels of potentially harmful inorganic elements. For example, PET_2, which is a multifunctional housing (also for electrical applications), showed low elemental mass fractions of the selected metal(oid)s (except for the less problematic Cu, Ti and Zn). In contrast, PVC_2 (transparent PVC-tubing suitable for application in food processing), was in good agreement with the specifications of the Food and Drug Administration (FDA) as well as the European Union (EU), as no considerable mass fractions were observed here.

#### 3.5.2. Metal(loid)s in leachates

Considerable amounts of the total elemental mass fractions leached out from the bulk material over time, with a higher proportion leached from UV-treated samples compared to DC samples. Particularly high extents of leaching were observed for Ni, Sb, Zn and As (Fig. 6). The corresponding figures and leached concentration data for the remaining 9 elements are given in Figs. S23 - S31 in SI_A and Table S4 in SI_B, respectively.

Element concentrations in the leachates co-varied with the corresponding mass fractions in the bulk material. Leachates from PE_1 (greenhouse cover foil), PE_3 (jerrycan), PE_4 (agricultural foil), PET_2 (multipurpose housing) and PVC_2 (transparent PVC-tubing) contained low metal(loid) concentrations – in accordance with the low mass fraction in the bulk material.

To evaluate if the levels might be problematic, the concentrations of the metal(loids) in the leachates were compared to the German limit values for drinking water (TrinkwV) [77]. The leachates from PET_1 (D-Sub plug housing) had the highest potential toxicity.

The limit values [77] were exceeded 15.6-times for As (limit value = 10 μg $L^{-1}$), 1.8-times for Cd (3 μg $L^{-1}$), 4.4-times for Ni (20 μg $L^{-1}$), 2.1-times for Pb (10 μg $L^{-1}$), and 2440-times for Sb (5 μg $L^{-1}$) by the measured concentrations of the UV leachates. The Sn concentrations were elevated in both UV and DC leachates. It must be kept in mind that the approach chosen for this study included highly elevated plastic loads in small water volumes under strong leaching conditions. Correspondingly, Chapter 3.7 expands on the relevance of the experimental scenario for drinking water.

In accordance with the high Sb mass fraction in the plastic samples (17 ± 3 g $kg^{-1}$ (($U$ ($k = 2$))), the concentration of Sb in the leachates of 24 ± 4 mg $L^{-1}$ was remarkably high. According to acute toxicity data sourced from temperate and tropical regional studies, the predicted no effect concentrations (PNECs) are 0.156 μg $L^{-1}$ and 0.195 μg $L^{-1}$ of Sb (III) [78]. In the tiered ecological risk assessment approach, the alga *Selenastrum capricornutum* was the most sensitive species ($EC_{50}$ = 0.75 mg $L^{-1}$) whereas the alga *Pseudokirchneriella subcapitata* was the most tolerant ($EC_{50}$ = 206 mg $L^{-1}$). Hence, the measured values in our extreme setting clearly exceeded the PNEC values for different freshwater species by more than two orders of magnitude [78]. The leachates from PE_2 and PVC_1 also exhibited elevated Sb concentrations. While the DC leachates of both materials did not exceed the limit value of the EU drinking water directive substantially, the UV treatment led to Sb concentrations > 600 μg $L^{-1}$ in the PE_2 leachate (over 60-times the limit value) and > 30 μg $L^{-1}$ in the PVC_1 leachate (over 3-times).

To conclude, metal(loid) release from the different consumer plastics varied strongly, within a range from almost 0 % (e.g. 0.0009 % Cu from PE_2) to 50 % to 100 % (e.g. Ni from PET 1, Zn from PET (both Fig. 6) and Ba from PVC_1 (Fig. S23 in SI_A)). Nevertheless, small shares can still result in relevant concentrations of potentially harmful element species. The current results indicated substantial leaching of potentially toxic metal(loid)s over relatively short periods, even in the absence of UV light.

This observation is consistent with the work of Novotna et al. [79], who showed rapid leaching of 10 elements from 16 types of

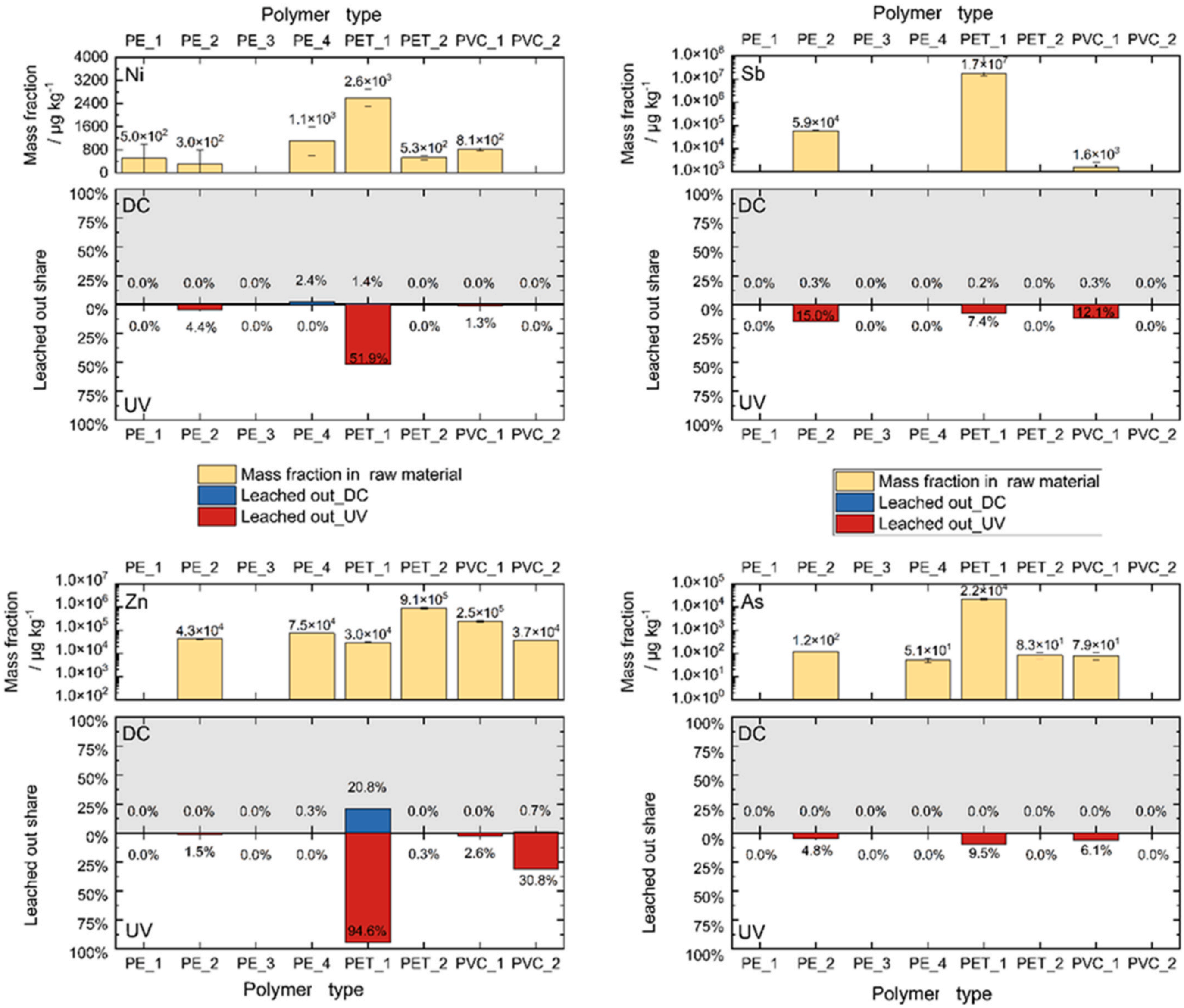


**Fig. 6.** Measured elemental mass fractions of Ni, Sb, Zn and As of all consumer plastics together with the relative leached share of the total elemental mass in DCs (grey area) and UV treatments.

microplastics prepared from consumer plastics into ultrapure water during the first two to three weeks followed by equilibrium partitioning between the plastics and the aqueous phase [79].

### 3.6. A brief toxicity profiling based on US EPA databases

In the following section, the potential toxicity of the leachates produced in the extreme setting of this study is briefly explored from a human health perspective using data readily available through the US EPA ToxVal database [80,81] and the ToxCast assay database [81,82]. In assessing the toxicological profiles of metal(loid)s from our leaching experiments, we mined the Maximum Contaminant Levels (MCL) from ToxVal as a benchmark for hazard identification (Fig. S32). Mercury, with an MCL of 2 µg $L^{-1}$, was found to be the most hazardous, closely followed by As, Cd, and Sb, which each possess an MCL at or below 10 µg $L^{-1}$. These elements are associated with substantial health risks even at low exposure levels. Uranium, with an MCL of 27.5 µg $L^{-1}$, alongside Cr and Ni, each with an MCL of 100 µg $L^{-1}$, also displayed considerable toxicity and warrant careful consideration. In contrast, Ba and Cu, with MCLs of 2000 µg $L^{-1}$ and 1300 µg $L^{-1}$, respectively, present a relatively lower toxicity but as Cu has high mass fractions in several plastic products it nevertheless requires monitoring to manage the cumulative health effects of metal(loid)s.

In parallel, the 15 organic additives detected in the extreme setting of our plastic leaching experiments using targeted analysis were cross-checked against ToxCast. The assays covered in this database are indicative of various biological endpoints, such as hormone receptor interactions, cellular processes, receptor and transcription factor activities, and overall receptor agonist/antagonist actions. Twelve identified additives exhibited biological activity across a broad spectrum of assays (Fig. S33). However, three additives — DPhP, Irgafos 126, and Tinuvin 327 — were not found in the database, although DPhP is a known metabolite of triphenylphosphate and developmental toxicity has been shown [83]. Notably, Cyanox 2246 in the high levels obtained in our leaching experiment would exhibit activity in 55 % of the tested assays, potentially raising toxicity concerns.

Additionally, TBBPA and HDEHP were active in 42 % and 34 % of the tested assays, respectively, suggesting a need for caution regarding their use. The diversity of these biological endpoints - spanning hormonal, genetic, cytotoxic, and cellular integrity aspects -illustrates the complex potential impacts of these additives.

Nevertheless, it must be acknowledged that the toxicological profile discussed above is merely a snapshot of a broader and more complex scenario. The additives detected with the targeted analysis in this study represent a small fraction of the complete chemical composition of the leachates. This toxicological profiling also does not account for the mixture toxicity that could result from the combination of these additives, nor does it consider the toxicity that may stem from phototransformation effects in the UV treatments or nano/microplastics released from the plastics into the leachates. The interactions within the complete leachate, including synergistic and antagonistic effects among constituents, remain uncharacterized. Furthermore, the concentrations in the leachates generated in our study were likely several orders of magnitude higher than what can be expected in the environment. Future toxicity studies should aim to capture the complex interplay of all components and their collective impact on human health and the environment using realistic settings. While cell line tests can indicate biological potential, tests on entire organisms are needed to investigate the potential indicated here and cover additional aspects such as possible biotransformation. Therefore, complementary studies including *in vitro* and *in vivo* testing will be needed to reveal effects of these mixtures that can neither be estimated/predicted from chemicals databases nor from the multiple analyses within our toolbox.

### *3.7. The extended coverage of our analytical toolbox*

Our work comprises plastic-related analyses from polymer identity to particle formation, metal(loid) determination, and targeted and non-targeted organic analyses, and it comprehensively addresses environmental concerns related with selected consumer plastic products. Our results of this UV leaching study showed that 1) the mechanical stress and elevated temperatures in the weathering chamber facilitated the release of microplastics from the pre-cut consumer products, 2) UV radiation enhanced the release of small particles, and 3) photodegradation and mechanical stress could induce further fragmentation. Furthermore, characterization using HRMS-based non-target screening 4) highlighted complex differences across plastic products and UV vs. DC samples, and 5) indicated increased leaching and the likely formation of possibly more bioavailable phototransformation products in UV-exposed samples. Targeted analysis 6) confirmed that vastly different chemical profiles could be expected from different plastic products and 7) indicated that leaching and transformation processes were enhanced under UV-exposure and depended on the specific chemical in question as well as the polymer type. Finally, metal(loid) analysis showed that 8) high quantities of metal(loid) additives could leach from consumer plastics into water and 9) UV radiation and elevated temperatures strongly increased the leaching.

In the current study, our comprehensive analytical toolbox was used to gain a multidimensional insight into potentially hazardous components of leachates from artificially UV-weathered plastics. The combined results of this study are illustrated in Fig. 7 (with details in Table S4 in SI_A). Visual and spectroscopic changes along with targeted analysis (organic and inorganic) and non-targeted mass spectrometric characterization shed light on a highly complex field that requires more than one or a few selected analytical techniques to obtain a comprehensive picture. In the case of PE_3, for example, the use of visual, spectroscopic, or metal(loid) data alone could have suggested that no changes had occurred in the material and that no hazardous components had leached. However, the targeted organic analysis revealed the release of an antioxidant and a flame retardant. In another case, the most severe visual changes were observed for PE_4, indicating the dissolution of the material, with distinct differences between the UV treatment and the DC control. However, all analytical techniques showed no noticeable, or only minor changes between the DC and the UV-treated samples. Moreover, the methods within the applied analytical toolbox showed potential for complementing and/or confirming each other. For example, leached plasticizer and flame-retardant components could be identified by the targeted organic analysis in the UV-treated PVC_1, which agreed well with its elevated content of Sb, used as a flame-retardant synergist. Similarly for PET_1, the results of the individual analyses supported and confirmed each other.

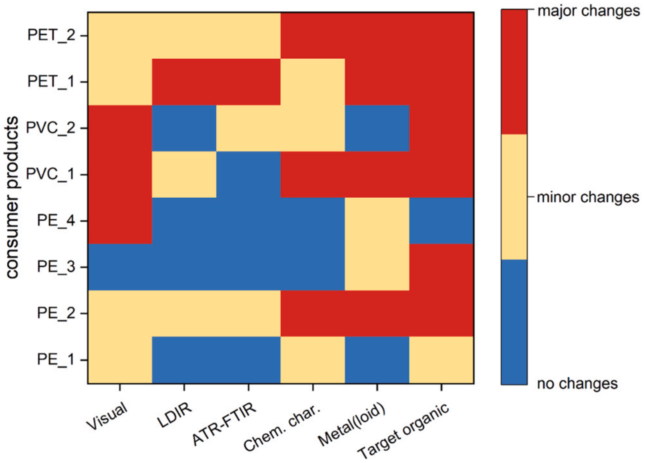


**Fig. 7.** Summary heat map of all analysis tools illustrating whether "no changes" (blue), "minor changes" (yellow) or "major changes" (red) were observed between the DC and the UV-treated samples.

It should be noted that an extreme setting, representing a worst-case scenario, was investigated, which is not realistic for environmental settings or matrices such as drinking water with low burdens of plastics. Chemicals were leached from new consumer plastics under comparably harsh conditions (strong UV light and elevated temperatures) using high plastic loads in comparably small water volumes and, therefore, slower processes and overall lower concentrations in the receiving phase may be expected in the considerably more complex natural environment. However, contact with abrasive materials like sand or animal shells can also facilitate mechanical degradation, and biofouling could either enhance (biofilms using the plastics as a carbon source) or slow down (shielding effect) degradation.

## 4. Conclusions

Ultimately, exposure to plastic particles, organic chemicals and potentially toxic elements represent a multiple stressor scenario common in the environment. Such complex settings require a range of analytical methods for adequate characterization. This pilot study highlights the chemical complexity of experimental leaching of plastic consumer products already under constant, simplified, and strong lab conditions and shows how extensive chemical characterization can provide valuable information on the leaching processes and on potential environmental and human health impacts. The experiments showed that common consumer products can release different toxicologically relevant trace metals and metalloids such as As, Cd, Sb, Sn or Pb. For the organic compounds, highly complex chemical compositions made up of several hundreds of relevant features were observed using LC-HRMS, only few of which could be investigated using targeted analysis, and clear indications of the formation of UV-transformation products were observed. Toxicity profiling indicated a generally high toxicological potential related with the different detected chemicals. This complexity of the leaching patterns generally calls for efforts to minimize and simplify chemicals added to plastics, to minimize the plastics' transfer into the environment as well as to pave the way for their recyclability, as discussed in the negotiations of the Global Plastics Treaty [84].

The analytical methods applied here were detailed, but not comprehensive. Plastic-associated organic chemicals include thousands of chemicals, of which only a small fraction was covered by the applied targeted analysis, and further unknown phototransformation products were likely formed during UV radiation. Future studies of the HRMS data following suspect and/or non-target screening approaches, which do not rely on the availability of analytical standards but are time- and resource intensive, would be a logical next step to address remaining blind spots. In addition, sample processing and matrix handling affecting the quantification methods should be further refined to allow the accurate quantification of the compounds detected during target analysis, which is essential for a future toxicological assessment. More complex leaching scenarios, e.g., involving different consumer product classes and natural leaching conditions (e.g., including river or seawater and biofouling) combined with bioanalytical tools (e.g., omics methods) for assessing the toxic potential of the leachates or their potential effects on the proteome level could further improve the future assessment of plastic leaching processes and the potential environmental and human health impacts of the leachates.

## Environmental implication

This pilot study illuminates the complex nature of chemicals and particles liberated from common consumer plastics when subjected to UV radiation mimicking prolonged sunlight exposure. We used complementary chemical analytical approaches to demonstrate the substantial release of complex chemical patterns and microplastics to water upon UV-induced weathering and photodegradation. Understanding these degradation processes is key to mitigating the environmental impact of plastic products during their lifetime and following disposal.

## Associated content

Author Information.
Notes.

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## CRediT authorship contribution statement

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## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

## Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.135256.

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