

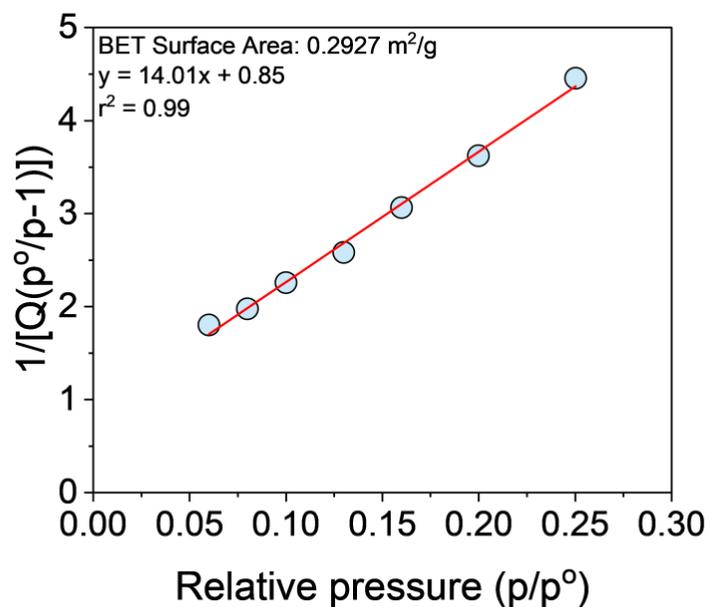
## **Uptake and release of perfluoroalkyl carboxylic acids (PFCAs) from macro and microplastics**

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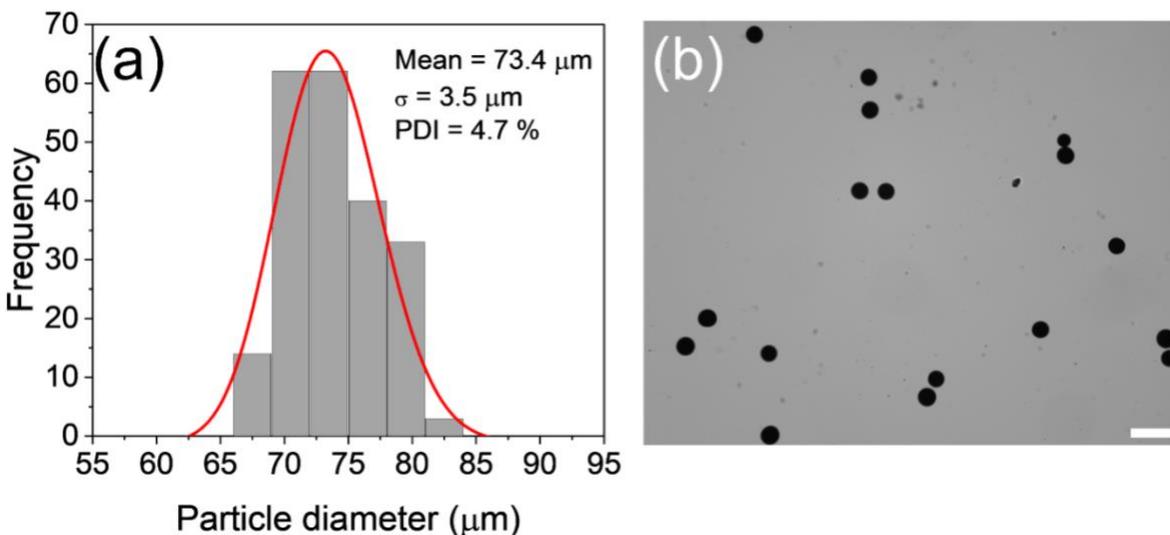
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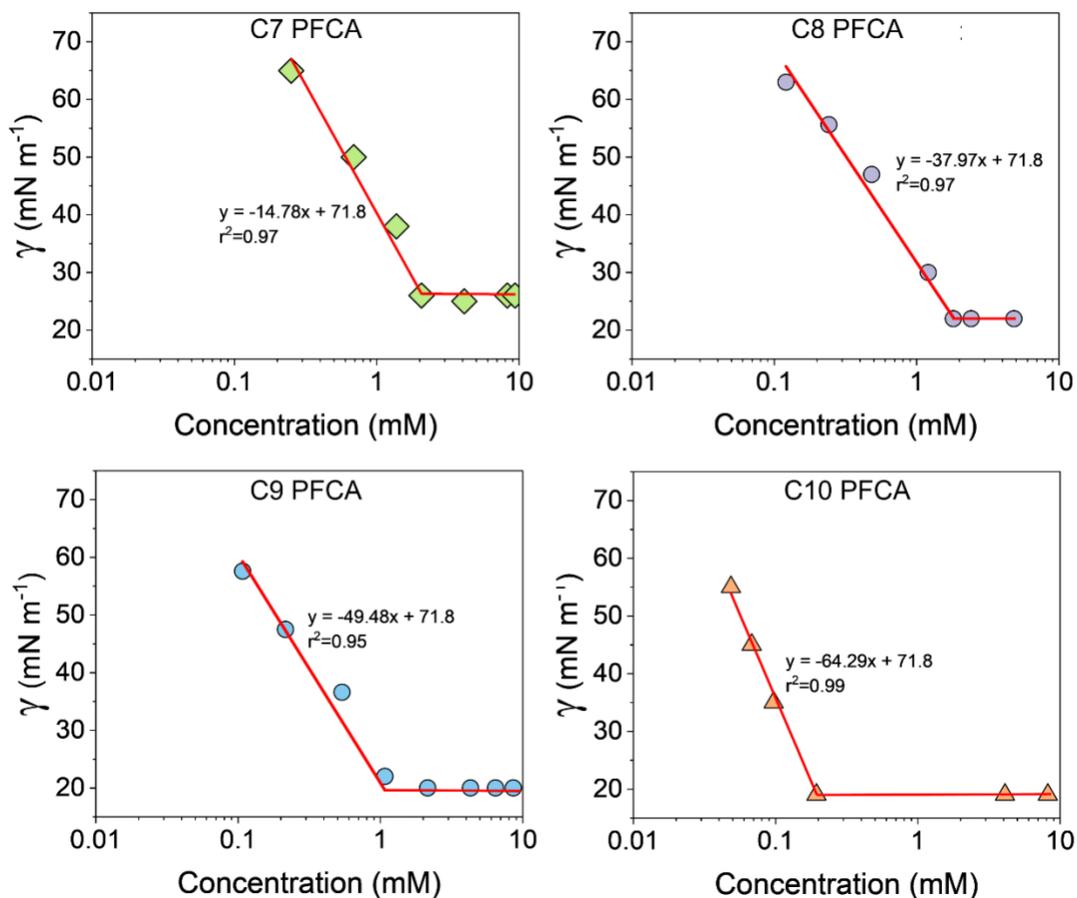
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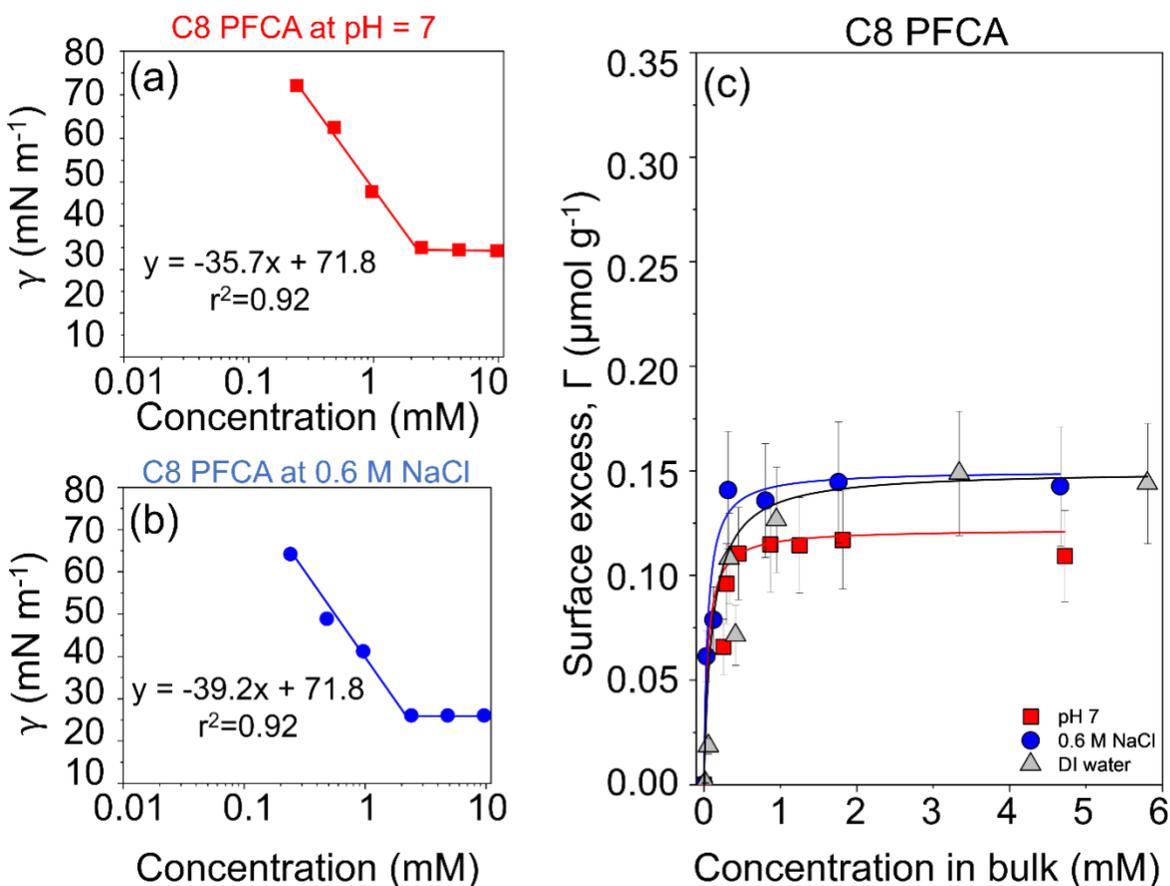
**Fig. S1.** Brunauer-Emmett-Teller (BET) plot of the model PE MPs. The surface area of the MPs was measured by N<sub>2</sub> adsorption-desorption using the Quantachrome AS-1 BET system. The surface areas were calculated from the adsorption isotherm by using a BET algorithm.



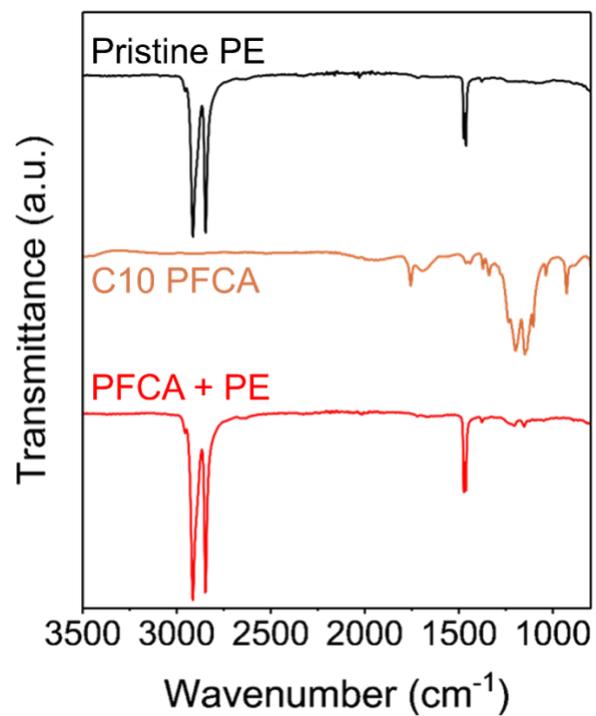
**Fig. S2.** (a) Particle size distribution and (b), optical microscopy image of the polyethylene microplastics used in the experiments. The size distribution in (a) is obtained by analyzing ten optical microscope images of the MPs (as shown in (b)). Here  $\sigma$  is the standard deviation in the microplastic diameter (size) as given by a log-normal distribution (red line) and PDI is the polydispersity index. Scale bar in (b) is 100 μm.



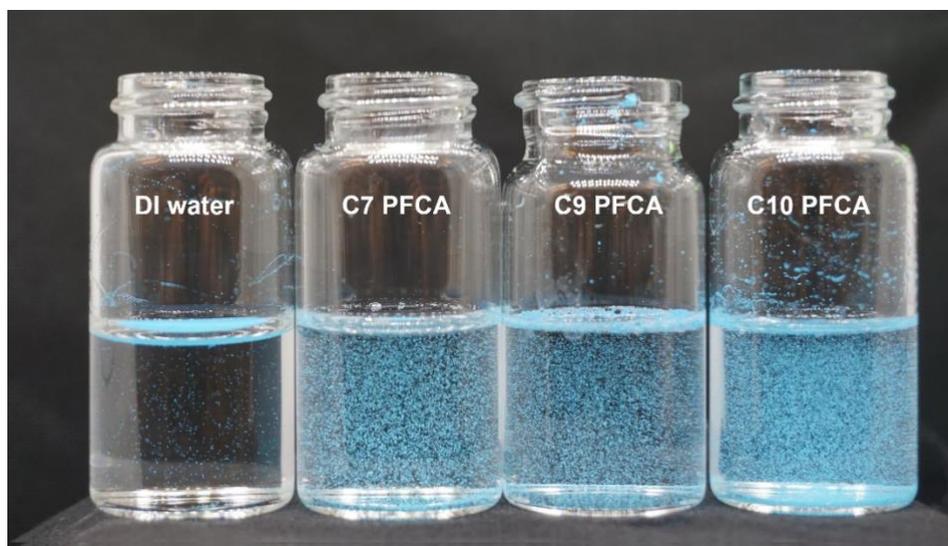
**Fig. S3** Surface tension calibration curves of (a) C7, (b) C8, (c) C9 and (d) C10 PFCAs. The points are the experimentally measured surface tension values of the PFCA solutions, and the red lines are the linear fits to the experimental data. The equation of line representing the surface tension decrease in the range up to the CMC is given in the plots and the  $r^2$  value is given to demonstrate the goodness-of-fit of the calibration curves.



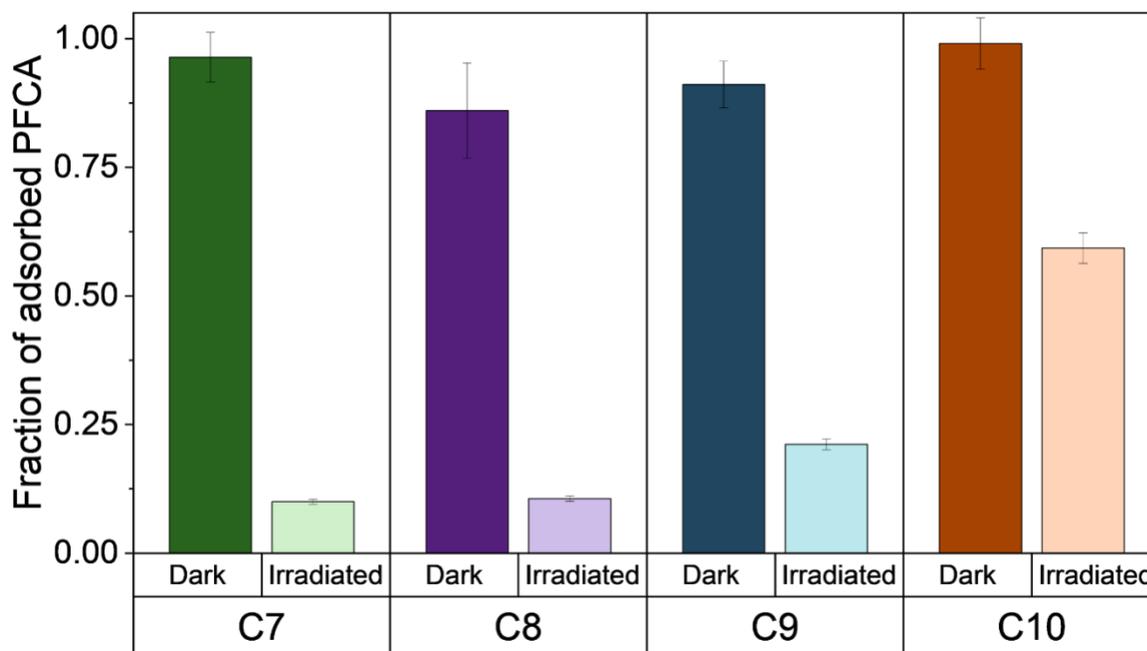
**Fig. S4** Surface tension calibration curves for C8 PFCA with controlled pH and high salinity. (a) Surface tension calibration curve of C8 PFCA in aqueous solution of adjusted to pH =7. (b) Surface tension calibration curve of C8 PFCA in solution containing 0.6 M NaCl. The points are the experimentally measured surface tension values of the PFCA solutions, and the lines are the linear fits to the experimental data. The equation of line representing the surface tension decrease in the range up to the CMC is given in the plots and the  $r^2$  value is given to demonstrate the goodness-of-fit of the calibration curves. (c) The adsorption isotherms for C8 PFCA binding onto the model PE MPs under elevated salinity and pH = 7. The points are the experimental data, and the lines represent the fits using Langmuir adsorption model. Error bars represent the standard deviation of the surface tension measurements obtained from the optical tensiometer, and corresponding values of the surface excess (described in methods section).



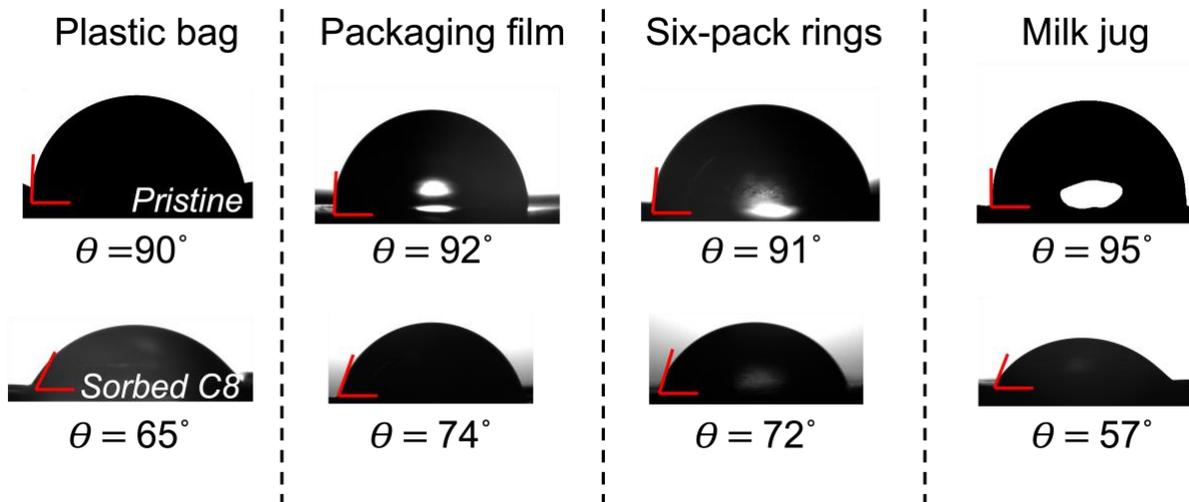
**Fig. S5.** Experimentally obtained ATR-FTIR spectra of pristine polyethylene (black), C10 PFCA (orange) and polyethylene with adsorbed C10 PFCA.



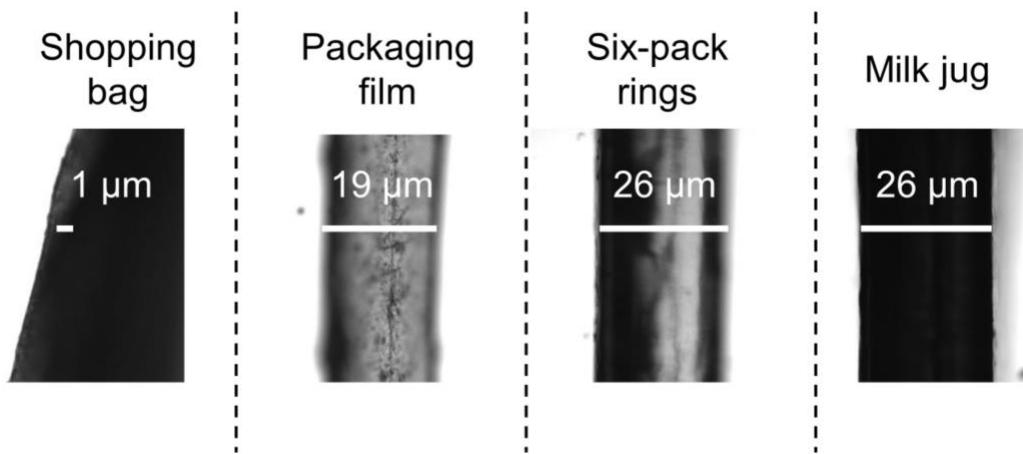
**Fig. S6** Change in dispersibility of model MPs upon the addition of PFCAs. The four vials contain approximately 50 mg of PE microplastics. Each vial contains a 5 mM solution of the respective PFCA solution as given in the image.



**Fig. S7** Surface excess values for each PFCA which remained adsorbed onto MPs after 10 days of equilibration in darkness compared to when exposed to UV irradiation. The error bars are the standard deviation of at least three repeat experiments.



**Fig. S8** Water contact angles of commodity plastics before and after C8 PFCA adsorption.  $\theta$  represents the experimentally measured contact angle of each sample. Images on the top row are the pristine commodity plastics and their experimentally obtained contact angles. The bottom row is after C8 PFCA is adsorbed on their surface.



**Fig. S9** Thickness of commodity plastics. Optical microscopy images taken of the four commodity plastics used in the experiments. The measurements obtained for each commodity plastic is as follows: Shopping bag  $< 1 \mu\text{m}$ , Packaging film =  $19 \mu\text{m}$ , six-pack rings =  $26 \mu\text{m}$  and milk jug =  $26 \mu\text{m}$