Environmental Degradation and Fragmentation of Microplastics: Dependence on Polymer Type, Humidity, UV Dose and Temperature

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Supporting Information

Scanning electron microscopy (SEM)

Polymer particles were prepared for Scanning Electron Microscope (SEM) (Apreo S, ThermoFisher Scientific) imaging by placing particles on a 12 mm SEM stub using ultra-smooth carbon adhesive tabs (Electron Microscopy Services). These polymer samples were prepared in a cleanroom and then gold sputter-coated at 12 mA for 600 s using a Denton Desk V sputter coater and gold target. These tabs were then placed in the SEM and imaged at various magnifications with settings of 5.0 kV beam and 25 pA current.

Differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC) (TA Instruments DSC 2500) was used to characterize thermal properties of the polymer samples. Samples were prepared by placing them in Tzero hermetic crimped pans. The polymer samples and pans were weighed and heated for 10°C/min for two heating and cooling cycles to specific ranges that were unique to each polymer in duplicate samples. TPU was not reported since it was an elastomer, PA-6 was heated from -50°C to 240°C, PP from -50°C to 200°C, HIPS from -50°C to 150°C, and LDPE from -50°C to 150°C. The resulting data files were then analyzed via TRIOS software for comparison to their pristine curves.

Attenuated total reflectance Fourier-transform infrared spectroscopy (ATR FT-IR)

The chemical composition of the microplastic surfaces before and after UV aging were analyzed by FT-IR spectroscopy to investigate changes in the carbonyl index, as described earlier.[1] The chemical compositions were determined using a Tensor 27, BRUKER FT-IR spectrometer with a diamond ATR accessory (platinum). The FT-IR spectra were recorded in the region of 4000-400 cm^{-1} with 30 scans at a resolution of 4 cm^{-1}. For determination of the carbonyl index, the ratio between two peaks was calculated:

HIPS 1720 cm^{-1} : 2920 cm^{-1}

LDPE 1715 cm^{-1} : 2900 cm^{-1}

PA-6 1715 cm^{-1} : 2860 cm^{-1}

PP 1725 cm^{-1} : 2918 cm^{-1}

TPU 1760 cm^{-1} : 2860 cm^{-1}

Gel permeation chromatography (GPC)

The molar mass distributions of the studied polymer powders before and after UV aging were determined by Gel Permeation Chromatography (GPC) as described earlier.[1]

For PA-6 and TPU, a modular GPC system with a combination of columns (HFIP-LG Guard column, 2 x PL HFIPgel columns, 300 x 7.5 mm, 3-100 µm, Agilent) was combined with a refractive index detector (8 µl, Agilent). The temperature was set to 35°C and the flow rate to 1.0 mL/min. Calibration was conducted using a polymethylmethacrylate calibration kit (with a known molar mass distribution) and 12 calibration points. Approximately 6 mg of each sample was dissolved in approximately 4 mL eluent (1,1,1,3,3,3 – Hexafluoro-2-propanol, Alfa Aesar) over 5 hours on a shaking table at room temperature. The resulting solutions were filtered through Millipore-Millex-FG 0.2 µm filters prior to injection.

For HIPS a modular GPC system with a combination of columns (PLgel Mixed-B guard column and 3 x PLgel Mixed-B columns, 300 x 7.5 mm, 10 µm, Agilent) was combined with a refractive index detector (8 µl, Agilent). The temperature was set to 35°C and the flow rate to 1.0 mL/min. The calibration was obtained with narrow molar mass standards (Polystyrene (PS) molar mass range 580-6870000 g/mol, Agilent). Extrapolation was used to estimate the molecular weight distribution outside the range of these calibration standards with respect to the exclusion and permeation limits. 2 mg of each sample was dissolved in approximately 4 mL eluent (Tetrahydrofuran, Fischer Chemical) overnight on a shaking table at room temperature. The resulting solutions were filtered through Chromafil Xtra PTFE FG 0.2 µm filters prior to injection.

For LDPE high temperature (HT-)GPC was used. The chromatographic system consisted of a PolymerChar GPC-IR (Valencia, Spain) high temperature GPC chromatograph equipped with an internal IR5 infra-red detector (IR5). The autosampler oven compartment was set at 160º Celsius and the column compartment was set at 150º Celsius. The columns used were 4 Agilent “Mixed A” 30cm 20-micron linear mixed-bed columns. The chromatographic solvent used was 1,2,4 trichlorobenzene and contained 200 ppm of butylated hydroxytoluene (BHT). The solvent source was nitrogen sparged. The injection volume used was 200 microliters and the flow rate was 1.0 milliliters/minute. Calibration of the GPC column set was performed with 21 narrow molecular weight distribution polystyrene standards with molecular weights ranging from 580 to 8,400,000 and were arranged in 6 “cocktail” mixtures with at least a decade of separation between individual molecular weights. The standards were purchased from Agilent Technologies.

The polystyrene standards were prepared at 0.025 grams in 50 milliliters of solvent for molecular weights equal to or greater than 1,000,000, and 0.05 grams in 50 milliliters of solvent for molecular weights less than 1,000,000. The polystyrene standards were pre-dissolved at 80 ºC with gentle agitation for 30 minutes then cooled and the room temperature solution is transferred cooled into the autosampler dissolution oven at 160ºC for 30 minutes. The polystyrene standard peak molecular weights were converted to polyethylene molecular weights using Equation 1 (as described in Williams and Ward, J. Polym. Sci., Polym. Let., 6, 621 (1968)).:

$$M_{polyethylene} = A \times \left(M_{polystyrene}\right)^{B} \qquad \text{(EQ1)}$$

where M is the molecular weight, A has a value of 0.4315 and B is equal to 1.0. A fifth order polynomial was used to fit the respective polyethylene-equivalent calibration points.

The total plate count of the GPC column set was performed with decane which was introduced into blank sample via a micropump controlled with the PolymerChar GPC-IR system. The plate count for the chromatographic system should be greater than 18,000 for the 4 Agilent “Mixed A” 30cm 20-micron linear mixed-bed columns.

Samples were prepared in a semi-automatic manner with the PolymerChar “Instrument Control” Software, wherein the samples were weight-targeted at 2 mg/ml, and the solvent (contained 200ppm BHT) was added to a pre nitrogen-sparged septa-capped vial, via the PolymerChar high temperature autosampler. The samples were dissolved for 2 hours at 160º Celsius under “low speed” shaking.

The calculations of M_n, M_w, and M_z were based on GPC results using the internal IR5 detector (measurement channel) of the PolymerChar GPC-IR chromatograph according to Equations 2-4, using PolymerChar GPCOne™ software, the baseline-subtracted IR chromatogram at each equally-spaced data collection point (i), and the polyethylene equivalent molecular weight obtained from the narrow standard calibration curve for the point (i) from Equation 1.

$$Mn = \frac{\sum^{i} IR_i}{\sum^{i}\left(IR_i / M_{polyethylene_i}\right)} \qquad \text{(EQ 2)}$$

$$Mw = \frac{\sum^{i}\left(IR_i * M_{polyethylene_i}\right)}{\sum^{i} IR_i} \qquad \text{(EQ 3)}$$

$$Mz = \frac{\sum^{i}\left(IR_i * M_{polyethylene_i}^{\ 2}\right)}{\sum^{i}\left(IR_i * M_{polyethylene_i}\right)} \qquad \text{(EQ 4)}$$

For PP Molecular Weight and Molecular Weight Distribution (MWD) was measured by way of GPC in 1,2,4-trichlorobenzene (TCB). Molecular weight parameters **(**$\overline{M}_n$**,** $\overline{M}_w$**,** $\overline{M}_z$**)** and molecular weight distributions for all the samples were measured by using a GPC-IR apparatus by PolymerChar, which was equipped with a column set of four PLgel Olexis mixed-bed (Polymer Laboratories) and an IR5 infrared detector (PolymerChar). The dimensions of the columns were 300 × 7.5 mm and their particle size 13 *μ*m. The mobile phase flow rate was kept at 1.0 mL/min. All the measurements were carried out at 150°C. Solution concentrations were 1.5 mg/mL (at 150°C) and 0.3 g/L of 2,6-diterbuthyl-*p*-chresole were added to prevent degradation. For GPC calculation, a universal calibration curve was obtained using 16 polystyrene (PS) standard samples supplied by PolymerChar (peak molecular weights ranging from 266 to 1220000). A third order polynomial fit was used for interpolating the experimental data and obtain the relevant calibration curve. Data acquisition and processing was done by using Empower 3 (Waters).

The Mark-Houwink relationship was used to determine the molecular weight distribution and the relevant average molecular weights: the *K* values were K_{PS} = 1.21 × 10^{-4} dL/g and K_{PP} = 1.90 × 10^{-4} dL/g for PS and polypropylene (PP) respectively, while the Mark-Houwink exponents α = 0.706 for PS and α = 0.725 for PP were used.

Fraunhofer light scattering

The size distribution as well as D10, D50 and D90 measurements were taken for all pristine cryomilled polymer particles as well as for three UV aged 2000 h particles (HIPS, TPU, PA-6) using a Mastersizer 3000 (Malvern, UK). These particles were sonicated, suspended in ethanol, and placed in the mixing receptacle at 2000 rpm until the obscuration was in the 2-10% range. The samples were measured 20-30 times and the last 10 measurements were used for analysis.

Single particle counter

Analogously to our previous study,[1] the secondary microplastic particles >1 µm were counted with an Abakus® Mobil Fluid laser particle counter (Klotz GmbH) equipped with a laser sensor LDS 2148(0) (1-139 µm). For this purpose, 0.7 mL of the NanoRelease dispersion was filled up to 500 mL with ultrapure water. For sensor protection the sample was filtered with a 190 µm pre-filter (nylon, Rotert). The

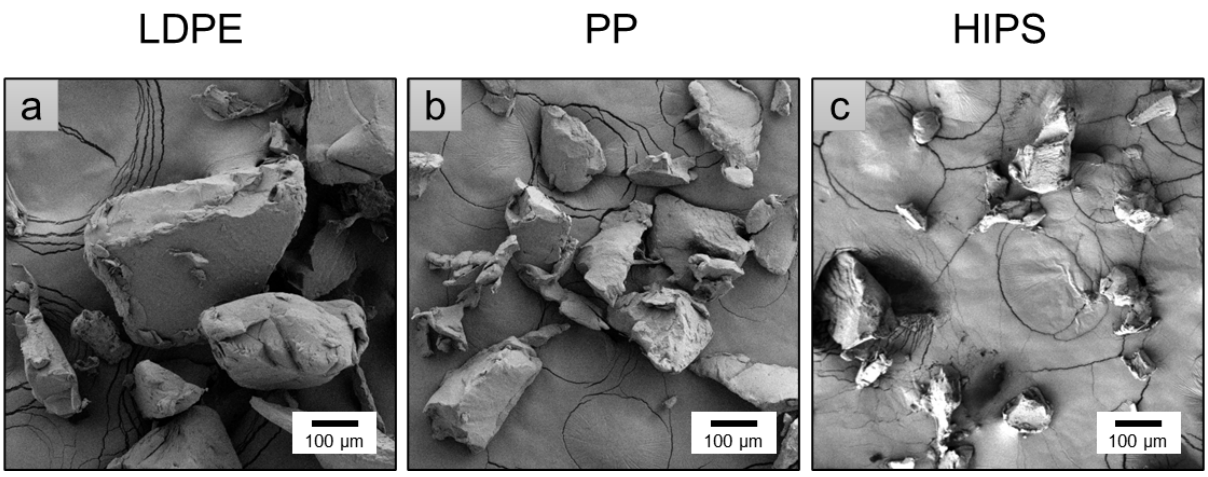
measurement was performed with 200 mL of the sample. Data evaluation was performed with C.A.R. Lab. Duplicate samples were measured and values for blank measurements were subtracted.

Analytical ultracentrifugation with refractive index detector (AUC-RI)

Analogously to our previous study,[1] Analytical Ultracentrifugation served to determine the concentration and particle size distribution of secondary nanoplastics. The AUC-Beckman XL centrifuge is equipped with an interference optical system synchronized to the centrifugal rotation, so that the colloidal sedimentation profile during centrifugation is monitored over time. By adjusting the centrifugation speed, the mass concentration and particle size distribution were determined in three overlapping size intervals of 10-150 nm (12 000 rpm ≙ 10 500 rcf), 40-800 nm (3 000 rpm ≙ 650 rcf) and 300-5000 nm (1 000 rpm ≙ 70 rcf); with 3 hours centrifugation time each. For data evaluation of the absorption profiles (resolved in time and radius) SedFit v14.0 was used with the corresponding polymer density (SI Table 1).[2] Duplicate samples were measured and values for blank measurements were subtracted. In the case of pristine LDPE ($\rho < 1$), D_2O instead of water was used to measure flotation instead of sedimentation. The density of LDPE increased due to UV aging,[3] enabling measurements by sedimentation.

Dissolved organic carbon (DOC)

To determine the concentration of released water-soluble organics, as described earlier,[1] the filtered NanoRelease sample was diluted with ultrapure water with dilution factors of 1:10 and 1:20 and total carbon (sample combusted catalytically on a Pt/Al_2O_3 catalyst in an oxygen stream at ca. 680 °C) and total inorganic carbon (sample acidified with 25 wt.-% phosphoric acid) were quantified as CO_2 via NDIR detection (Shimadzu TOC-L). DOC was calculated as the difference between total carbon and total inorganic carbon. Due to lack of substance, it was not possible to conduct duplicate measurements.



SI Figure 1: Scanning electron microscope (SEM) images showing the shapes and sizes of the polymer powders used for UV aging experiments: (a) LDPE, (b) PP, (c) HIPS. Images of the investigated PA-6 and TPU_ether_arom have been published in the SI of our previous study.[1]

SI Table 1: Physical-chemical properties of the investigated polymer powders. Data for the investigated PA-6 and TPU_ether_arom are published in the SI of our previous study.[1]

Polymer	Density [g/cm³][a]	T_g [°C][b]	T_m [°C][b]	Mean particle size d50 [µm][c]	M_n [g/mol][d]	M_w [g/mol][d]
LDPE	0.917	-	104.56 ± 0.05	420	9 830	63 600
PP	0.905	-	164.01 ± 0.15	294	24 958	276 458
HIPS	1.04	95.64 ± 0.03	-	178	62 000	199 000

[a] Values adopted from material safety datasheets of polymer materials.
[b] Values determined by DSC, 2nd heating curve, 10 K/min.
[c] Values determined by Fraunhofer light scattering, ISO13320.
[d] Values determined by GPC (see method description above).

SI Table 2: Parameters used in the three aging protocols investigated.

Aging protocol	**Kalahari**	**ISO4892, RH = 50%**	**ISO4892, RH = 75%**
Wavelength range	300-400 nm	300-400 nm	300-400 nm
UV intensity	60 W/m²	60 W/m²	60 W/m²
BST	90°C	65°C	65°C
Rel. humidity	28%	50%	75%

SI Table 3: Carbonyl index values measured for all polymers, all aging conditions.

Polymer	UV protocol	UV irradiation [h]	Carbonyl index
LDPE	pristine	0	0.001 ± 0.001
	Kalahari	1000	0.145 ± 0.008
		1500	0.171 ± 0.025
		2000	0.239 ± 0.018
	ISO4892, RH = 50%	1000	0.164 ± 0.010
		1500	0.160 ± 0.010
		2000	0.231 ± 0.005
	ISO4892, RH = 75%	1000	0.072 ± 0.003
		1500	0.123 ± 0.006
		2000	0.192 ± 0.006
PP	pristine	0	0.004 ± 0.001
	Kalahari	1000	0.010 ± 0.001
		1500	0.009 ± 0.001
		2000	0.011 ± 0.001
	ISO4892, RH = 50%	1000	0.012 ± 0.002
		1500	0.007 ± 0.001
		2000	0.012 ± 0.002
	ISO4892, RH = 75%	1000	0.012 ± 0.003
		1500	0.014 ± 0.002
		2000	0.015 ± 0.002
HIPS	pristine	0	0.027 ± 0.014
	Kalahari	1000	0.565 ± 0.084
		1500	0.755 ± 0.111
		2000	0.735 ± 0.102
	ISO4892, RH = 50%	1000	0.559 ± 0.043
		1500	0.910 ± 0.194
		2000	0.900 ± 0.102
	ISO4892, RH = 75%	1000	0.544 ± 0.114
		1500	0.626 ± 0.073
		2000	0.770 ± 0.109
PA-6	pristine	0	0.034 ± 0.011
	Kalahari	1000	0.256 ± 0.008
		1500	0.309 ± 0.015
		2000	0.411 ± 0.013
	ISO4892, RH = 50%	1000	0.309 ± 0.011
		1500	0.363 ± 0.016
		2000	0.400 ± 0.013
	ISO4892, RH = 75%	1000	0.323 ± 0.016
		1500	0.611 ± 0.084
		2000	0.562 ± 0.141
TPU	pristine	0	0.042 ± 0.003
	Kalahari	1000	0.450 ± 0.137
		1500	0.494 ± 0.108
		2000	0.671 ± 0.190
	ISO4892, RH = 50%	1000	0.450 ± 0.112
		1500	0.798 ± 0.382
		2000	1.383 ± 0.370
	ISO4892, RH = 75%	1000	0.317 ± 0.077
		1500	0.747 ± 0.266
		2000	0.994 ± 0.333

SI Table 4: Polydispersity (PDI), mass (M_w) and number (M_n) average molecular weight of the sol fraction of pristine and UV aged microplastics determined by GPC. Crosslinking after UV aging was observed for TPU and LDPE.

Polymer	UV protocol	UV irradiation [h]	M_n [g/mol]	M_w [g/mol]	PDI	Insoluble gel fraction [%]
PP	pristine	0	24958	276458	11.1	-
	Kalahari	2000	23335	228984	9.8	-
	ISO4892, RH = 50%	2000	21610	218589	10.1	-
	ISO4892, RH = 75%	2000	21355	219836	10.3	-
LDPE	pristine	0	9 830	63 600	-	-
	Kalahari	2000	1 230	1 870	-	52
	ISO4892, RH = 50%	2000	2 480	6 800	-	43
	ISO4892, RH = 75%	2000	1 520	2 770	-	40
HIPS	pristine	0	62 000	199 000	3.2	-
	Kalahari	2000	19 700	195 000	9.9	-
	ISO4892, RH = 50%	2000	15 500	189 000	12.2	-
	ISO4892, RH = 75%	2000	16 800	171 000	10.2	-
PA-6	pristine	0	16400	57900	3.5	-
	Kalahari	2000	2770	12000	4.3	-
	ISO4892, RH = 50%	2000	2350	8990	3.8	-
	ISO4892, RH = 75%	2000	2130	8130	3.8	-
TPU	pristine	0	32600	137000	4.2	-
	Kalahari	2000	1860	11400	6.1	73
	ISO4892, RH = 50%	2000	1940	14600	7.5	71
	ISO4892, RH = 75%	2000	2140	19200	9.0	69

SI Table 5: GPC molecular weight results of UV aged LDPE and TPU (2000h).

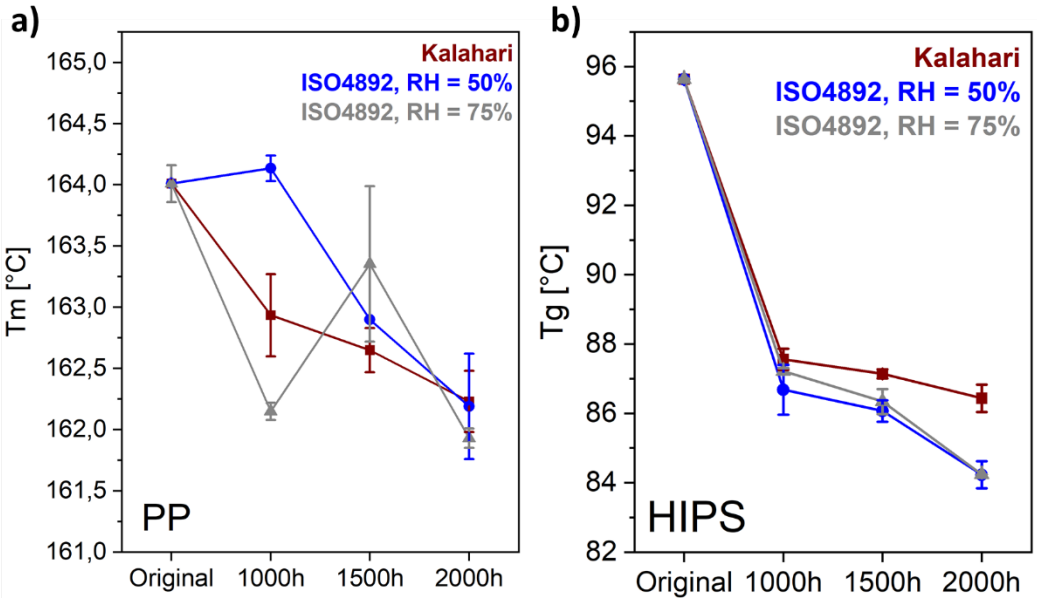
LINEAR LDPE equivalent values	M_w estimate (g/mol)	M_n estimate (g/mol)	Insoluble gel fraction* (%)
Kalahari	2 060 000	2 360	52
ISO4892, RH = 50%	1 730 000	5 370	43
ISO4892, RH = 75%	1 600 000	2 860	40

LINEAR TPU equivalent values	M_w estimate (g/mol)	M_n estimate (g/mol)	Insoluble gel fraction* (%)
Kalahari	1 463 000	6 870	73
ISO4892, RH = 50%	1 424 000	6 670	71
ISO4892, RH = 75%	1 386 000	6 890	69

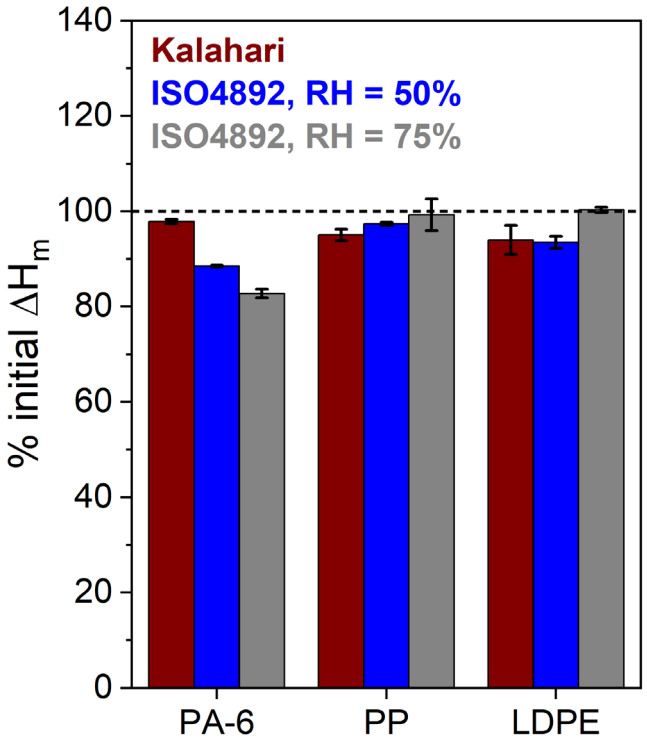
* Insoluble gel fraction comprises network polymer that is crosslinked beyond the gel point and was calculated as (100 - mass recovery)%, in which the mass recovery of the sample was calculated from the GPC-IR5 measurement channel chromatogram. The Mw and Mn values of the insoluble gel fraction were both conservatively estimated to be 3 983 000 g/mol (LDPE) and 2 000 000 g/mol (TPU), based on the column exclusion limit reported by the vendors in linear PE/TPU equivalent values. In reality, the molecular weight of the insoluble gel fraction is infinite. The M_w and M_n values were calculated according to the following equations:

$$M_{w,estimate} = f\left(M_{w,sol} + M_{w,gel}\right) = w_{sol} * M_{w,sol} + (1 - w_{sol}) * M_{w,gel,estimate}$$

$$M_{n,estimate} = f\left(M_{n,sol} + M_{n,gel}\right) = \frac{1}{\frac{w_{sol}}{M_{n,sol}} + \frac{1 - w_{sol}}{M_{n,gel,estimate}}}$$



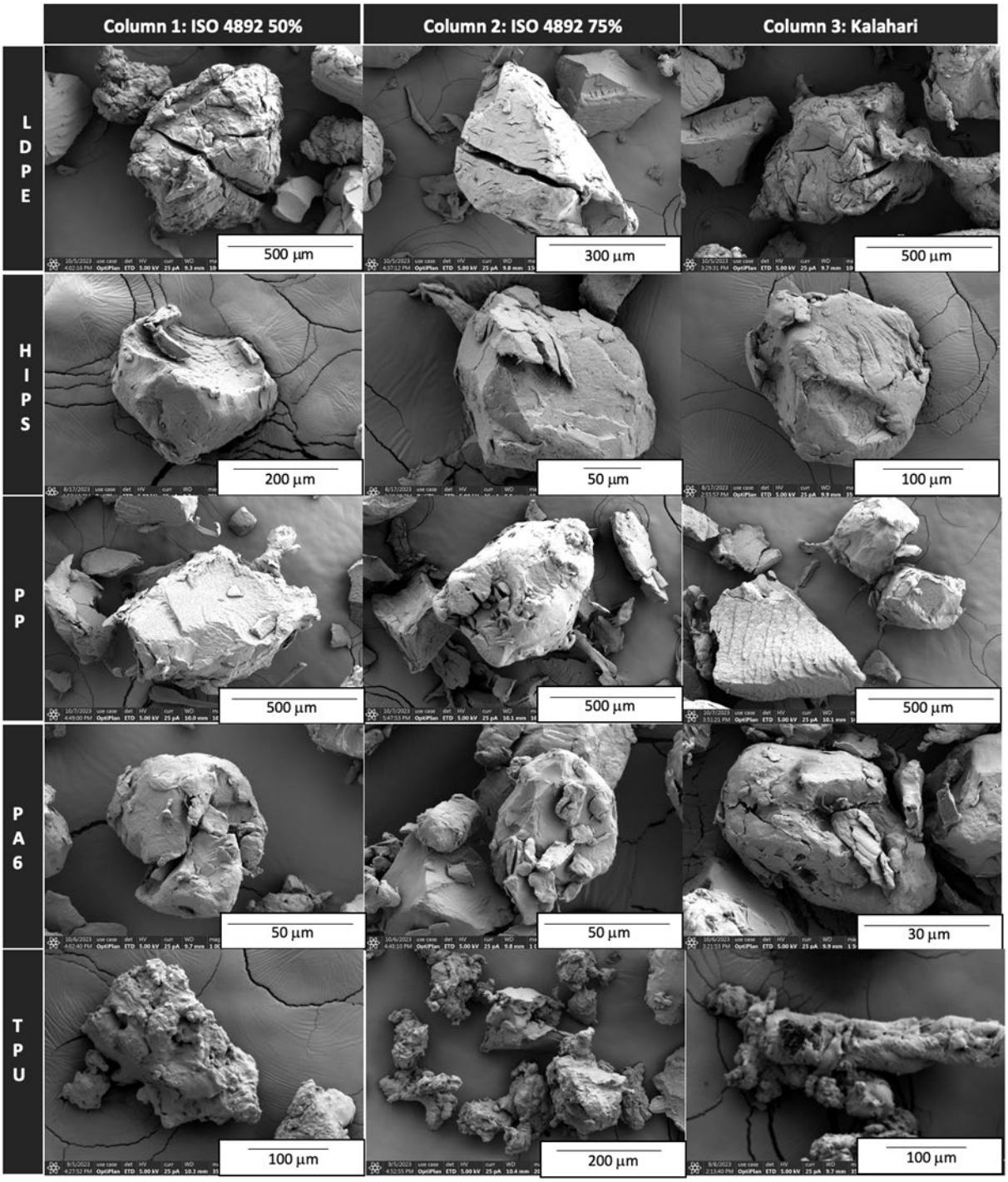
SI Figure 2: Changes in thermal properties of PP and HIPS after UV aging; a) melting point decrease of PP, b) glass transition temperature decrease of HIPS.



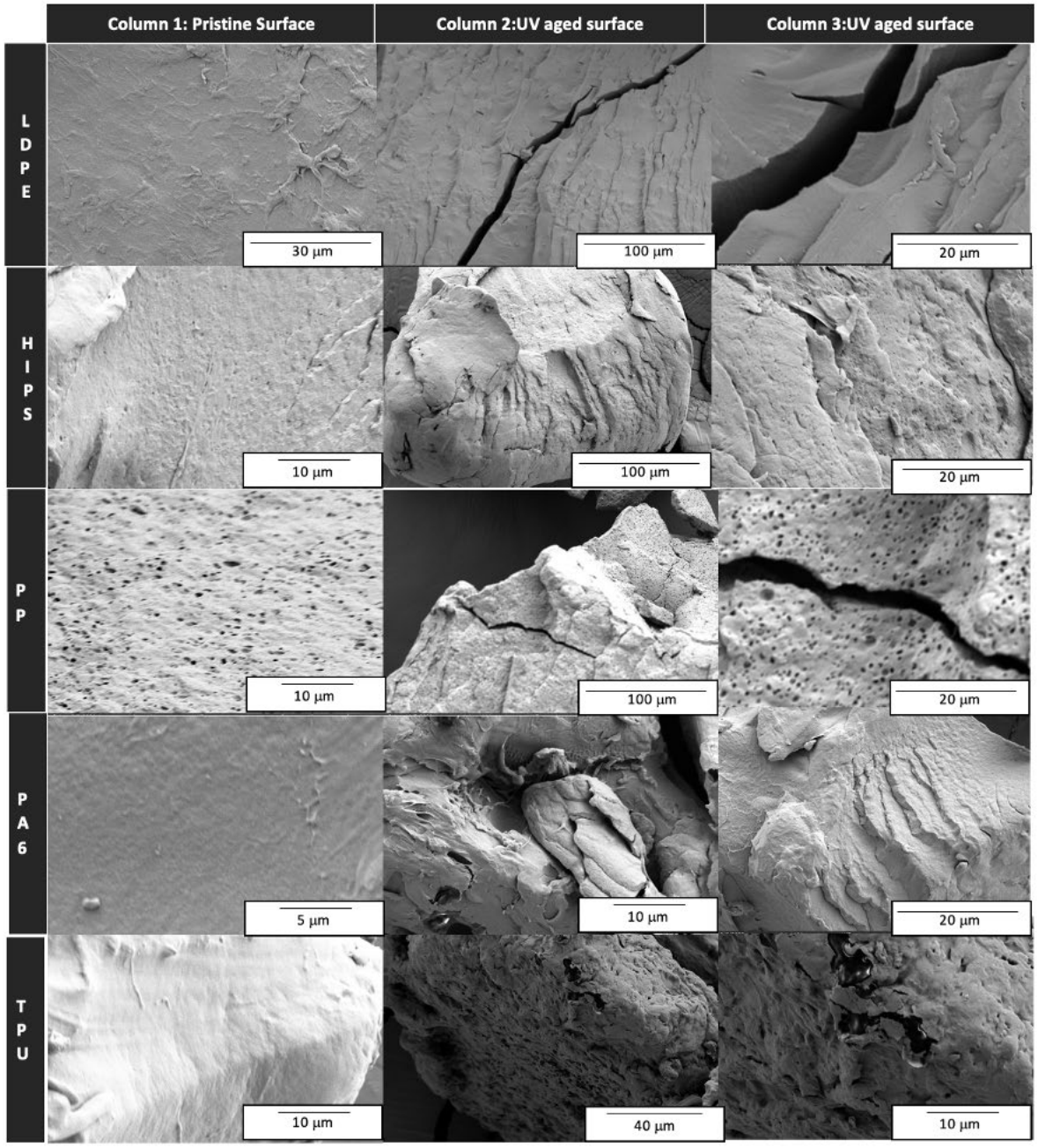
SI Figure 3: Percentage of initial ΔH_m (2^{nd} heating curve) after UV aging of PA-6, PP and LDPE by different UV aging protocols.

SI Table 6: Thermal properties of the investigated polymers after UV aging by three different protocols, duplicate measurements. Results were obtained by DSC data evaluation of the 2nd heating curve. Additional results (*) were obtained by DSC data evaluation of the 1st heating curve.

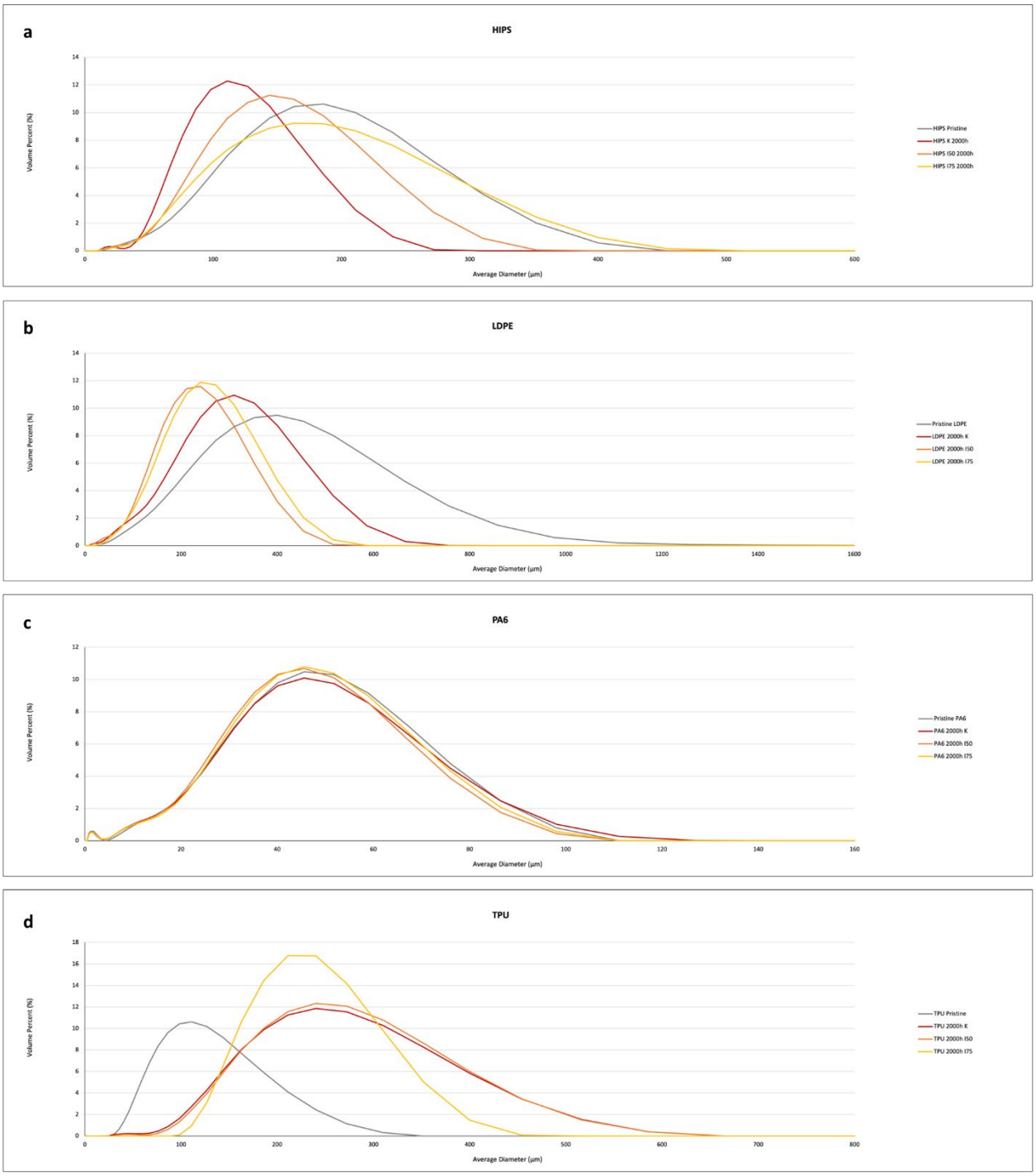
Polymer	UV protocol	UV irradiation [h]	T_g [°C]	T_m [°C]	ΔH_m [J/g]	Heating curve
LDPE	pristine	0	-	104.56 ± 0.05	131.24 ± 0.72	2nd
	Kalahari	1000	-	107.00 ± 0.29	118.70 ± 3.53	2nd
		1500	-	106.94 ± 0.31	121.52 ± 1.71	2nd
		2000	-	107.66 ± 0.38	123.38 ± 4.05	2nd
		2000*	-	111.07 ± 0.63	138.08 ± 1.94	1st
	ISO4892, RH = 50%	1000	-	104.50 ± 0.19	124.84 ± 0.73	2nd
		1500	-	105.98 ± 0.15	124.72 ± 0.09	2nd
		2000	-	107.41 ± 0.16	122.69 ± 1.69	2nd
		2000*	-	109.72 ± 0.05	135.29 ± 0.79	1st
	ISO4892, RH = 75%	1000	-	104.75 ± 0.09	117.78 ± 4.61	2nd
		1500	-	105.76 ± 0.47	124.65 ± 1.24	2nd
		2000	-	106.45 ± 0.46	131.66 ± 0.75	2nd
		2000*	-	108.03 ± 0.75	145.98 ± 1.89	1st
PP	pristine	0	-	164.01 ± 0.15	83.49 ± 1.42	2nd
	Kalahari	1000	-	162.94 ± 0.34	81.73 ± 2.26	2nd
		1500	-	162.65 ± 0.18	78.89	2nd
		2000	-	162.23 ± 0.25	79.35 ± 1.05	2nd
		2000*	-	165.91 ± 0.1	78.32 ± 1.57	1st
	ISO4892, RH = 50%	1000	-	164.14 ± 0.11	76.99 ± 0.55	2nd
		1500	-	162.90	74.75	2nd
		2000	-	162.19 ± 0.43	81.32 ± 0.26	2nd
		2000*	-	165.55 ± 0.24	76.039 ± 0.28	1st
	ISO4892, RH = 75%	1000	-	162.15 ± 0.07	79.33 ± 0.35	2nd
		1500	-	163.36 ± 0.64	82.39 ± 0.41	2nd
		2000	-	161.93 ± 0.08	82.88 ± 2.76	2nd
		2000*	-	165.60	69.02	1st
HIPS	pristine	0	95.64 ± 0.03	-	-	2nd
	Kalahari	1000	87.56 ± 0.31	-	-	2nd
		1500	87.14 ± 0.13	-	-	2nd
		2000	86.44 ± 0.40	-	-	2nd
	ISO4892, RH = 50%	1000	86.68 ± 0.72	-	-	2nd
		1500	86.07 ± 0.31	-	-	2nd
		2000	84.23 ± 0.39	-	-	2nd
	ISO4892, RH = 75%	1000	87.22 ± 0.09	-	-	2nd
		1500	86.34 ± 0.36	-	-	2nd
		2000	84.24 ± 0.01	-	-	2nd
PA-6	pristine	0	-	217.88 ± 3.09	93.14 ± 3.86	2nd
	Kalahari	1000	-	210.07 ± 0.77	91.68 ± 0.35	2nd
		1500	-	202.62 ± 4.19	88.47 ± 0.11	2nd
		2000	-	200.98 ± 0.01	91.15 ± 0.41	2nd
	ISO4892, RH = 50%	1000	-	198.64 ± 2.90	95.64 ± 2.70	2nd
		1500	-	200.44 ± 1.31	90.87 ± 3.61	2nd
		2000	-	198.51 ± 1.82	82.42 ± 0.14	2nd
	ISO4892, RH = 75%	1000	-	206.77 ± 0.21	97.57 ± 0.25	2nd
		1500	-	201.08 ± 0.49	84.65 ± 0.24	2nd
		2000	-	194.35 ± 0.83	77.08 ± 0.75	2nd



SI Figure 4: SEM images of UV aged polymers for each UV aging protocol, all at 2000 h duration.



SI Figure 5: SEM images of UV aged polymer surfaces all at Kalahari 2000 h duration.



SI Figure 6: Particle size distribution of investigated polymer powders before and after 2000 h UV aging by 3 different protocols. A) HIPS, b) LDPE, c) PA-6, d) TPU. Size distributions were determined by Fraunhofer diffraction.

SI Table 7: Particle counts per gram LDPE for size fractions from 1 to 139 µm measured for all conditions.

		pristine	Kalahari		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
LDPE	1	258117 ± 118761	6555136 ± 115231	103045589 ± 12133572	107080275 ± 19665653
	2	53936 ± 24175	1909661 ± 108989	26267586 ± 2443481	27500361 ± 5322867
	3	25553 ± 11386	1099656 ± 84767	13228383 ± 1063983	13709753 ± 2439114
	4	14472 ± 5617	739461 ± 73639	7783706 ± 580122	8055053 ± 1334997
	5	9289 ± 2711	540306 ± 60778	5008636 ± 337742	5177994 ± 797617
	6	5714 ± 1392	413758 ± 49686	3454661 ± 213744	3535781 ± 500258
	7	5117 ± 1183	342467 ± 46894	2566125 ± 144908	2650483 ± 347650
	8	3147 ± 242	311806 ± 43900	2125964 ± 129014	2178875 ± 252642
	9	2997 ± 514	306933 ± 50794	1943628 ± 116028	1989975 ± 192808
	10	1206 ± 78	139639 ± 24017	814008 ± 47286	829136 ± 60203
	11	1394 ± 156	142183 ± 24217	795189 ± 49756	810831 ± 54358
	12	2419 ± 175	393717 ± 77939	1907925 ± 94075	1993914 ± 96236
	15	378 ± 56	126225 ± 25097	525272 ± 27617	576761 ± 8167
	18	306 ± 83	86014 ± 18458	334917 ± 19417	377139 ± 1578
	21	194 ± 56	63050 ± 13133	240508 ± 16897	271825 ± 4214
	25	97 ± 69	60075 ± 14325	214958 ± 20097	258147 ± 12881
	30	69 ± 14	27808 ± 6503	102708 ± 16681	127689 ± 12606
	35	0 ± 0	12953 ± 2953	46342 ± 11714	64494 ± 5839
	40	42 ± 14	3314 ± 897	13319 ± 4681	19375 ± 1986
	45	0 ± 0	2244 ± 328	9000 ± 3444	12967 ± 1283
	50	0 ± 0	1528 ± 56	5819 ± 2792	9458 ± 736
	55	0 ± 0	1167 ± 83	3972 ± 1972	6383 ± 561
	60	0 ± 0	639 ± 28	2583 ± 1500	4764 ± 292
	65	14 ± 14	883 ± 133	3111 ± 1917	5486 ± 403
	75	0 ± 0	250 ± 28	875 ± 431	1681 ± 236
	80	0 ± 0	56 ± 0	625 ± 514	1278 ± 0
	85	0 ± 0	56 ± 28	542 ± 264	861 ± 56
	90	0 ± 0	97 ± 69	444 ± 278	1194 ± 139
	100	0 ± 0	56 ± 28	208 ± 153	583 ± 56
	110	0 ± 0	0 ± 0	181 ± 97	500 ± 222
	125	0 ± 0	14 ± 14	42 ± 42	208 ± 14
	139	0 ± 0	14 ± 14	97 ± 97	194 ± 83

		pristine	ISO4892, RH = 50%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
LDPE	1	258117 ± 118761	25228356 ± 10887900	42167922 ± 4869861	47099211 ± 1688406
	2	53936 ± 24175	7819408 ± 2682636	12971122 ± 699194	13965175 ± 188114
	3	25553 ± 11386	4373575 ± 1280164	7340431 ± 119808	7268503 ± 406631
	4	14472 ± 5617	2815392 ± 706881	4721467 ± 98000	4407558 ± 389753
	5	9289 ± 2711	1972608 ± 436836	3283061 ± 152272	2939631 ± 327825
	6	5714 ± 1392	1465144 ± 305378	2425308 ± 171081	2088828 ± 272128
	7	5117 ± 1183	1156789 ± 231667	1906364 ± 164942	1613764 ± 249114
	8	3147 ± 242	1016586 ± 190047	1660108 ± 171497	1382525 ± 244131
	9	2997 ± 514	993383 ± 197178	1600186 ± 162569	1323664 ± 258703
	10	1206 ± 78	430764 ± 89214	688611 ± 67961	565142 ± 121081
	11	1394 ± 156	440592 ± 97319	688542 ± 64642	570369 ± 125458
	12	2419 ± 175	1160064 ± 296103	1740919 ± 125964	1426311 ± 346217
	15	378 ± 56	350128 ± 110711	489758 ± 9036	407503 ± 103669
	18	306 ± 83	235489 ± 87522	310553 ± 2092	261328 ± 62561
	21	194 ± 56	163581 ± 75069	214697 ± 9381	185933 ± 40767
	25	97 ± 69	151733 ± 79144	182489 ± 14833	168506 ± 27561
	30	69 ± 14	68675 ± 43981	77728 ± 9072	82947 ± 8392
	35	0 ± 0	32347 ± 23292	33336 ± 8792	40892 ± 975
	40	42 ± 14	9431 ± 7375	9239 ± 2333	12333 ± 639
	45	0 ± 0	7647 ± 6147	5911 ± 1394	9272 ± 756
	50	0 ± 0	5264 ± 4625	3528 ± 1361	6167 ± 361
	55	0 ± 0	3486 ± 2986	2653 ± 1097	4731 ± 325
	60	0 ± 0	2556 ± 2389	1889 ± 500	2842 ± 964
	65	14 ± 14	3139 ± 2694	1897 ± 492	4278 ± 1333
	75	0 ± 0	1014 ± 958	1000 ± 583	1444 ± 333
	80	0 ± 0	736 ± 597	611 ± 333	1111 ± 361
	85	0 ± 0	778 ± 611	375 ± 125	833 ± 333
	90	0 ± 0	833 ± 833	722 ± 361	1028 ± 194
	100	0 ± 0	458 ± 403	236 ± 153	458 ± 208
	110	0 ± 0	236 ± 236	306 ± 222	403 ± 69
	125	0 ± 0	319 ± 319	208 ± 153	222 ± 28
	139	0 ± 0	611 ± 611	208 ± 181	250 ± 56

		pristine	ISO4892, RH = 75%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
LDPE	1	258117 ± 118761	17331953 ± 3169175	41343258 ± 19789969	146605911 ± 11846783
	2	53936 ± 24175	4854583 ± 820422	11417522 ± 5351744	36128775 ± 3144936
	3	25553 ± 11386	2713686 ± 436992	6196336 ± 2834975	17474814 ± 1730186
	4	14472 ± 5617	1737853 ± 275975	3903358 ± 1747686	10010069 ± 977486
	5	9289 ± 2711	1212242 ± 191369	2687786 ± 1150686	6295739 ± 593383
	6	5714 ± 1392	893300 ± 134728	1929483 ± 806567	4265039 ± 387717
	7	5117 ± 1183	709225 ± 102475	1505911 ± 599800	3124447 ± 278108
	8	3147 ± 242	607447 ± 95647	1277250 ± 490139	2573733 ± 214833
	9	2997 ± 514	585831 ± 85525	1195267 ± 430600	2348822 ± 194506
	10	1206 ± 78	246003 ± 32975	507058 ± 174819	965361 ± 79161
	11	1394 ± 156	245736 ± 33408	492361 ± 165728	946533 ± 85628
	12	2419 ± 175	622594 ± 87733	1220092 ± 367519	2307011 ± 173794
	15	378 ± 56	179756 ± 25372	334772 ± 81467	656414 ± 43619
	18	306 ± 83	113436 ± 17136	214536 ± 40647	427736 ± 25097
	21	194 ± 56	75856 ± 11178	149164 ± 24703	313342 ± 15353
	25	97 ± 69	64533 ± 8694	128569 ± 9886	296319 ± 10931
	30	69 ± 14	25272 ± 4917	56536 ± 808	157792 ± 1653
	35	0 ± 0	8283 ± 1072	23844 ± 4372	81300 ± 811
	40	42 ± 14	1861 ± 222	5561 ± 1461	26917 ± 389
	45	0 ± 0	1056 ± 111	4447 ± 1375	19306 ± 83
	50	0 ± 0	542 ± 14	2108 ± 942	14125 ± 403
	55	0 ± 0	292 ± 14	1667 ± 833	9750 ± 194
	60	0 ± 0	181 ± 42	911 ± 550	7528 ± 222
	65	14 ± 14	153 ± 14	911 ± 578	9306 ± 750
	75	0 ± 0	56 ± 0	264 ± 181	3050 ± 328
	80	0 ± 0	28 ± 28	194 ± 111	2292 ± 97
	85	0 ± 0	0 ± 0	97 ± 69	1611 ± 194
	90	0 ± 0	28 ± 28	139 ± 28	2264 ± 292
	100	0 ± 0	0 ± 0	42 ± 14	1097 ± 42
	110	0 ± 0	0 ± 0	14 ± 14	833 ± 28
	125	0 ± 0	0 ± 0	28 ± 28	542 ± 125
	139	0 ± 0	0 ± 0	14 ± 14	500 ± 28

SI Table 8: Particle counts per gram PP for size fractions from 1 to 139 µm measured for all conditions.

		pristine	Kalahari		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
PP	1	298497 ± 56842	1409931 ± 881464	793622 ± 246917	3287444 ± 2031456
	2	59908 ± 8825	342944 ± 211683	201608 ± 69569	736667 ± 449122
	3	30967 ± 5200	183642 ± 109731	114122 ± 39467	368414 ± 219725
	4	20169 ± 3019	122881 ± 70969	75486 ± 22797	222517 ± 130383
	5	14769 ± 2514	84750 ± 45544	60056 ± 17917	151172 ± 84933
	6	11022 ± 1578	66389 ± 32533	47642 ± 12586	111514 ± 60703
	7	10219 ± 1519	56983 ± 27850	40828 ± 9617	87486 ± 47225
	8	8236 ± 519	49961 ± 21200	39817 ± 8172	78628 ± 39972
	9	9661 ± 950	51033 ± 21144	42864 ± 6514	78975 ± 37636
	10	4533 ± 283	24075 ± 10225	19844 ± 2856	35181 ± 17431
	11	4669 ± 564	24342 ± 8281	19044 ± 2428	36758 ± 17297
	12	14425 ± 1764	65381 ± 19342	55106 ± 5772	103489 ± 48861
	15	5939 ± 411	19028 ± 5683	17425 ± 2019	34475 ± 16525
	18	3533 ± 61	12558 ± 3919	10828 ± 1967	23986 ± 9792
	21	2919 ± 153	8531 ± 2347	8125 ± 986	18133 ± 7756
	25	3275 ± 192	6411 ± 2022	8161 ± 1728	18708 ± 8597
	30	1489 ± 28	2942 ± 758	3083 ± 528	7875 ± 2653
	35	828 ± 6	694 ± 250	1278 ± 222	3292 ± 1542
	40	183 ± 17	161 ± 22	333 ± 28	792 ± 347
	45	42 ± 14	111 ± 83	153 ± 69	583 ± 333
	50	42 ± 42	125 ± 42	139 ± 28	194 ± 56
	55	56 ± 28	0 ± 0	42 ± 14	278 ± 167
	60	14 ± 14	8 ± 8	42 ± 14	153 ± 97
	65	8 ± 8	0 ± 0	56 ± 0	111 ± 56
	75	0 ± 0	0 ± 0	42 ± 14	14 ± 14
	80	0 ± 6	0 ± 0	0 ± 0	14 ± 14
	85	0 ± 6	0 ± 0	0 ± 0	0 ± 0
	90	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	100	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	110	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	125	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	139	0 ± 0	0 ± 0	0 ± 0	0 ± 0

		pristine	ISO4892, RH = 50%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
PP	1	298497 ± 56842	1582553 ± 421181	1703947 ± 616836	6078100 ± 3183022
	2	59908 ± 8825	400486 ± 129436	423656 ± 158100	1738433 ± 937878
	3	30967 ± 5200	227456 ± 74928	235953 ± 86275	931008 ± 511608
	4	20169 ± 3019	158461 ± 52317	156833 ± 55650	568572 ± 305650
	5	14769 ± 2514	120306 ± 39589	115894 ± 41650	377744 ± 200622
	6	11022 ± 1578	97653 ± 32397	91103 ± 30719	265675 ± 139703
	7	10219 ± 1519	85661 ± 25733	75250 ± 24611	198847 ± 100619
	8	8236 ± 519	79197 ± 24681	67764 ± 22381	162431 ± 81092
	9	9661 ± 950	86369 ± 22981	69453 ± 21131	147986 ± 69514
	10	4533 ± 283	37036 ± 8047	31667 ± 9256	59378 ± 28511
	11	4669 ± 564	39019 ± 7525	33717 ± 10333	56644 ± 25761
	12	14425 ± 1764	92797 ± 19103	76144 ± 22139	128603 ± 58919
	15	5939 ± 411	22153 ± 5536	18522 ± 5439	29800 ± 13522
	18	3533 ± 61	13167 ± 2711	11194 ± 3572	15344 ± 7061
	21	2919 ± 153	7500 ± 1339	5578 ± 1967	8567 ± 4189
	25	3275 ± 192	4772 ± 1194	3950 ± 1594	5983 ± 2856
	30	1489 ± 28	1536 ± 397	1250 ± 333	1694 ± 861
	35	828 ± 6	550 ± 6	417 ± 194	689 ± 244
	40	183 ± 17	139 ± 56	97 ± 14	83 ± 28
	45	42 ± 14	111 ± 28	125 ± 14	78 ± 22
	50	42 ± 42	42 ± 14	56 ± 28	28 ± 0
	55	56 ± 28	111 ± 28	28 ± 28	0 ± 0
	60	14 ± 14	42 ± 14	0 ± 0	14 ± 14
	65	8 ± 8	42 ± 42	14 ± 14	42 ± 42
	75	0 ± 0	42 ± 14	0 ± 0	14 ± 14
	80	0 ± 6	0 ± 0	0 ± 0	0 ± 0
	85	0 ± 6	0 ± 0	0 ± 0	0 ± 0
	90	0 ± 0	0 ± 0	28 ± 0	28 ± 28
	100	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	110	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	125	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	139	0 ± 0	0 ± 0	0 ± 0	0 ± 0

		pristine	ISO4892, RH = 75%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
PP	1	298497 ± 56842	955975 ± 92764	1364647 ± 415547	4514350 ± 2075439
	2	59908 ± 8825	193061 ± 24667	309703 ± 93753	992397 ± 439608
	3	30967 ± 5200	98533 ± 8083	168100 ± 48411	500789 ± 212278
	4	20169 ± 3019	65503 ± 3558	111164 ± 33131	303722 ± 122439
	5	14769 ± 2514	46172 ± 2778	81275 ± 23936	204200 ± 78572
	6	11022 ± 1578	34711 ± 2067	64700 ± 18206	148397 ± 51714
	7	10219 ± 1519	30075 ± 892	54953 ± 17397	119328 ± 42783
	8	8236 ± 519	26531 ± 1514	51083 ± 15183	103317 ± 34872
	9	9661 ± 950	28306 ± 1106	55786 ± 17842	103672 ± 30267
	10	4533 ± 283	13417 ± 167	25133 ± 8783	48014 ± 11764
	11	4669 ± 564	14081 ± 492	27342 ± 9325	49553 ± 13131
	12	14425 ± 1764	39306 ± 667	76294 ± 29100	135239 ± 30578
	15	5939 ± 411	12278 ± 722	24478 ± 10511	42278 ± 5583
	18	3533 ± 61	7842 ± 731	17550 ± 7867	28322 ± 2833
	21	2919 ± 153	6244 ± 783	11944 ± 6000	20272 ± 2394
	25	3275 ± 192	4808 ± 586	11008 ± 5686	17408 ± 114
	30	1489 ± 28	2383 ± 644	4439 ± 2061	7147 ± 686
	35	828 ± 6	653 ± 208	1764 ± 903	3008 ± 936
	40	183 ± 17	222 ± 139	361 ± 28	522 ± 117
	45	42 ± 14	194 ± 28	208 ± 69	333 ± 0
	50	42 ± 42	69 ± 14	97 ± 97	181 ± 97
	55	56 ± 28	14 ± 14	97 ± 69	125 ± 14
	60	14 ± 14	0 ± 0	28 ± 28	69 ± 14
	65	8 ± 8	14 ± 14	14 ± 14	14 ± 14
	75	0 ± 0	0 ± 0	0 ± 0	28 ± 28
	80	0 ± 6	0 ± 0	0 ± 0	14 ± 14
	85	0 ± 6	0 ± 0	14 ± 14	0 ± 0
	90	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	100	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	110	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	125	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	139	0 ± 0	0 ± 0	0 ± 0	0 ± 0

SI Table 9: Particle counts per gram PA-6 for size fractions from 1 to 139 µm measured for all conditions.

		pristine	Kalahari		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
PA-6	1	58401803 ± 4340775	76807289 ± NA	101393150 ± NA	61640622 ± NA
	2	25122092 ± 954314	30178172 ± NA	32335233 ± NA	27028394 ± NA
	3	17665242 ± 338569	20538839 ± NA	19229067 ± NA	18824061 ± NA
	4	14410142 ± 118008	16188528 ± NA	13778094 ± NA	14860139 ± NA
	5	12590572 ± 11222	13706750 ± NA	10876717 ± NA	12540639 ± NA
	6	11357975 ± 10131	12081361 ± NA	9047433 ± NA	10989500 ± NA
	7	10792869 ± 67314	11312372 ± NA	8075428 ± NA	10201539 ± NA
	8	11190381 ± 78669	11499828 ± NA	7969722 ± NA	10303578 ± NA
	9	12808981 ± 114919	13011072 ± NA	8684956 ± NA	11571211 ± NA
	10	6217628 ± 56472	6238489 ± NA	4038194 ± NA	5563822 ± NA
	11	6689986 ± 68175	6682900 ± NA	4208589 ± NA	5977122 ± NA
	12	19145800 ± 180178	19247006 ± NA	11627600 ± NA	17140617 ± NA
	15	6196172 ± 58089	6432672 ± NA	3814978 ± NA	5719089 ± NA
	18	4172661 ± 34967	4507383 ± NA	2637528 ± NA	3952189 ± NA
	21	3109947 ± 26581	3466767 ± NA	2031750 ± NA	3050433 ± NA
	25	2986433 ± 33417	3517094 ± NA	1995583 ± NA	3087622 ± NA
	30	1526994 ± 19494	1970756 ± NA	1003194 ± NA	1649672 ± NA
	35	734328 ± 21411	1021789 ± NA	408750 ± NA	807511 ± NA
	40	191217 ± 7550	275806 ± NA	80889 ± NA	204528 ± NA
	45	114142 ± 4392	167889 ± NA	40111 ± NA	120306 ± NA
	50	63208 ± 4625	90139 ± NA	15889 ± NA	63750 ± NA
	55	30056 ± 1222	43639 ± NA	6417 ± NA	32194 ± NA
	60	13861 ± 611	20306 ± NA	2611 ± NA	13306 ± NA
	65	8264 ± 569	11694 ± NA	1139 ± NA	7639 ± NA
	75	792 ± 69	1500 ± NA	111 ± NA	861 ± NA
	80	486 ± 69	611 ± NA	28 ± NA	444 ± NA
	85	181 ± 69	361 ± NA	56 ± NA	167 ± NA
	90	139 ± 28	306 ± NA	0 ± NA	111 ± NA
	100	14 ± 14	56 ± NA	0 ± NA	0 ± NA
	110	0 ± 0	28 ± NA	0 ± NA	0 ± NA
	125	0 ± 0	0 ± NA	0 ± NA	28 ± NA
	139	0 ± 0	0 ± NA	0 ± NA	0 ± NA

		pristine	ISO4892, RH = 50%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
PA-6	1	58401803 ± 4340775	43869144 ± 1653783	61572417 ± 5357761	92549592 ± 395692
	2	25122092 ± 954314	21362681 ± 151608	28354539 ± 1435533	33345639 ± 614944
	3	17665242 ± 338569	16539600 ± 122578	20436367 ± 742567	21177294 ± 648011
	4	14410142 ± 118008	14212369 ± 215586	16456136 ± 478875	15022958 ± 651136
	5	12590572 ± 11222	12816286 ± 292169	14076839 ± 363022	11473133 ± 614994
	6	11357975 ± 10131	11790514 ± 323625	12437425 ± 328353	9225381 ± 564497
	7	10792869 ± 67314	11342708 ± 366492	11545228 ± 374717	7940342 ± 552303
	8	11190381 ± 78669	11826933 ± 448611	11608181 ± 448975	7453472 ± 548778
	9	12808981 ± 114919	13521822 ± 620311	12823242 ± 663419	7753194 ± 628900
	10	6217628 ± 56472	6587344 ± 366683	6061783 ± 422422	3499003 ± 298669
	11	6689986 ± 68175	7090569 ± 445969	6355425 ± 555619	3579133 ± 332500
	12	19145800 ± 180178	20279028 ± 1509044	16971375 ± 2576842	9237081 ± 1118758
	15	6196172 ± 58089	6439161 ± 625944	4872106 ± 1409828	2764169 ± 478597
	18	4172661 ± 34967	4150436 ± 446892	2966872 ± 1238806	1822917 ± 387472
	21	3109947 ± 26581	2830869 ± 352014	1944303 ± 1089992	1330008 ± 354353
	25	2986433 ± 33417	2175933 ± 258106	1562919 ± 1150781	1237356 ± 448450
	30	1526994 ± 19494	692336 ± 35636	592403 ± 522658	604733 ± 297794
	35	734328 ± 21411	180097 ± 33958	185669 ± 175942	258958 ± 153514
	40	191217 ± 7550	26803 ± 11525	28806 ± 28028	55653 ± 36486
	45	114142 ± 4392	13067 ± 6856	12278 ± 11944	29722 ± 18944
	50	63208 ± 4625	5133 ± 3578	4319 ± 4153	12944 ± 8694
	55	30056 ± 1222	2244 ± 1411	1708 ± 1625	5028 ± 3167
	60	13861 ± 611	792 ± 542	792 ± 764	1833 ± 1417
	65	8264 ± 569	361 ± 278	250 ± 250	1069 ± 764
	75	792 ± 69	14 ± 14	14 ± 14	83 ± 56
	80	486 ± 69	28 ± 0	14 ± 14	0 ± 0
	85	181 ± 69	14 ± 14	0 ± 0	42 ± 14
	90	139 ± 28	22 ± 6	14 ± 14	14 ± 14
	100	14 ± 14	0 ± 0	0 ± 0	0 ± 0
	110	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	125	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	139	0 ± 0	0 ± 0	0 ± 0	14 ± 14

		pristine	ISO4892, RH = 75%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
PA-6	1	58401803 ± 4340775	62356622 ± NA	56678239 ± NA	66830739 ± NA
	2	25122092 ± 954314	25389844 ± NA	17368694 ± NA	24306033 ± NA
	3	17665242 ± 338569	17716939 ± NA	10321378 ± NA	15560856 ± NA
	4	14410142 ± 118008	14243700 ± NA	7344228 ± NA	11341094 ± NA
	5	12590572 ± 11222	12208700 ± NA	5661472 ± NA	8878539 ± NA
	6	11357975 ± 10131	10866278 ± NA	4678144 ± NA	7409072 ± NA
	7	10792869 ± 67314	10158633 ± NA	4115978 ± NA	6638733 ± NA
	8	11190381 ± 78669	10327517 ± NA	4029306 ± NA	6538372 ± NA
	9	12808981 ± 114919	11581317 ± NA	4386872 ± NA	7205961 ± NA
	10	6217628 ± 56472	5529300 ± NA	2028156 ± NA	3448306 ± NA
	11	6689986 ± 68175	5900100 ± NA	2124239 ± NA	3692500 ± NA
	12	19145800 ± 180178	16752622 ± NA	5999822 ± NA	10799728 ± NA
	15	6196172 ± 58089	5473411 ± NA	1990944 ± NA	3739361 ± NA
	18	4172661 ± 34967	3762989 ± NA	1402622 ± NA	2728539 ± NA
	21	3109947 ± 26581	2861272 ± NA	1083983 ± NA	2199756 ± NA
	25	2986433 ± 33417	2850967 ± NA	1062383 ± NA	2408833 ± NA
	30	1526994 ± 19494	1547561 ± NA	508828 ± NA	1424161 ± NA
	35	734328 ± 21411	788639 ± NA	201833 ± NA	766728 ± NA
	40	191217 ± 7550	207300 ± NA	40222 ± NA	201444 ± NA
	45	114142 ± 4392	126617 ± NA	17694 ± NA	116639 ± NA
	50	63208 ± 4625	68239 ± NA	7500 ± NA	62194 ± NA
	55	30056 ± 1222	32878 ± NA	2917 ± NA	29389 ± NA
	60	13861 ± 611	15611 ± NA	750 ± NA	12972 ± NA
	65	8264 ± 569	8111 ± NA	472 ± NA	6861 ± NA
	75	792 ± 69	1167 ± NA	111 ± NA	750 ± NA
	80	486 ± 69	250 ± NA	56 ± NA	306 ± NA
	85	181 ± 69	222 ± NA	28 ± NA	28 ± NA
	90	139 ± 28	100 ± NA	0 ± NA	56 ± NA
	100	14 ± 14	28 ± NA	0 ± NA	0 ± NA
	110	0 ± 0	0 ± NA	0 ± NA	28 ± NA
	125	0 ± 0	0 ± NA	0 ± NA	0 ± NA
	139	0 ± 0	0 ± NA	0 ± NA	0 ± NA

SI Table 10: Particle counts per gram HIPS for size fractions from 1 to 139 µm measured for all conditions.

		pristine	Kalahari		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
HIPS	1	1042297 ± 267675	321500328 ± 15465633	228606328 ± 16836172	388898261 ± 11150822
	2	258286 ± 43769	60501339 ± 2532750	40764047 ± 3833469	71840139 ± 4106078
	3	133739 ± 7900	25584742 ± 865175	17180753 ± 1618381	29153292 ± 1809708
	4	78122 ± 233	13578261 ± 372650	9175186 ± 828831	14978619 ± 896253
	5	49206 ± 5344	8203886 ± 180469	5588375 ± 461114	8855200 ± 502967
	6	34442 ± 4969	5460983 ± 94489	3743578 ± 272789	5822497 ± 317697
	7	26144 ± 4644	3996150 ± 50822	2764464 ± 201731	4221397 ± 202097
	8	23261 ± 5539	3267544 ± 34400	2295569 ± 147892	3452072 ± 150461
	9	22300 ± 5272	2970831 ± 17147	2122328 ± 120128	3132847 ± 109908
	10	9511 ± 2650	1224872 ± 289	897269 ± 53753	1296114 ± 37886
	11	9181 ± 3014	1204031 ± 4131	888528 ± 47228	1276072 ± 25239
	12	22344 ± 7372	2994828 ± 46856	2259092 ± 97886	3190542 ± 20825
	15	6922 ± 2311	876178 ± 30956	690294 ± 20178	951625 ± 13036
	18	4356 ± 1578	575003 ± 17347	473550 ± 8922	634253 ± 22514
	21	3014 ± 1042	422794 ± 24011	360264 ± 1542	471869 ± 25147
	25	2722 ± 722	409067 ± 15894	357939 ± 9644	473306 ± 48306
	30	958 ± 181	213647 ± 13547	201153 ± 8792	255375 ± 37792
	35	333 ± 111	117592 ± 7075	115475 ± 11525	147472 ± 32889
	40	56 ± 0	38639 ± 722	39792 ± 4042	47189 ± 9911
	45	28 ± 28	27986 ± 236	28744 ± 4339	37153 ± 11375
	50	0 ± 0	21417 ± 56	21639 ± 3000	27861 ± 8361
	55	42 ± 14	15278 ± 500	16528 ± 2167	19425 ± 5814
	60	0 ± 0	10653 ± 458	12531 ± 1664	15619 ± 5381
	65	14 ± 14	13528 ± 1028	15328 ± 3867	19778 ± 7444
	75	0 ± 0	3972 ± 528	4508 ± 1158	5639 ± 2500
	80	0 ± 0	2889 ± 361	3431 ± 736	4078 ± 1633
	85	0 ± 0	2014 ± 42	2292 ± 847	2667 ± 889
	90	0 ± 0	2514 ± 125	2819 ± 708	3833 ± 1778
	100	0 ± 0	556 ± 56	1125 ± 319	1361 ± 472
	110	0 ± 0	486 ± 97	806 ± 444	1153 ± 542
	125	0 ± 0	208 ± 14	306 ± 139	556 ± 250
	139	0 ± 0	139 ± 28	239 ± 150	208 ± 97

		pristine	ISO4892, RH = 50%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
HIPS	1	1042297 ± 267675	262731892 ± 1528153	216072656 ± 48998906	215829778 ± 53154361
	2	258286 ± 43769	76764253 ± 1547231	47545503 ± 12010047	51662803 ± 13561153
	3	133739 ± 7900	36723808 ± 1261914	21315667 ± 5346344	25314514 ± 6385486
	4	78122 ± 233	20685736 ± 917058	11513006 ± 2833311	14815494 ± 3511528
	5	49206 ± 5344	12934528 ± 650394	6974058 ± 1648819	9594586 ± 2068986
	6	34442 ± 4969	8708294 ± 500244	4548619 ± 1051908	6657181 ± 1264069
	7	26144 ± 4644	6340089 ± 391150	3250931 ± 706697	5004731 ± 830847
	8	23261 ± 5539	5063353 ± 325658	2543667 ± 533839	4149692 ± 558069
	9	22300 ± 5272	4452275 ± 303425	2177292 ± 422564	3765517 ± 377639
	10	9511 ± 2650	1758386 ± 120697	844761 ± 159394	1539875 ± 102153
	11	9181 ± 3014	1640917 ± 121533	787842 ± 142825	1478944 ± 69111
	12	22344 ± 7372	3473164 ± 206797	1632550 ± 279967	3443094 ± 71667
	15	6922 ± 2311	741089 ± 27394	339619 ± 60375	898306 ± 92306
	18	4356 ± 1578	378686 ± 8608	166450 ± 30194	544347 ± 82986
	21	3014 ± 1042	211775 ± 1408	87028 ± 14278	368917 ± 76028
	25	2722 ± 722	137089 ± 10283	45950 ± 8483	310250 ± 81083
	30	958 ± 181	45492 ± 9125	10167 ± 2000	140514 ± 43319
	35	333 ± 111	16997 ± 5358	2522 ± 256	65583 ± 24389
	40	56 ± 0	5078 ± 2672	444 ± 83	17958 ± 7625
	45	28 ± 28	3322 ± 1444	194 ± 56	13819 ± 5903
	50	0 ± 0	2681 ± 1486	83 ± 28	9611 ± 4278
	55	42 ± 14	2114 ± 1419	97 ± 69	6903 ± 3681
	60	0 ± 0	1444 ± 1139	153 ± 14	4861 ± 2222
	65	14 ± 14	1958 ± 1403	83 ± 0	6347 ± 4014
	75	0 ± 0	819 ± 653	28 ± 0	2139 ± 1472
	80	0 ± 0	542 ± 375	0 ± 0	1208 ± 1014
	85	0 ± 0	503 ± 347	14 ± 14	1333 ± 861
	90	0 ± 0	514 ± 431	0 ± 0	1569 ± 1264
	100	0 ± 0	208 ± 181	0 ± 0	819 ± 736
	110	0 ± 0	181 ± 181	0 ± 0	569 ± 542
	125	0 ± 0	111 ± 111	0 ± 0	389 ± 333
	139	0 ± 0	42 ± 42	0 ± 0	361 ± 333

		pristine	ISO4892, RH = 75%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
HIPS	1	1042297 ± 267675	196429131 ± 28260869	257872658 ± 54462936	460327111 ± 160930872
	2	258286 ± 43769	38955658 ± 5275236	48776631 ± 10718614	124367814 ± 66429008
	3	133739 ± 7900	17276975 ± 2099958	20405797 ± 4217875	56539456 ± 32229350
	4	78122 ± 233	9499292 ± 1035469	10719411 ± 2071278	30021369 ± 17271142
	5	49206 ± 5344	5890494 ± 588561	6432158 ± 1182692	17716911 ± 10106217
	6	34442 ± 4969	3993611 ± 355561	4259208 ± 727503	11416572 ± 6404472
	7	26144 ± 4644	2974528 ± 245817	3121572 ± 518206	8118258 ± 4460342
	8	23261 ± 5539	2474550 ± 188011	2554483 ± 409044	6413242 ± 3431758
	9	22300 ± 5272	2301072 ± 145039	2327211 ± 341711	5621100 ± 2926239
	10	9511 ± 2650	972906 ± 54083	959633 ± 127828	2255281 ± 1131864
	11	9181 ± 3014	966539 ± 42294	943403 ± 120986	2139808 ± 1054264
	12	22344 ± 7372	2517969 ± 57575	2341858 ± 252808	5027372 ± 2360067
	15	6922 ± 2311	784806 ± 16606	687275 ± 57392	1368581 ± 591131
	18	4356 ± 1578	530925 ± 28881	455047 ± 24897	855772 ± 346811
	21	3014 ± 1042	409475 ± 32958	345842 ± 14408	598178 ± 225756
	25	2722 ± 722	413419 ± 51892	346672 ± 5722	540011 ± 174672
	30	958 ± 181	237350 ± 46294	195806 ± 306	264847 ± 68319
	35	333 ± 111	136458 ± 38958	117864 ± 4419	135253 ± 20486
	40	56 ± 0	44953 ± 15325	40417 ± 1944	40986 ± 3347
	45	28 ± 28	34514 ± 14014	32167 ± 2889	31347 ± 1014
	50	0 ± 0	26042 ± 11042	25097 ± 2264	22747 ± 1075
	55	42 ± 14	20050 ± 8478	20111 ± 2306	16161 ± 1578
	60	0 ± 0	14931 ± 7014	14764 ± 1208	11333 ± 2167
	65	14 ± 14	18431 ± 8736	21431 ± 1458	15306 ± 4167
	75	0 ± 0	6278 ± 3444	6833 ± 722	4917 ± 2083
	80	0 ± 0	4306 ± 2389	5042 ± 597	3458 ± 1458
	85	0 ± 0	2972 ± 1500	3972 ± 361	2639 ± 1361
	90	0 ± 0	3556 ± 2028	5042 ± 514	3597 ± 1764
	100	0 ± 0	1444 ± 1083	2194 ± 389	1500 ± 944
	110	0 ± 0	1361 ± 889	1667 ± 222	1250 ± 889
	125	0 ± 0	514 ± 403	611 ± 139	625 ± 514
	139	0 ± 0	472 ± 306	556 ± 56	597 ± 514

SI Table 11: Particle counts per gram TPU for size fractions from 1 to 139 µm measured for all conditions.

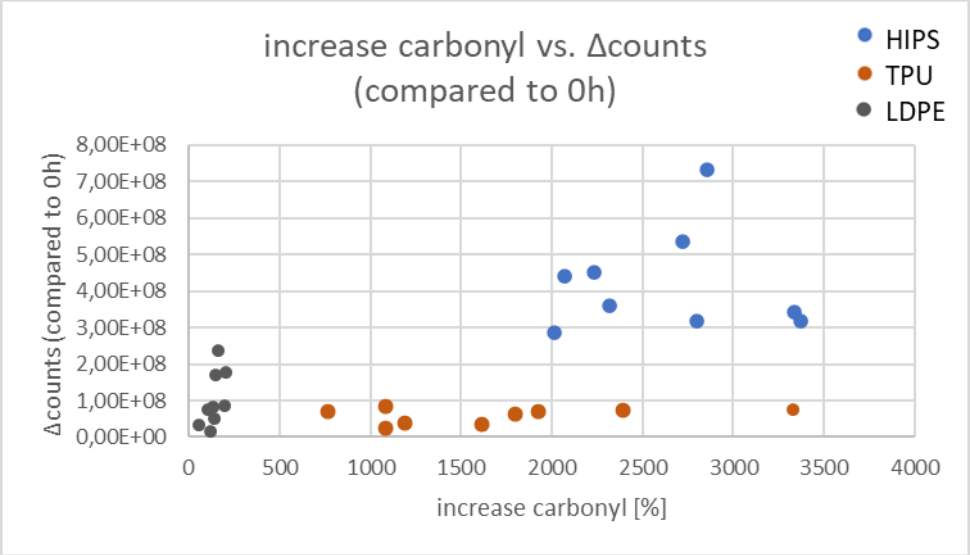
		pristine	Kalahari		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
TPU	1	242272 ± 123394	16908125 ± 3814097	26526503 ± 3194014	32088722 ± 4300439
	2	37081 ± 10308	3889697 ± 267742	5830278 ± 971133	3622383 ± 235989
	3	16810 ± 3399	1788792 ± 77925	2706019 ± 424281	1263950 ± 207511
	4	9385 ± 824	944442 ± 122681	1476267 ± 204589	597022 ± 126622
	5	6481 ± 481	548772 ± 95961	893089 ± 95917	329000 ± 77144
	6	4838 ± 657	344919 ± 78353	577756 ± 47494	204664 ± 51636
	7	4046 ± 24	228250 ± 53667	407292 ± 30247	136897 ± 36914
	8	4199 ± 546	165839 ± 44150	299042 ± 8497	102033 ± 26717
	9	4078 ± 411	134078 ± 38628	256072 ± 756	85600 ± 17906
	10	1988 ± 224	51658 ± 15553	95372 ± 2017	32825 ± 6253
	11	1628 ± 267	46558 ± 11942	88511 ± 928	29592 ± 5758
	12	4717 ± 1800	103878 ± 22044	182806 ± 239	60469 ± 10469
	15	1222 ± 417	26594 ± 3506	44217 ± 1939	13092 ± 2981
	18	839 ± 394	16358 ± 2247	25836 ± 1197	6886 ± 1953
	21	575 ± 353	11331 ± 481	17458 ± 1292	3828 ± 1272
	25	803 ± 331	9456 ± 411	13875 ± 514	2447 ± 1069
	30	258 ± 64	3675 ± 19	4861 ± 56	847 ± 347
	35	129 ± 115	1153 ± 181	1806 ± 222	250 ± 139
	40	35 ± 21	292 ± 69	292 ± 42	83 ± 56
	45	42 ± 42	111 ± 28	208 ± 14	42 ± 14
	50	7 ± 7	125 ± 14	56 ± 28	56 ± 0
	55	0 ± 0	139 ± 56	42 ± 14	14 ± 14
	60	14 ± 14	42 ± 14	28 ± 28	28 ± 0
	65	14 ± 14	56 ± 0	14 ± 14	0 ± 0
	75	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	80	0 ± 0	14 ± 14	0 ± 0	0 ± 0
	85	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	90	7 ± 7	0 ± 0	0 ± 0	0 ± 0
	100	0 ± 0	0 ± 0	14 ± 14	0 ± 0
	110	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	125	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	139	0 ± 0	0 ± 0	0 ± 0	14 ± 14

		pristine	ISO4892, RH = 50%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
TPU	1	242272 ± 123394	48900419 ± 13969558	46652658 ± 2034936	47614831 ± 12849669
	2	37081 ± 10308	7380336 ± 3718858	9858753 ± 916925	10224608 ± 3249181
	3	16810 ± 3399	2802206 ± 1478667	4744056 ± 539822	4998431 ± 1510814
	4	9385 ± 824	1335964 ± 675981	2761997 ± 358814	2961503 ± 848431
	5	6481 ± 481	740681 ± 330581	1788575 ± 244642	1936758 ± 505636
	6	4838 ± 657	458667 ± 166667	1204367 ± 179078	1326453 ± 318175
	7	4046 ± 24	305503 ± 72886	888903 ± 134903	969864 ± 214181
	8	4199 ± 546	241911 ± 35822	712169 ± 121669	779217 ± 155700
	9	4078 ± 411	215744 ± 2383	628886 ± 108425	673144 ± 125089
	10	1988 ± 224	88753 ± 7547	248747 ± 41253	266306 ± 41917
	11	1628 ± 267	86006 ± 11622	236997 ± 41142	242194 ± 32211
	12	4717 ± 1800	216658 ± 53919	486972 ± 83794	507339 ± 66844
	15	1222 ± 417	61458 ± 21175	104547 ± 21547	102781 ± 10447
	18	839 ± 394	38381 ± 15408	52908 ± 11764	51947 ± 6375
	21	575 ± 353	27578 ± 11228	28714 ± 6336	27733 ± 3017
	25	803 ± 331	26539 ± 12456	18531 ± 5558	17408 ± 2636
	30	258 ± 64	13842 ± 7203	6114 ± 2053	5792 ± 1125
	35	129 ± 115	6575 ± 3825	2042 ± 569	1906 ± 361
	40	35 ± 21	1981 ± 1231	481 ± 131	417 ± 56
	45	42 ± 42	1097 ± 625	319 ± 69	222 ± 56
	50	7 ± 7	1167 ± 750	194 ± 139	181 ± 14
	55	0 ± 0	528 ± 306	42 ± 42	111 ± 0
	60	14 ± 14	347 ± 292	97 ± 69	42 ± 14
	65	14 ± 14	625 ± 431	97 ± 14	28 ± 0
	75	0 ± 0	167 ± 111	14 ± 14	28 ± 0
	80	0 ± 0	42 ± 42	28 ± 0	14 ± 14
	85	0 ± 0	83 ± 83	14 ± 14	0 ± 0
	90	7 ± 7	56 ± 56	14 ± 14	14 ± 14
	100	0 ± 0	42 ± 42	0 ± 0	14 ± 14
	110	0 ± 0	28 ± 28	0 ± 0	0 ± 0
	125	0 ± 0	28 ± 28	0 ± 0	0 ± 0
	139	0 ± 0	14 ± 14	0 ± 0	0 ± 0

		pristine	ISO4892, RH = 75%		
Polymer	size [µm]	0 h	1000 h	1500 h	2000 h
TPU	1	242272 ± 123394	52686561 ± 2084039	49540775 ± 14044414	53677611 ± 2268378
	2	37081 ± 10308	9768792 ± 1065531	7420206 ± 2416656	9537269 ± 722397
	3	16810 ± 3399	3878294 ± 522344	2838494 ± 784950	4188858 ± 454703
	4	9385 ± 824	1822042 ± 245097	1364544 ± 285800	2272283 ± 279133
	5	6481 ± 481	961900 ± 123072	763253 ± 108364	1368319 ± 193897
	6	4838 ± 657	551836 ± 67069	475603 ± 33653	900906 ± 119639
	7	4046 ± 24	351919 ± 39336	328997 ± 4914	646447 ± 83236
	8	4199 ± 546	244444 ± 30094	253894 ± 13878	510500 ± 51533
	9	4078 ± 411	188069 ± 27236	220706 ± 21011	444675 ± 37414
	10	1988 ± 224	67692 ± 9692	88642 ± 12753	175631 ± 10575
	11	1628 ± 267	63142 ± 8803	84122 ± 14039	164053 ± 7975
	12	4717 ± 1800	131056 ± 24078	195303 ± 35642	379319 ± 7158
	15	1222 ± 417	31739 ± 6906	51653 ± 11542	94089 ± 2406
	18	839 ± 394	17244 ± 3244	31542 ± 7431	54856 ± 994
	21	575 ± 353	10869 ± 2314	20903 ± 6292	34386 ± 2897
	25	803 ± 331	7014 ± 1653	17328 ± 6033	25000 ± 1722
	30	258 ± 64	2514 ± 625	6431 ± 3181	8417 ± 722
	35	129 ± 115	722 ± 0	2764 ± 1292	3153 ± 514
	40	35 ± 21	125 ± 42	514 ± 292	542 ± 42
	45	42 ± 42	69 ± 14	486 ± 431	292 ± 42
	50	7 ± 7	14 ± 14	264 ± 153	111 ± 0
	55	0 ± 0	14 ± 14	153 ± 125	83 ± 0
	60	14 ± 14	0 ± 0	28 ± 28	69 ± 14
	65	14 ± 14	0 ± 0	56 ± 56	0 ± 0
	75	0 ± 0	0 ± 0	28 ± 28	0 ± 0
	80	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	85	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	90	7 ± 7	0 ± 0	0 ± 0	0 ± 0
	100	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	110	0 ± 0	0 ± 0	0 ± 0	14 ± 14
	125	0 ± 0	0 ± 0	0 ± 0	0 ± 0
	139	0 ± 0	0 ± 0	0 ± 0	0 ± 0

SI Table 12: Secondary micro- (1-139 µm), nanoplastic (0.01-0.99 µm) and DOC (< 0.02 µm) masses per gram polymer obtained for pristine and 2000h aged microplastics after NanoRelease tests.

Polymer	Aging	Microplastics [mg/$g_{polymer}$]	Nanoplastics [mg/$g_{polymer}$]	Dissolved [mg/$g_{polymer}$]
PA-6	pristine	193.33 ± 3.27	0.08	2.57
	Kalahari	193.03	0.37 ± 0.21	28.78
	ISO4892, RH = 50%	81.88 ± 24.47	2.36 ± 1.30	47.44
	ISO4892, RH = 75%	151.63	0.61 ± 0.09	47.44
HIPS	pristine	0.16 ± 0.04	0.02	< 0.08
	Kalahari	47.02 ± 8.46	2.46 ± 1.34	4.36
	ISO4892, RH = 50%	30.69 ± 6.71	1.36 ± 0.42	10.89
	ISO4892, RH = 75%	55.44 ± 10.32	3.40 ± 0.96	3.73
PP	pristine	0.13 ± 0.00	0.05 ± 0.01	0.11
	Kalahari	0.86 ± 0.40	0.12 ± 0.04	0.17
	ISO4892, RH = 50%	0.62 ± 0.29	0.12 ± 0.02	0.15
	ISO4892, RH = 75%	0.89 ± 0.06	0.22 ± 0.03	0.19
LDPE	pristine	0.02 ± 0.00	0 ± 0	< 0.08
	Kalahari	19.45 ± 0.14	2.26 ± 0.12	5.29
	ISO4892, RH = 50%	13.56 ± 0.60	0.52 ± 0.01	12.44
	ISO4892, RH = 75%	26.08 ± 0.64	1.15 ± 0.37	2.96
TPU	pristine	0.05 ± 0.01	0.27	0.08
	Kalahari	0.48 ± 0.13	0.80 ± 0.08	7.23
	ISO4892, RH = 50%	3.07 ± 0.54	0.84 ± 0.00	12.44
	ISO4892, RH = 75%	2.61 ± 0.07	1.45 ± 0.26	7.39



SI Figure 7: Correlation-plot of Δcounts of microplastics vs. carbonyl index increase [%] for HIPS, TPU and LDPE.

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