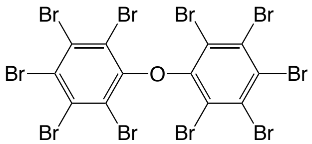
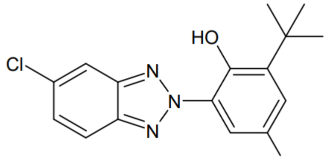
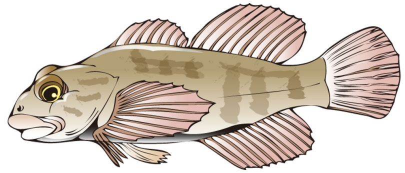
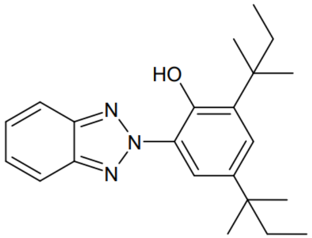
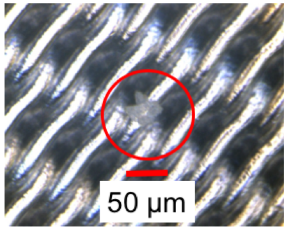
Plastic-mediated long-range transport of additives in marine environments and their bioaccumulation through plastic ingestion

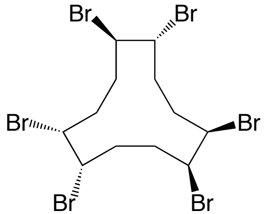


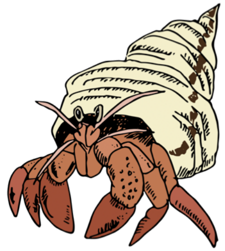












Hideshige TAKADA

Laboratory of Organic Geochemistry (LOG)
Tokyo University of Agriculture and Technology

International Pellet Watch

Presented on
“POPs in plastic”
April 24, 2023

IPCP International Panel on Chemical Pollution

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Mr. Taischi Takano
Ms. Natsuki Hirai
Ms. Rei Sakurai
Ms. Fumika Kashiwada

Volunteers for IPW
Volunteers for IPEN

Topics

Long-range transport of Benzotriazole-type UV stabilizers (BUVSs) via mm-size microplastics

Microplastic-mediated bioaccumulation of plastic additives

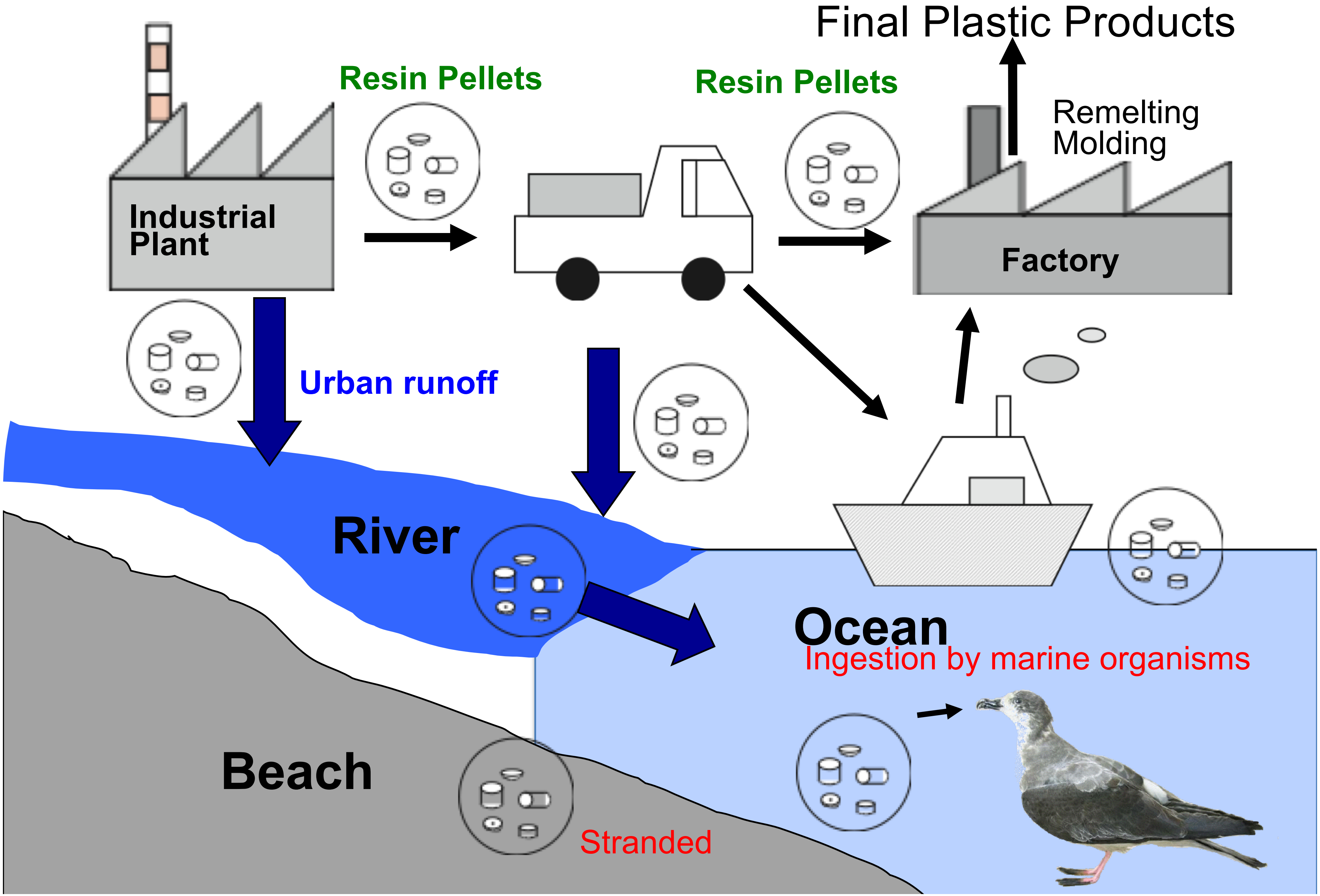
Topics

Long-range transport of Benzotriazole-type UV stabilizers (BUVSs) via mm-size microplastics

Microplastic-mediated bioaccumulation of plastic additives

Plastic resin pellets : feedstock of plastic products

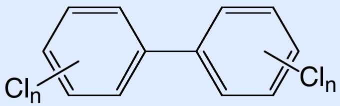
Resin pellets, industrial feedstock of user plastics, are spilled during transport and manufacturing and they are widely distributed in the ocean



Plastic resin pellets are stranded on beaches across the globe

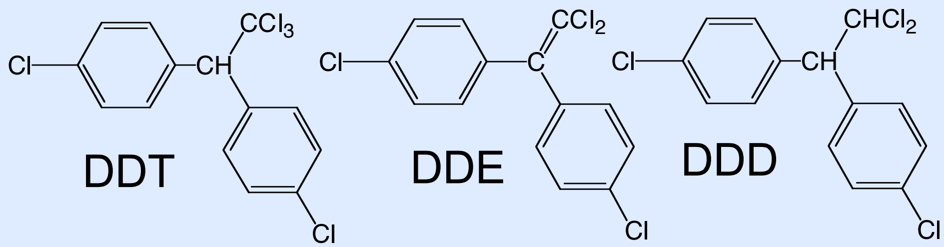
POPs are sorbed to plastic pellet from surrounding seawater

PCBs



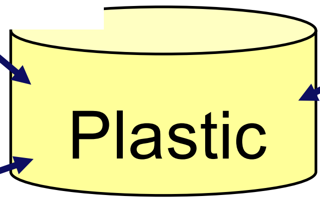
- Industrial products for a variety of uses including dielectric fluid, heat medium, and lubricants.
- Endocrine disrupting chemicals

DDTs

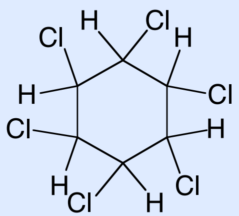


- DDT and its metabolites such as DDE and DDD.
- DDT was used as insecticides
- Endocrine disrupting chemicals

adsorption from ambient seawater



HCH



- Insecticide

$-\!(CH_2-CH_2)_n-$

Polyethylene (PE)

$-\!(CH_2-CH(CH_3))_n-$

Polyprorylene (PP)

Concentration factor is ~ 10^5 to ~10^6.

International Pellet Watch

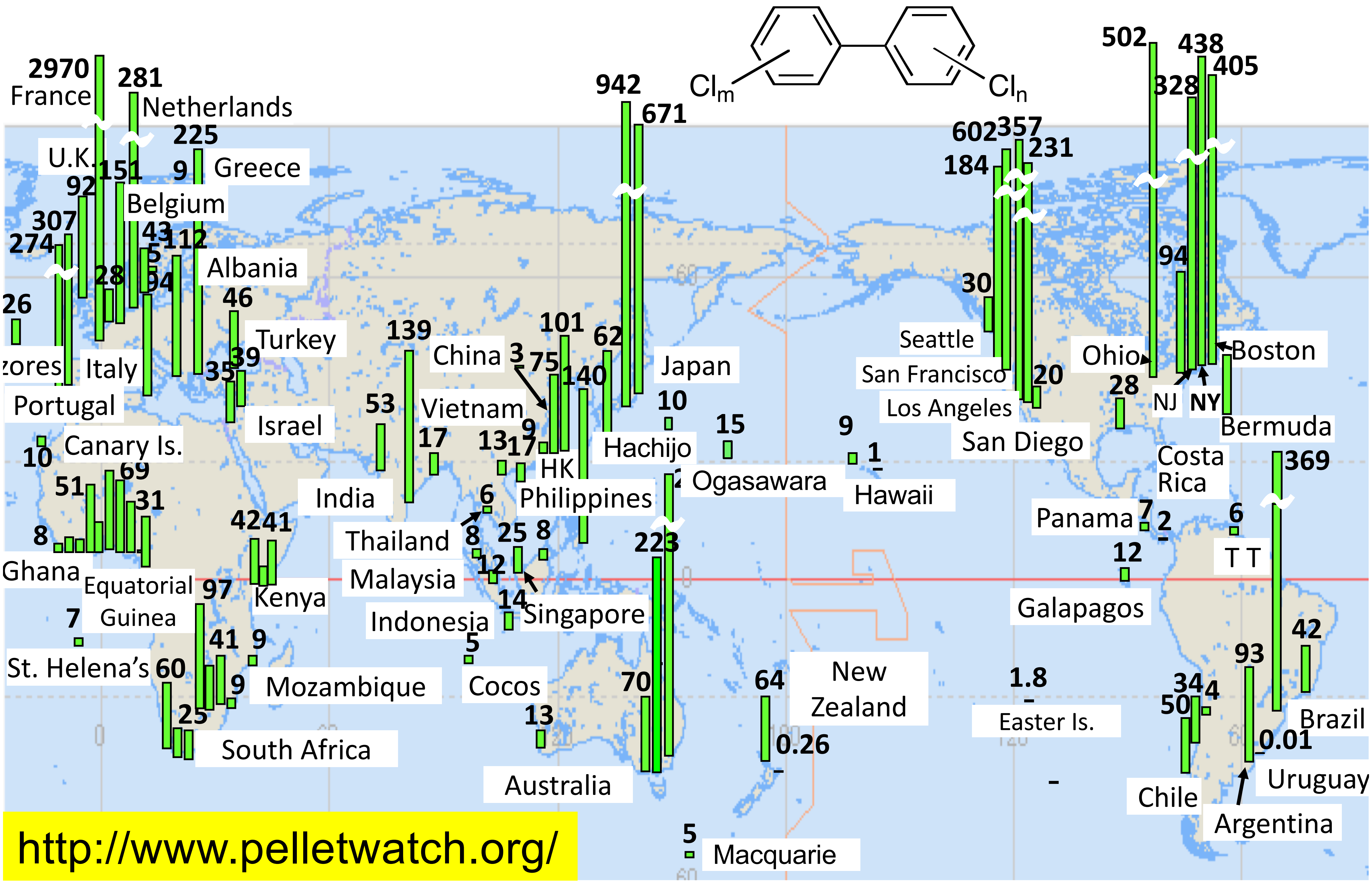
Call for pellets through website, journals, SNS.

since 2005

Laboratory of Organic Geochemistry (LOG)
Tokyo University of Agriculture and Technology, Japan

Plastic resin pellet from various areas in the world

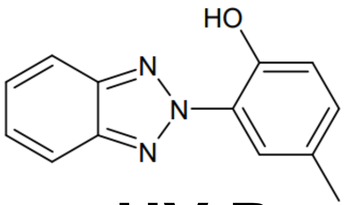
Monitoring of sorbed POPs



http://www.pelletwatch.org/

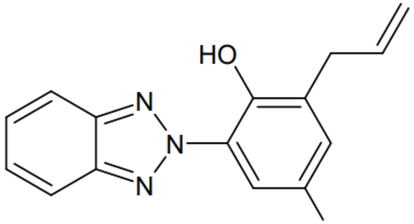
PCBs concentrations in beached plastic pellets (ng/g)

Benzotriazole-type UV stabilizers (BUVSs)



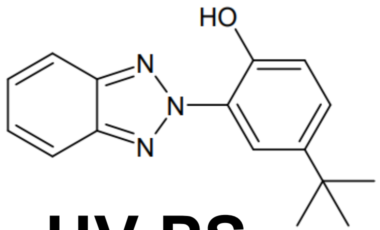
UV-P

logKow = 3.00



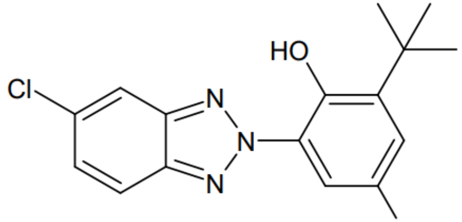
UV-9

logKow = 3.16



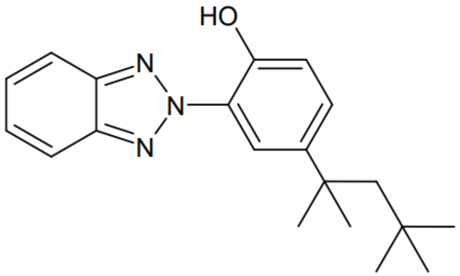
UV-PS

logKow = 4.36



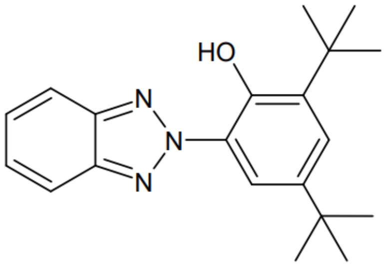
UV-326

logKow = 5.55



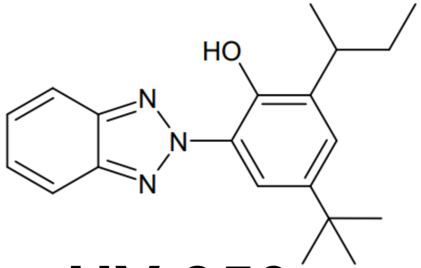
UV-329

logKow = 6.21



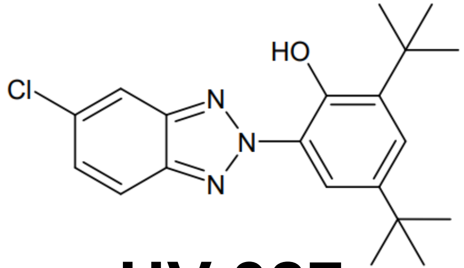
UV-320

logKow = 6.27



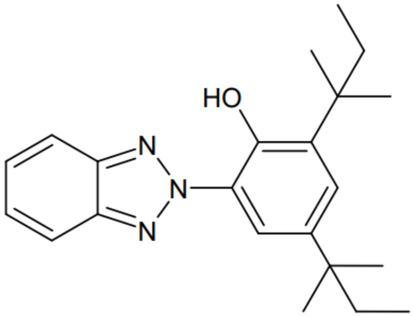
UV-350

logKow = 6.51



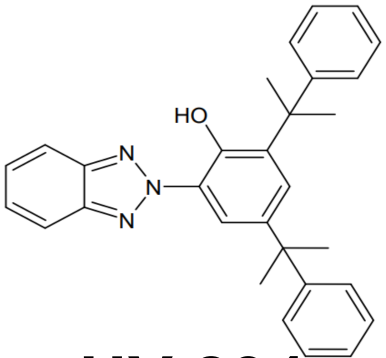
UV-327

logKow = 6.91



UV-328

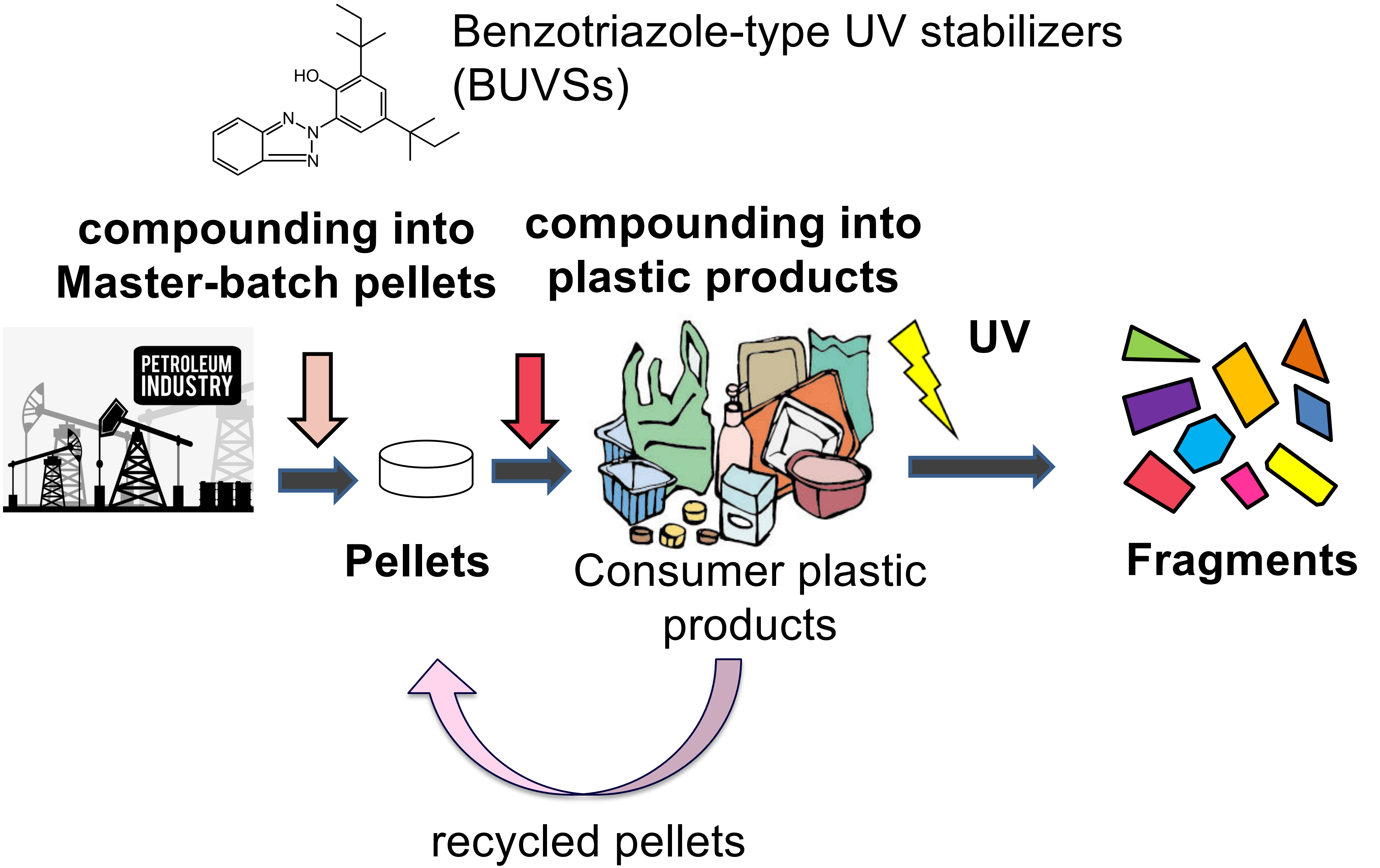
logKow = 7.25



UV-234

logKow = 7.67

UV stabilizers can be found in in plastic resin pellets : Master-batch pellets or recycled pellets



sorting

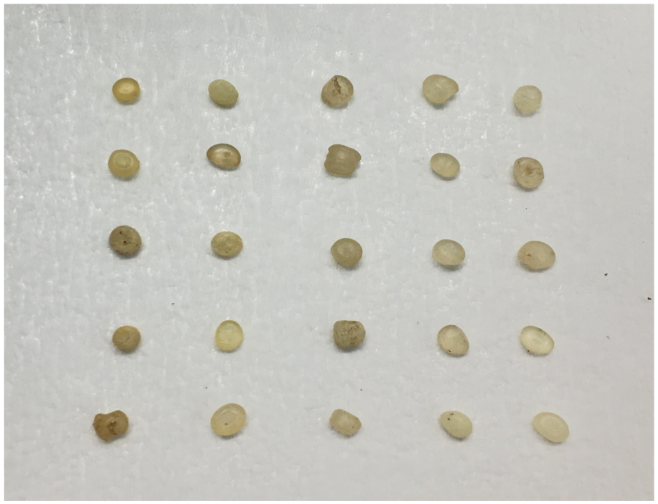
Polyethylene (PE)

Yellowing

More sorptive to POPs

sorting

Polypropylene (PP)



Less resistant to weathering

More UV-stabilizers required.

37 locations across the globe

Each sample contains 25 – 50 pellets

Analytical Procedure of BUVSs in plastic resin pellets

Pellets (25 - 50 pieces)

Soaking Extraction
Hexane 24 h x 3 times

surrogates
(^{13}C-BUVSs)

Acetylation
with acetic anhydride and pyridine

Silica gel column chromatography
10% H_2O deactivated

DCM/Hexane

DCM
(1% methanol)

GC-MS

Extremely High concentrations of BUVSs were found in pellets from 14 samples including remote islands among 37 samples

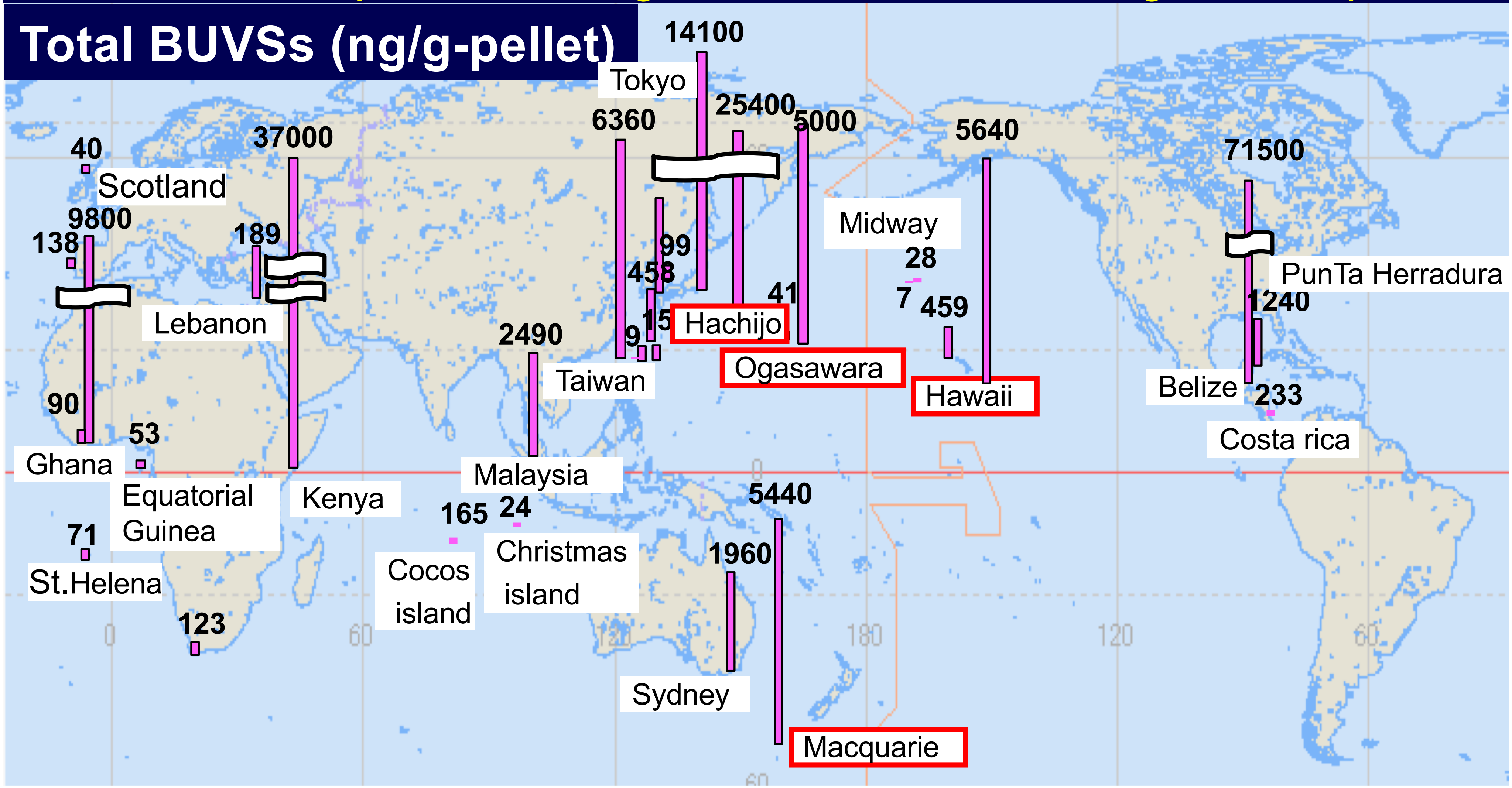
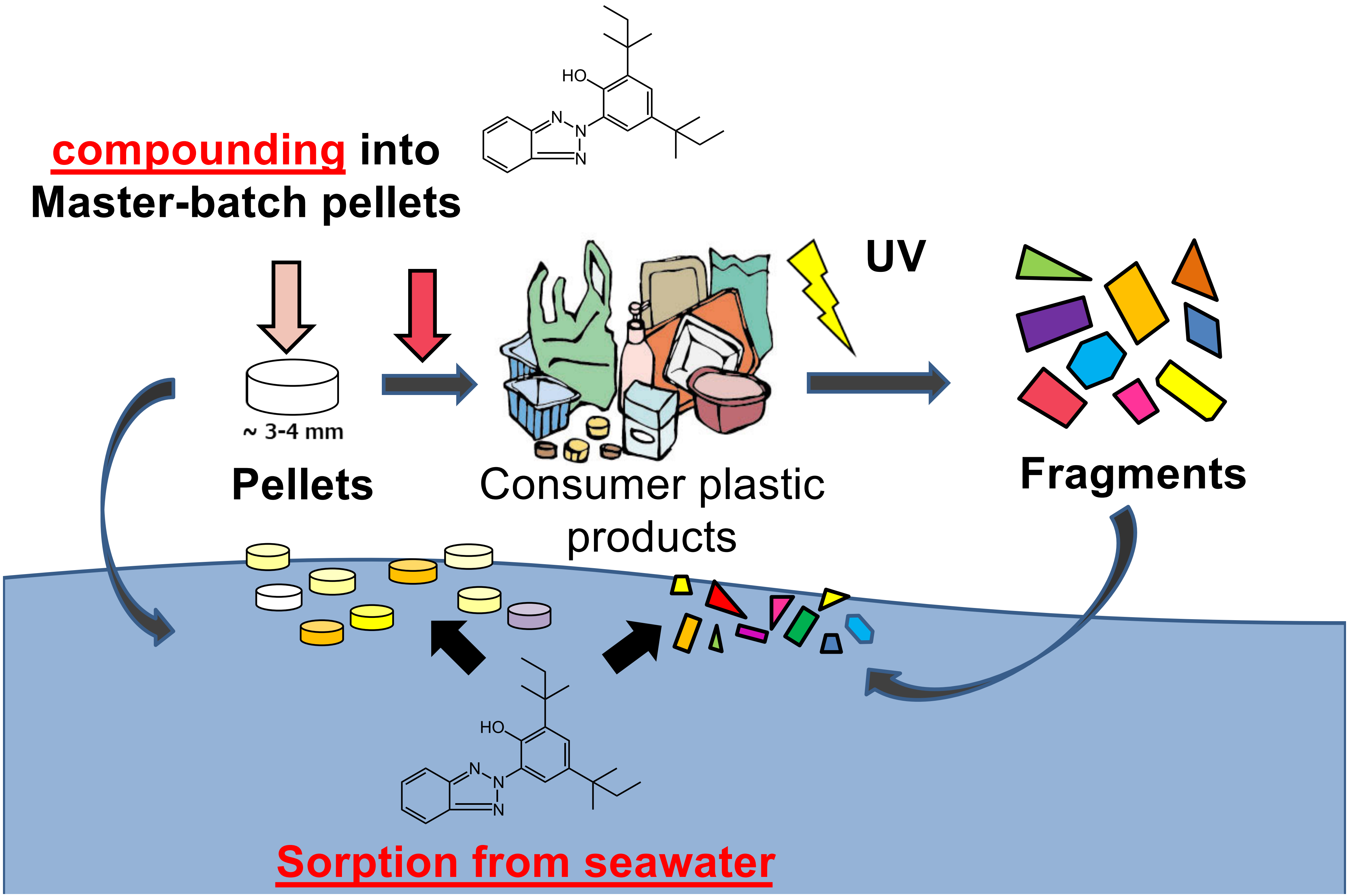


Fig.1. Concentration of total benzotriazole UV stabilizers (BUVSs) in polypropylene (PP) pellets on world beaches(ng/g-pellet)

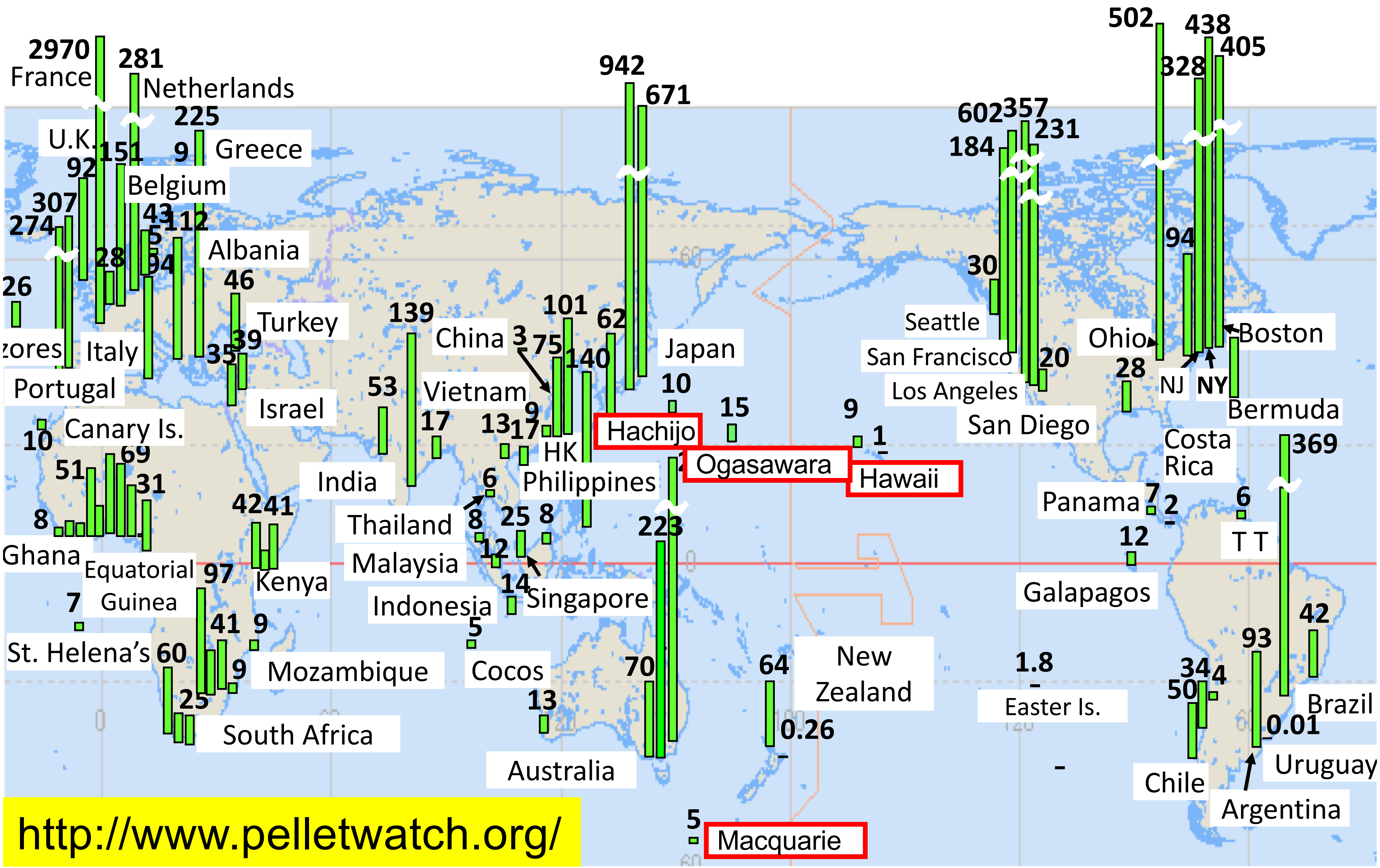
Total BUVSs : sum of UVP, UVPS, UV329, UV9, UV320, UV350, UV326, UV327, UV328, UV234.

Matsunaga et al. (2023)
to be presented on Annual meeting of Japan Society for Environmental Chemistry (JEC)

UV stabilizers can be originated via sorption form seawater in addition to master batch pellets and recycled pellets



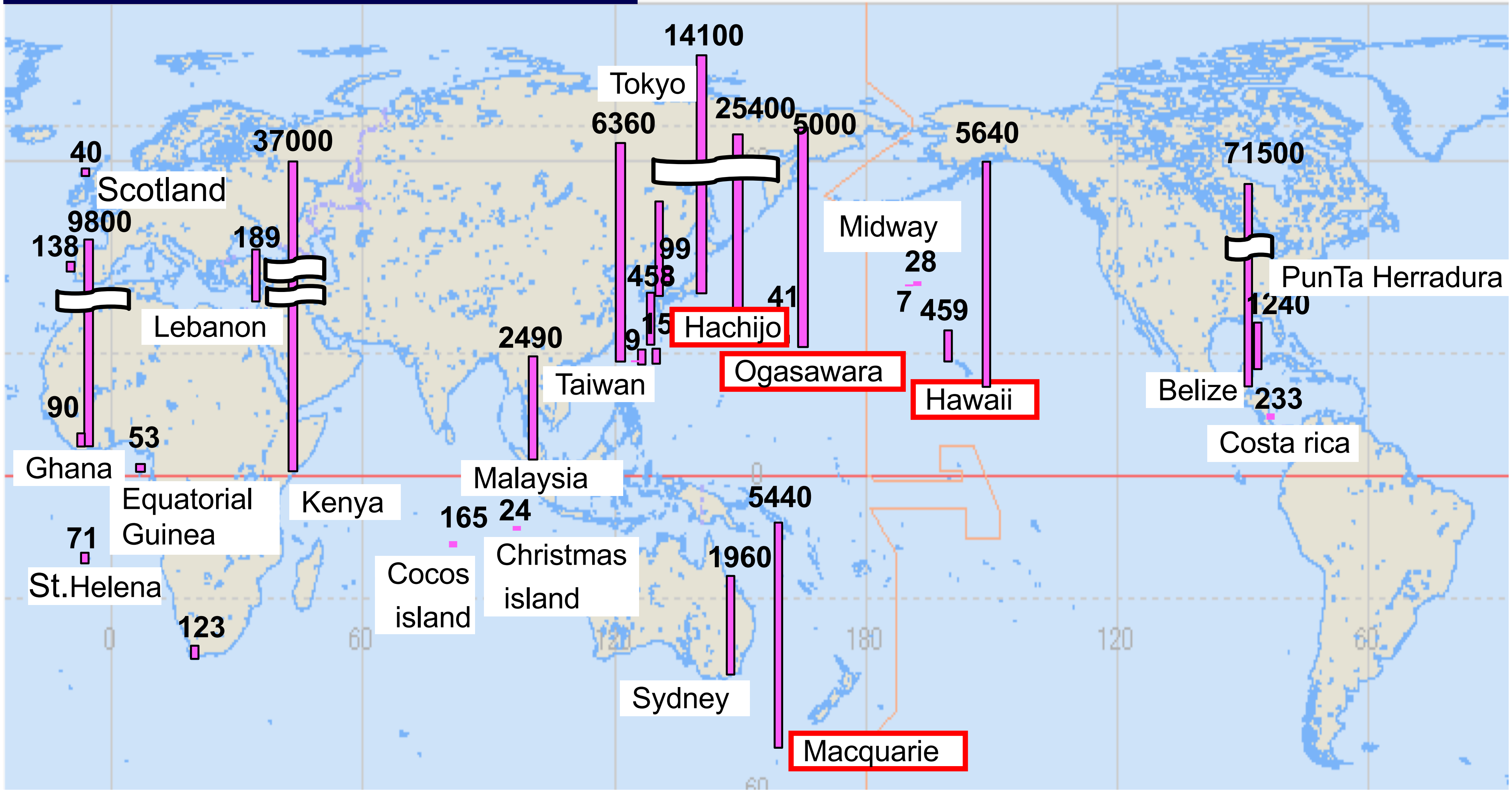
Trace level of PCBs were detected in pellets from remote islands



PCBs concentrations in beached plastic pellets (ng/g)

Plastic additives are long-range-transported across the borders

Total BUVSs (ng/g-pellet)



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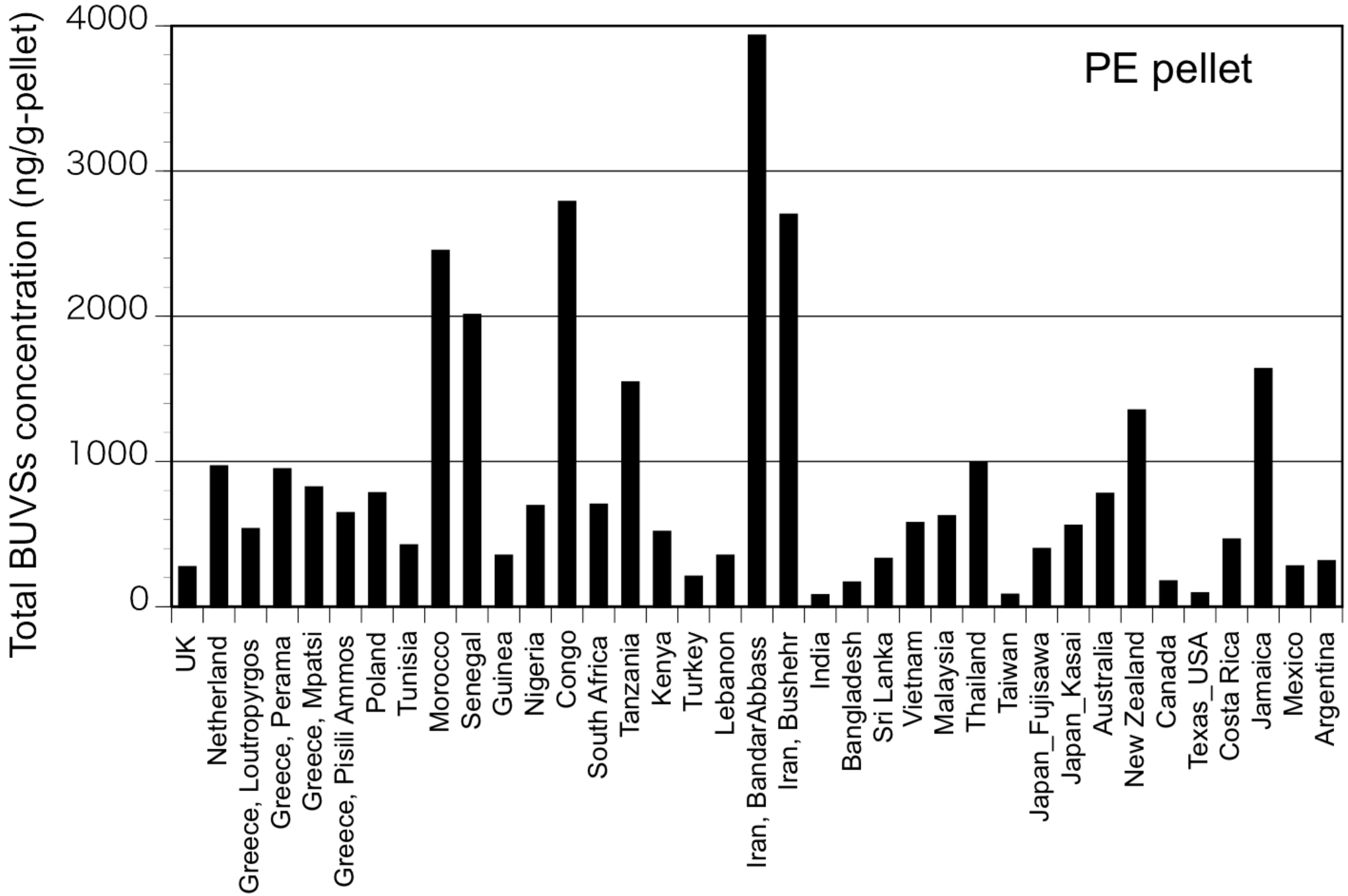


Fig.3. Total benzotriazole UV stabilizers (BUVSs) in polyethylene (PE) pellets.
Total BUVSs : sum of UVP, UVPS, UV329, UV9, UV320, UV350, UV326, UV327, UV328, UV234.

Karlsson et al. (2021)
IPEN report

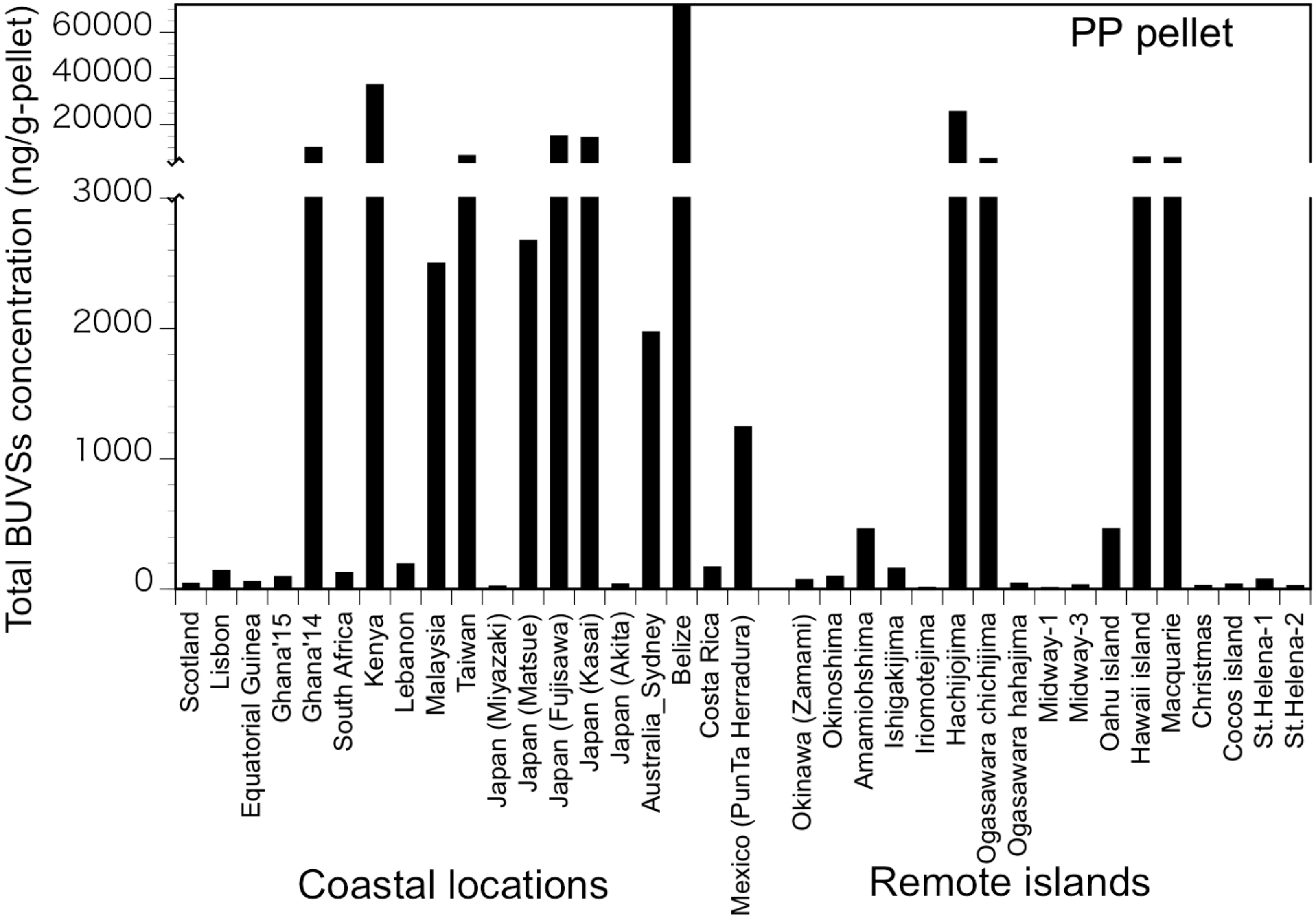


Fig.2. Total benzotriazole UV stabilizers (BUVSs) in polypropylene (PP) pellets.
Total BUVSs : sum of UVP, UVPS, UV329, UV9, UV320, UV350, UV326, UV327, UV328, UV234.

Matsunaga et al. (2023)
to be presented on Annual meeting of Japan Society for Environmental Chemistry (JEC)

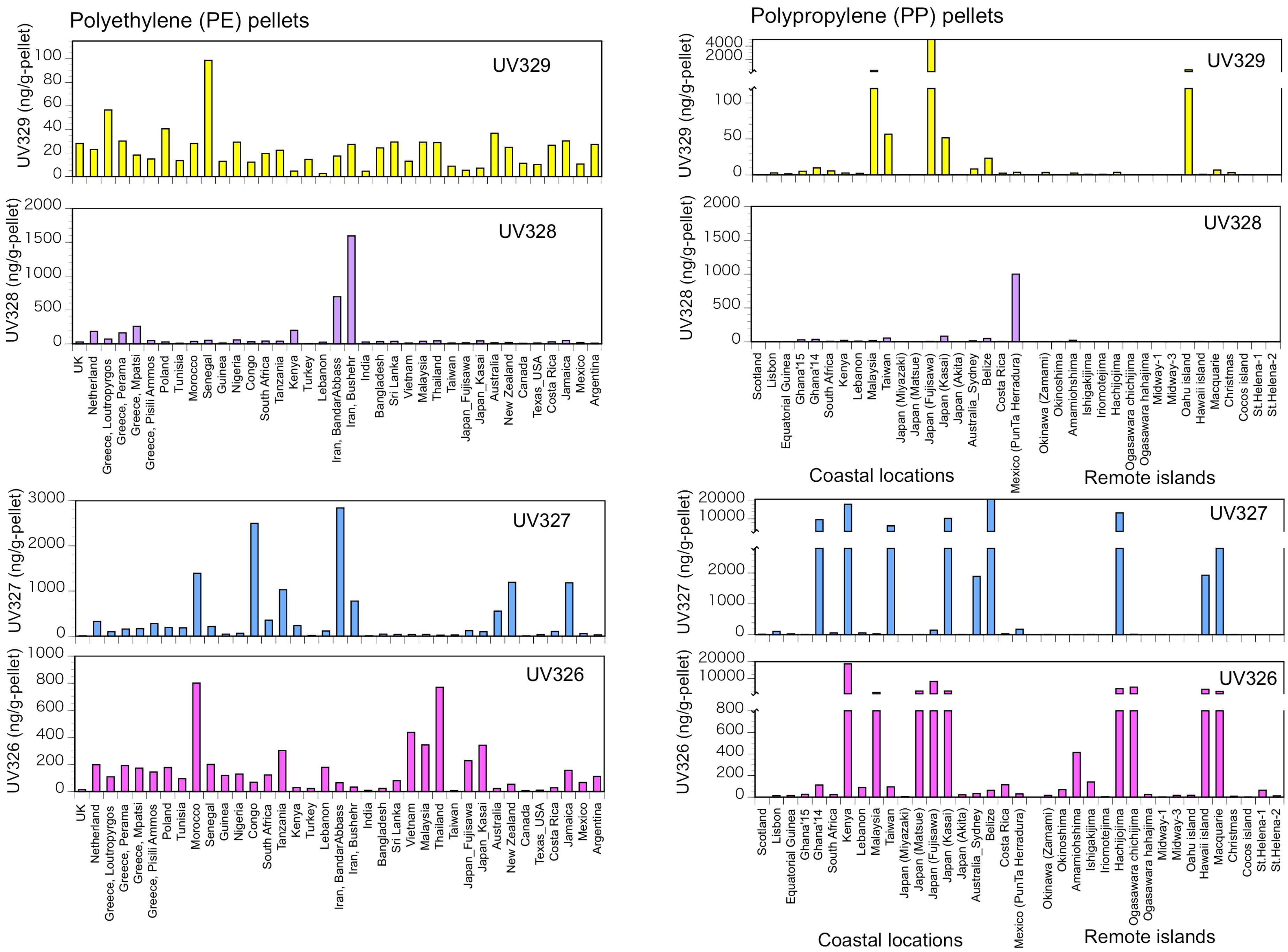


Fig.4. Concentrations of individual BUVSs in plastic resin pellets.
Left : Polyethylene (PE) pellets
Right : Polypropylene (PP) pellets.

PE data : Karlsson et al. (2021) IPEN report

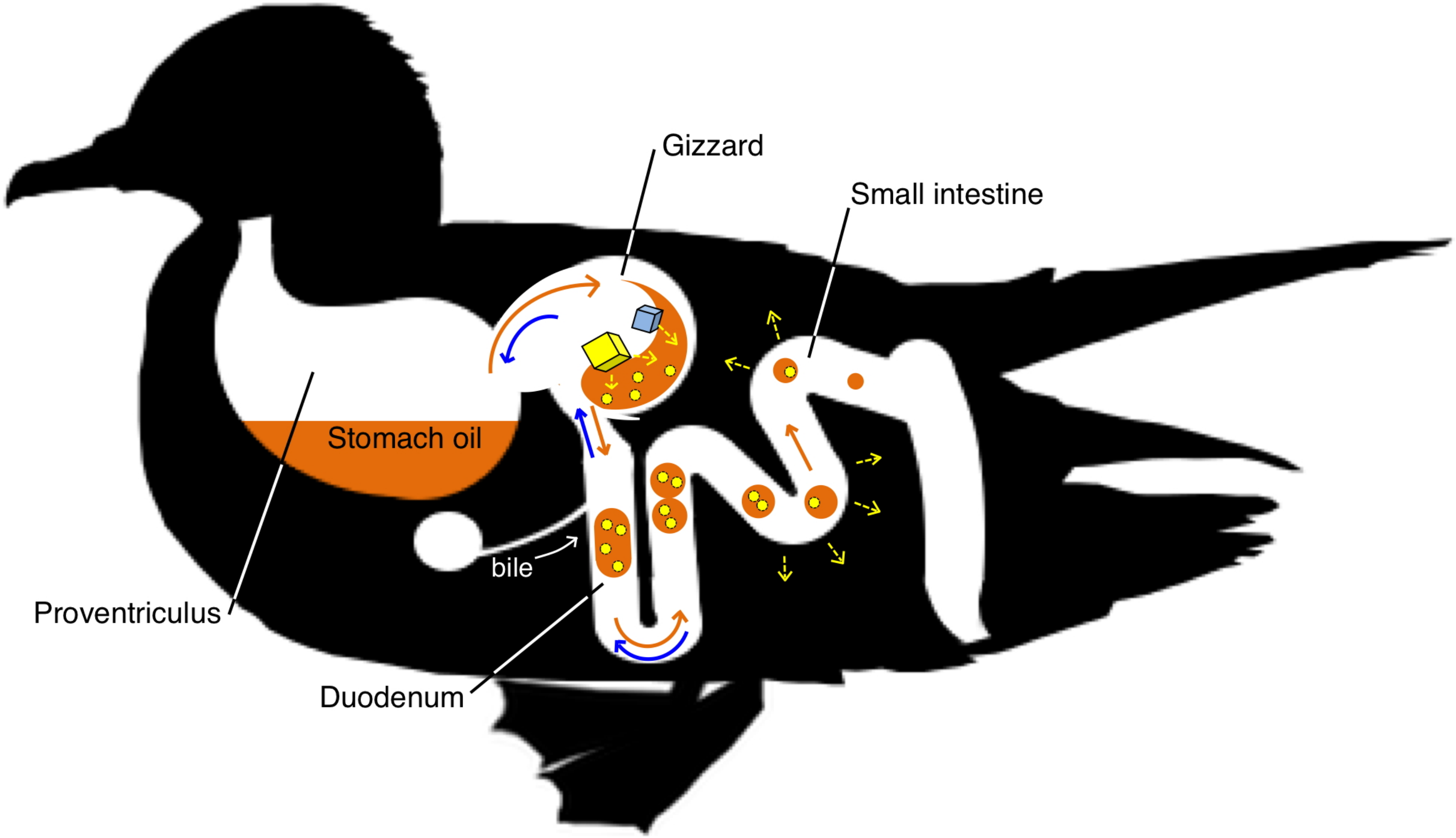
PP data : Matsunaga et al. (2023)

Topics

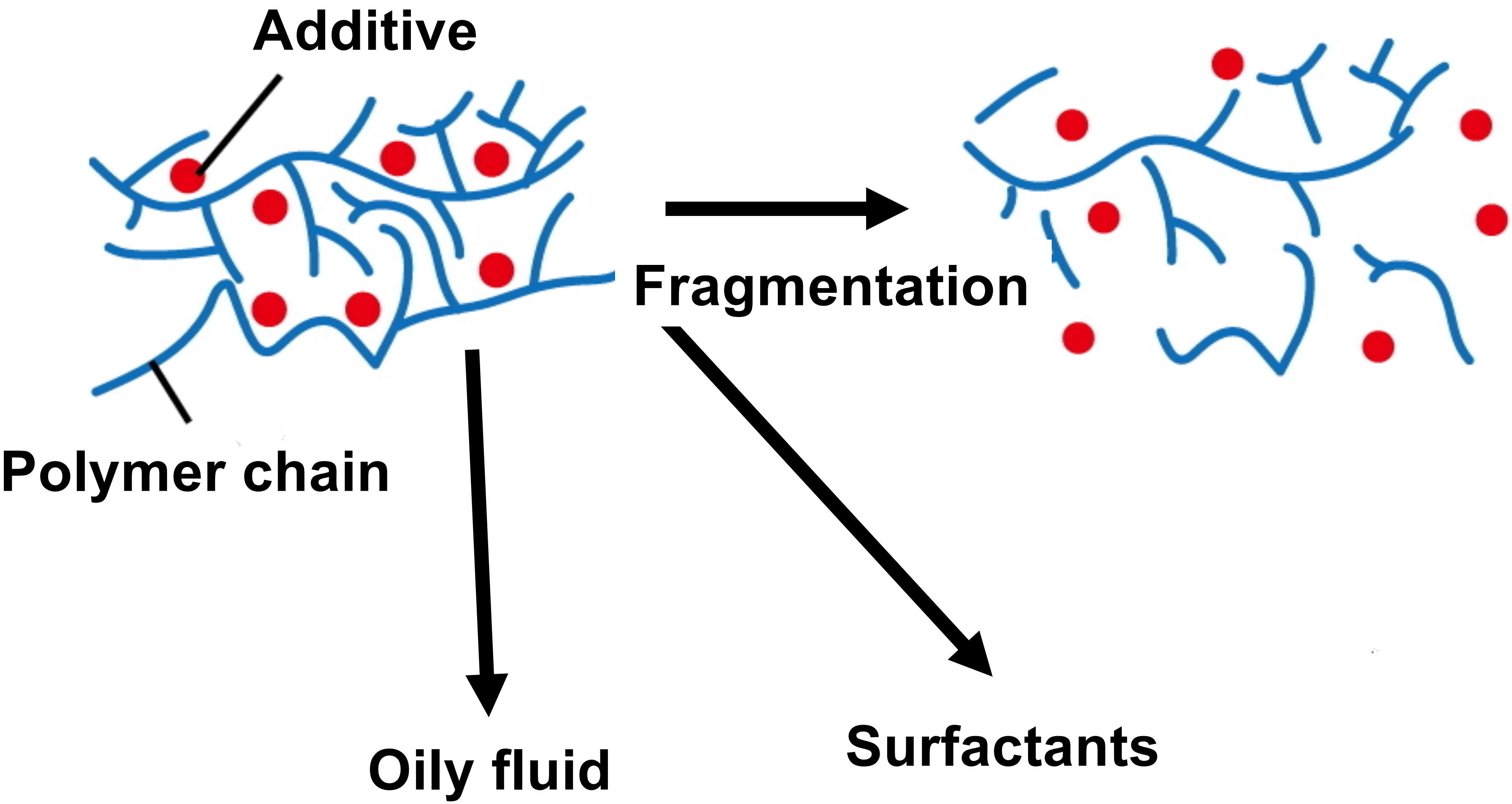
Long-range transport of Benzotriazole-type UV stabilizers (BUVSs) via mm-size microplastics

Microplastic-mediated bioaccumulation of plastic additives

Hazardous chemicals can be transferred and accumulated in tissue and organ of marine organisms which ingest plastics



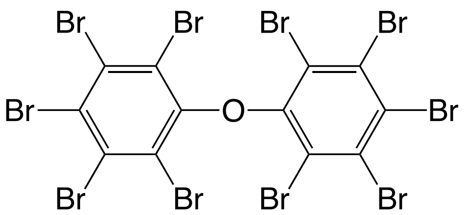
Tanaka, K., Yamashita, R., and Takada, H., *Transfer of hazardous chemicals from ingested plastics to higher-trophic level organisms,* in *Hazardous chemicals associated with plastics in environment,* H. Takada and H.K. Karapanagioti, Editor. 2018, Springer Berlin Heidelberg: p. 267–280.


Additive
Polymer chain
Fragmentation
Oily fluid
Surfactants

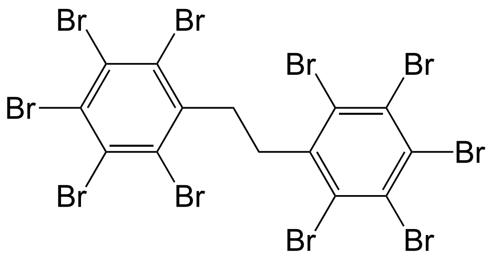
Fate of plastic additives in the ocean

Are the additives transferred and accumulated into biological tissue/organ, when plastics are ingested by marine organisms?

BFRs



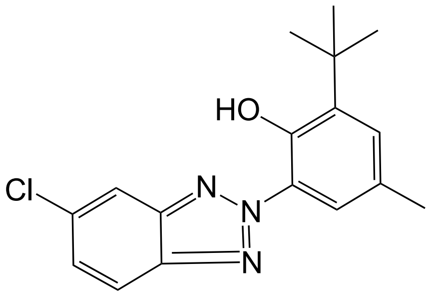
BDE209 (log K_{ow} : 12.11)



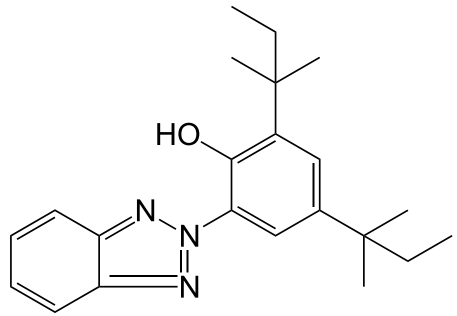
DBDPE (log K_{ow} : 13.64)

Highly hydrophobic

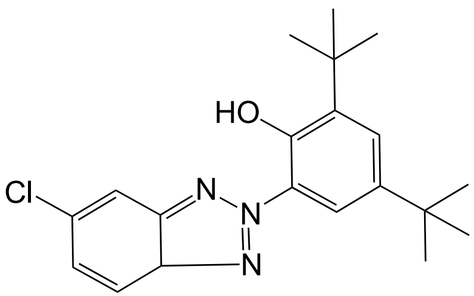
Benzotriazoles UV stabilizers



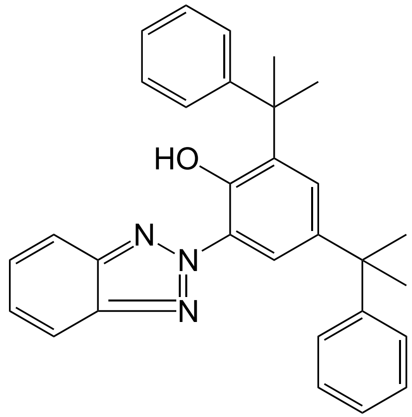
UV326 (log K_{ow} : 5.55)



UV328 (log K_{ow} : 7.25)

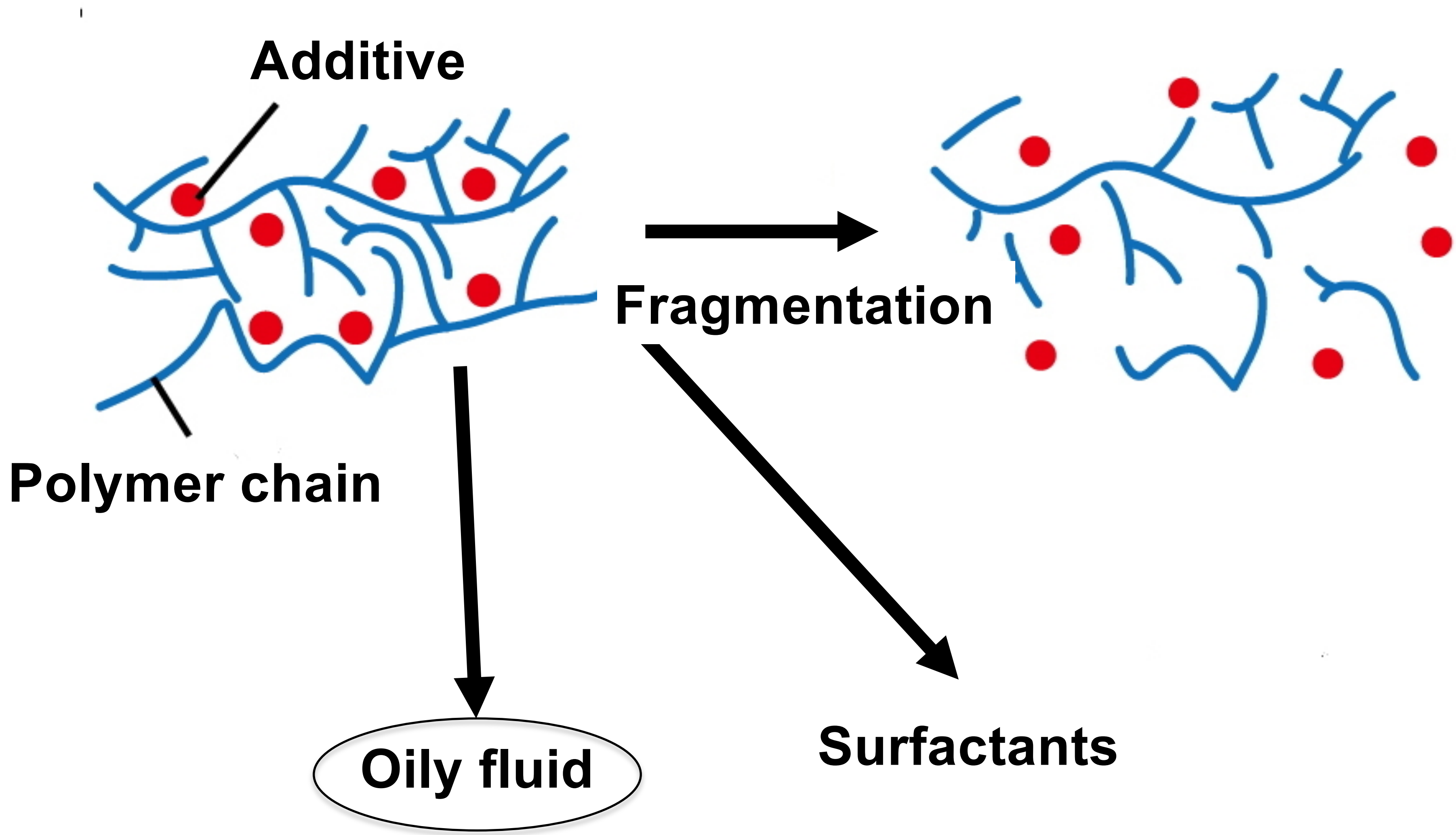


UV327 (log K_{ow} : 6.91)



UV234 (log K_{ow} : 7.67)

Moderately hydrophobic

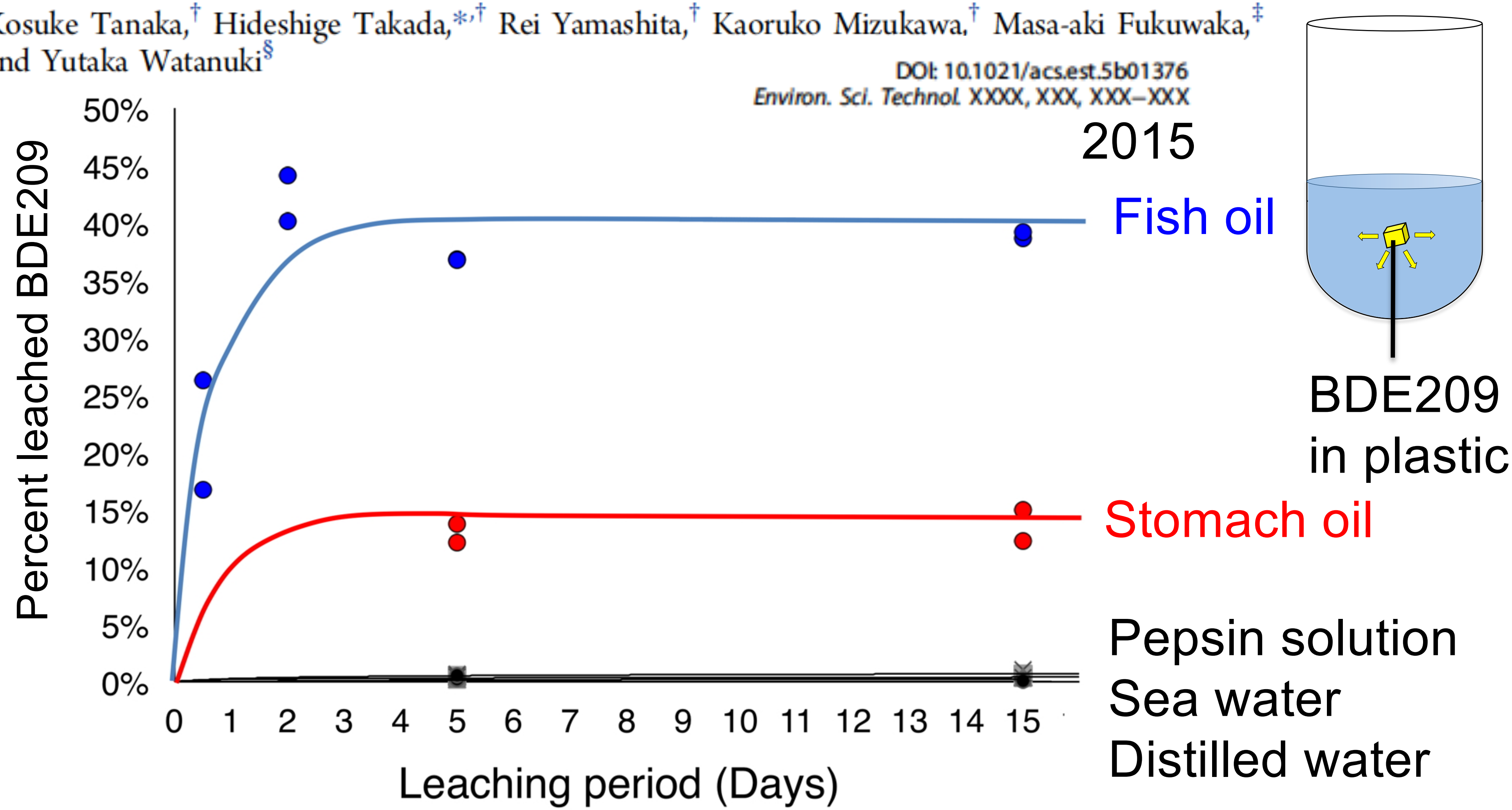

Additive
Polymer chain
Fragmentation
Oily fluid
Surfactants

Oily components in digestive tract facilitates leaching of hydrophobic additives

Facilitated Leaching of Additive-Derived PBDEs from Plastic by Seabirds' Stomach Oil and Accumulation in Tissues

Kosuke Tanaka,[†] Hideshige Takada,[*,†] Rei Yamashita,[†] Kaoruko Mizukawa,[†] Masa-aki Fukuwaka,[‡] and Yutaka Watanuki[§]

DOI: 10.1021/acs.est.5b01376
Environ. Sci. Technol. XXXX, XXX, XXX–XXX



Oily components in digestive fluid facilitate the leaching of additives from plastics

Environmental Science & Technology

Article

pubs.acs.org/est

Facilitated Leaching of Additive-Derived PBDEs from Plastic by Seabirds' Stomach Oil and Accumulation in Tissues

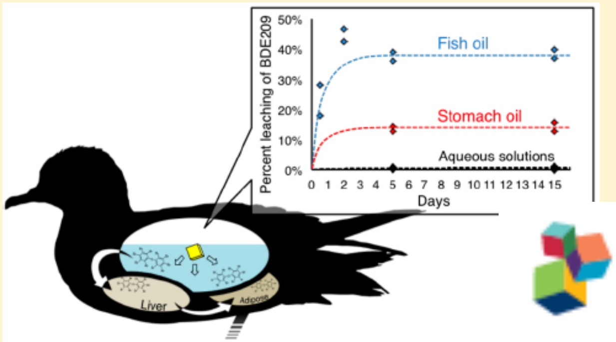
Kosuke Tanaka,[†] Hideshige Takada,[*,†] Rei Yamashita,[†] Kaoruko Mizukawa,[†] Masa-aki Fukuwaka,[‡] and Yutaka Watanuki[§]

[†]Laboratory of Organic Geochemistry, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183-8509, Japan
[‡]Hokkaido National Fisheries Research Institute, Fisheries Research Agency, Kushiro, Hokkaido 085-0802, Japan
[§]Faculty of Fisheries, Hokkaido University, Hakodate, Hokkaido 041-8611, Japan

2015

Supporting Information

ABSTRACT: Our previous study suggested the transfer of polybrominated diphenyl ether (PBDE) flame retardants from ingested plastics to seabirds' tissues. To understand how the PBDEs are transferred, we studied leaching from plastics into digestive fluids. We hypothesized that stomach oil, which is present in the digestive tract of birds in the order Procellariiformes, acts as an organic solvent, facilitating the leaching of hydrophobic chemicals. Pieces of plastic compounded with deca-BDE were soaked in several leaching solutions. Trace amounts were leached into distilled water, seawater, and acidic pepsin solution. In contrast, over 20 times as much material was leached into stomach oil, and over 50 times as much into fish oil (a major component of stomach oil). Analysis of abdominal adipose, liver tissue, and ingested plastics from 18 wild seabirds collected from the North Pacific Ocean showed the occurrence c tissues and the ingested plastics in three of the birds, suggesting transfer from the plastic their tissues, the dominance of BDE207 over other nona-BDE isomers suggested biologi Model calculation of PBDE exposure to birds based on the results of the leaching experir suggested the dominance of plastic-mediated internal exposure to BDE209 over exposu



frontiers in Environmental Science

Transfer of Additive Chemicals From Marine Plastic Debris to the Stomach Oil of Northern Fulmars

Susanne Kühn[1], Andy M. Booth[2], Lisbet Sørensen[2], Albert van Oyen[3] and Jan A. van Franeker[1]*

[1] Wageningen Marine Research, Den Helder, Netherlands, [2] Department (Ocean, Trondheim, Norway, [3] Carat GmbH, Bocholt, Germany

ORIGINAL RESEARCH
published: 19 August 2020
doi: 10.3389/fenvs.2020.00138

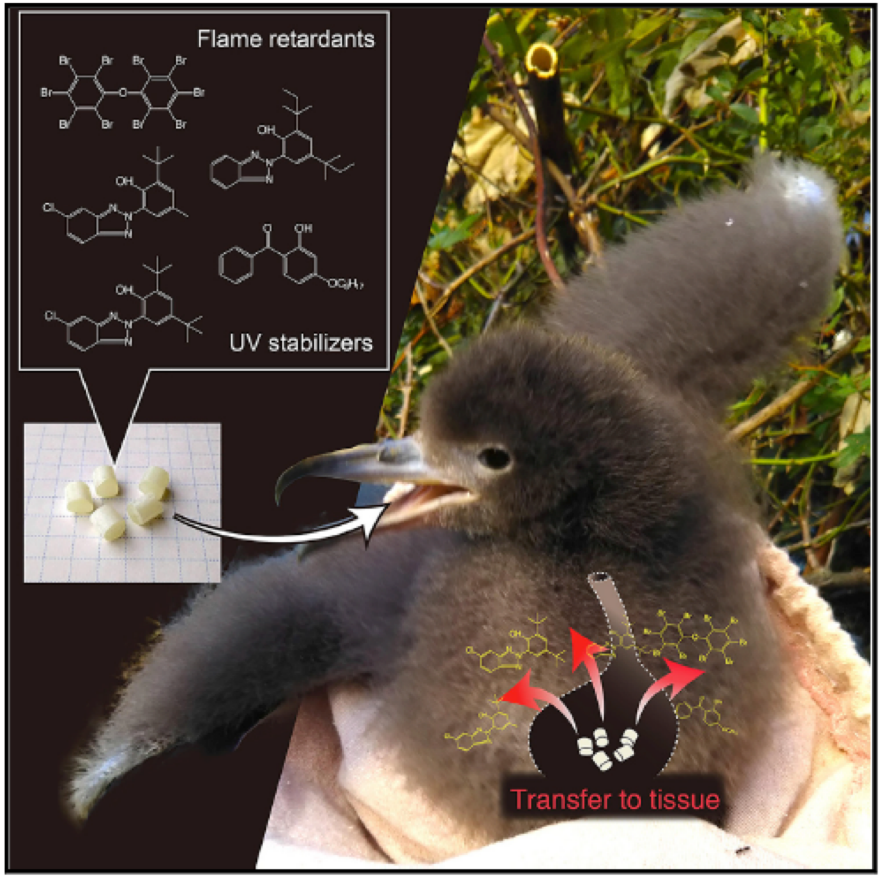
Feeding experiment confirmed the transfer of additives from ingested plastics to the organs of seabirds

Current Biology

Report

In Vivo Accumulation of Plastic-Derived Chemicals into Seabird Tissues

Graphical Abstract



Authors

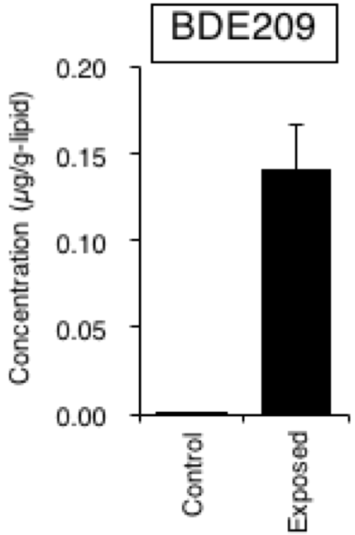
Kosuke Tanaka, Yutaka Watanuki, Hideshige Takada, ..., Michelle Hester, Yoshinori Ikenaka, Shouta M.M. Nakayama

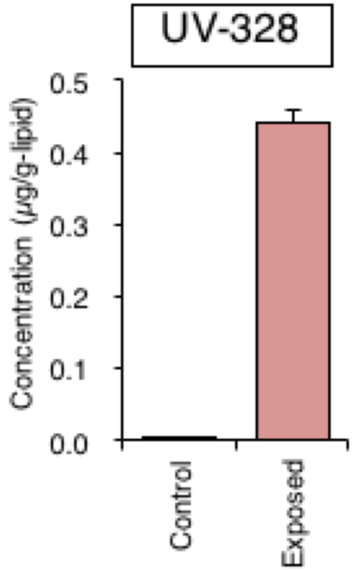
Correspondence

shige@cc.tuat.ac.jp

In Brief

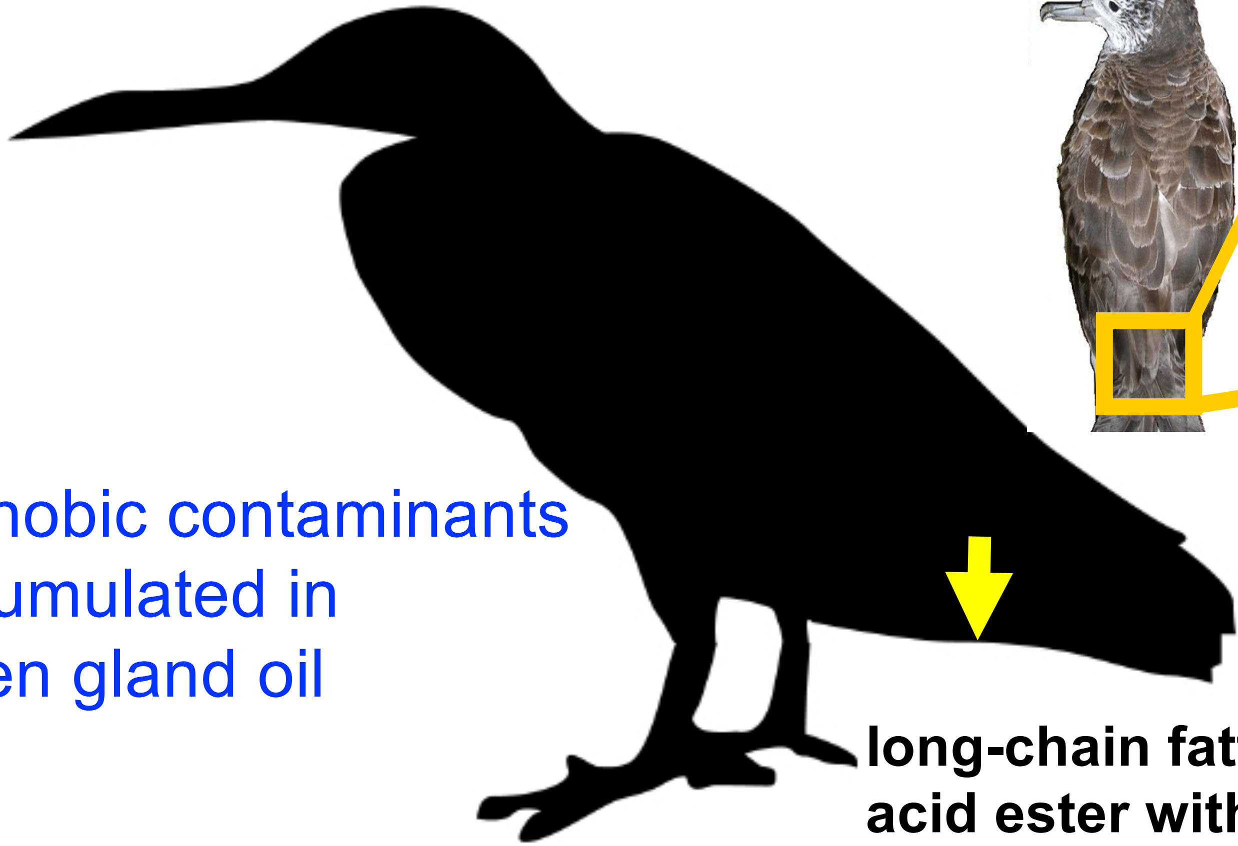
Tanaka et al. show that feeding additive-laced plastic pellets to seabirds results in the accumulation of chemical additives in liver and adipose tissue at 10^1–10^5 times above baseline. These findings demonstrate seabird exposure to plastic additives and additives' importance as emerging pollution sources.





Tanaka et al., 2020, Current Biology *30*, 1–6
February 24, 2020
https://doi.org/10.1016/j.cub.2019.12.037

Preen gland oil : oil excreted from an organ situated at the tail of birds



Hydrophobic contaminants are accumulated in the preen gland oil

long-chain fatty acid ester with higher alcohol

water-proof property to feathers of seabirds

Non-invasive approach

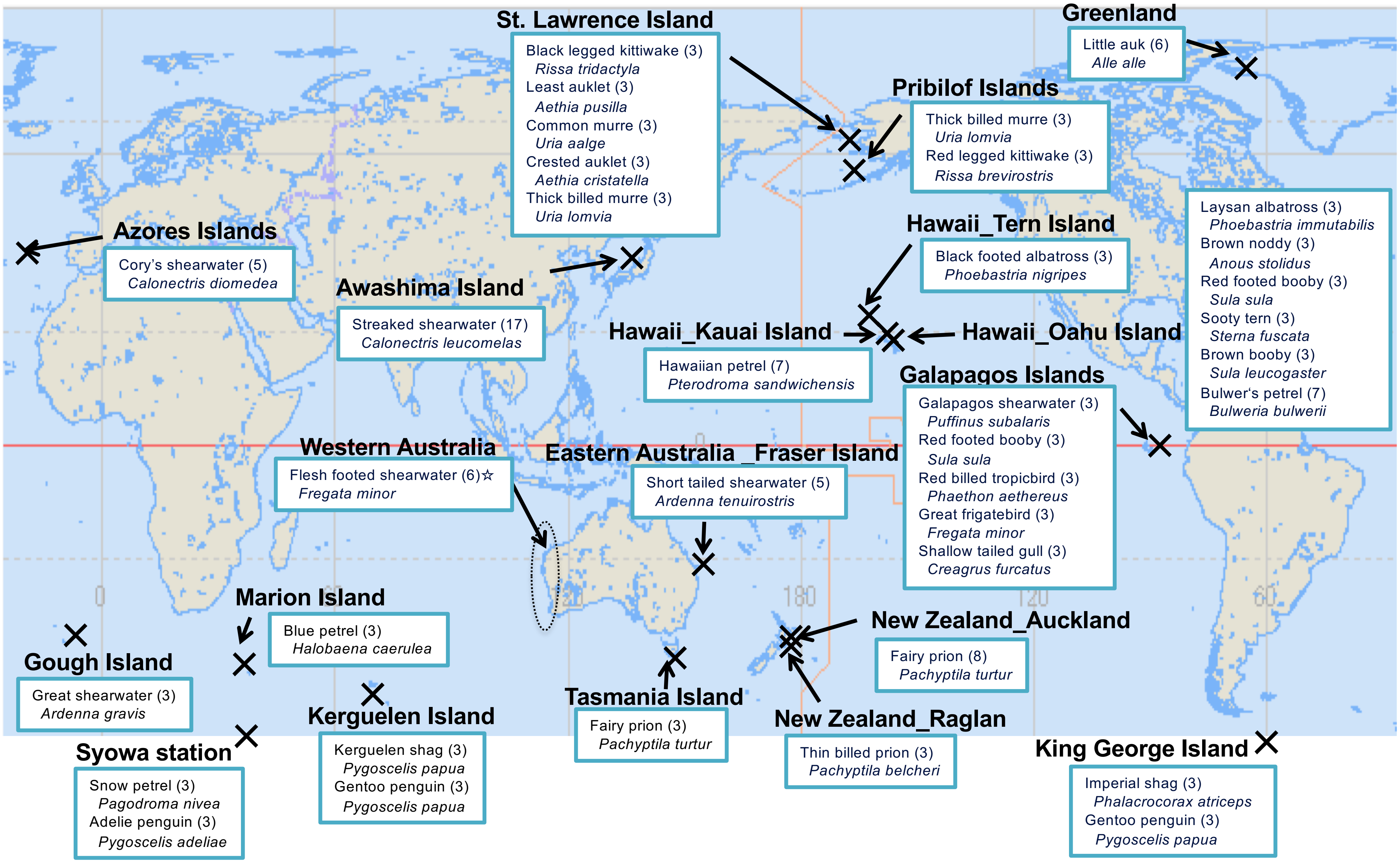
DOI: https://doi.org/10.5985/emcr.20210009
Environmental Monitoring and Contaminants Research Vol.1, pp.97–112, 2021
https://emcr-journal.org/
Article

Plastic additives and legacy persistent organic pollutants in the preen gland oil of seabirds sampled across the globe

Rei YAMASHITA[1,2], Nagako HIKI[1], Fumika KASHIWADA[1], Hideshige TAKADA[1]*, Kaoruko MIZUKAWA[1], Britta Denise HARDESTY[3,4], Lauren ROMAN[3,5], David HYRENBACH[6], Peter G. RYAN[7], Ben J. DILLEY[7], Juan Pablo MUÑOZ-PÉREZ[8,9], Carlos A. VALLE[8], Christopher K. PHAM[10], João FRIAS[11], Bungo NISHIZAWA[12], Akinori TAKAHASHI[12], Jean-Baptiste THIEBOT[12], Alexis WILL[13], Nobuo KOKUBUN[12], Yuuki Y. WATANABE[12], Takashi YAMAMOTO[14,15], Kozue SHIOMI[12,16], Ui SHIMABUKURO[17] and Yutaka WATANUKI[18]

Wide species of seabirds globally collected

145 individuals of 31 species from 16 locations in the world



Benzotriazole UV stabilizers were detected in preen gland oil from ~ 40 % of seabirds globally collected

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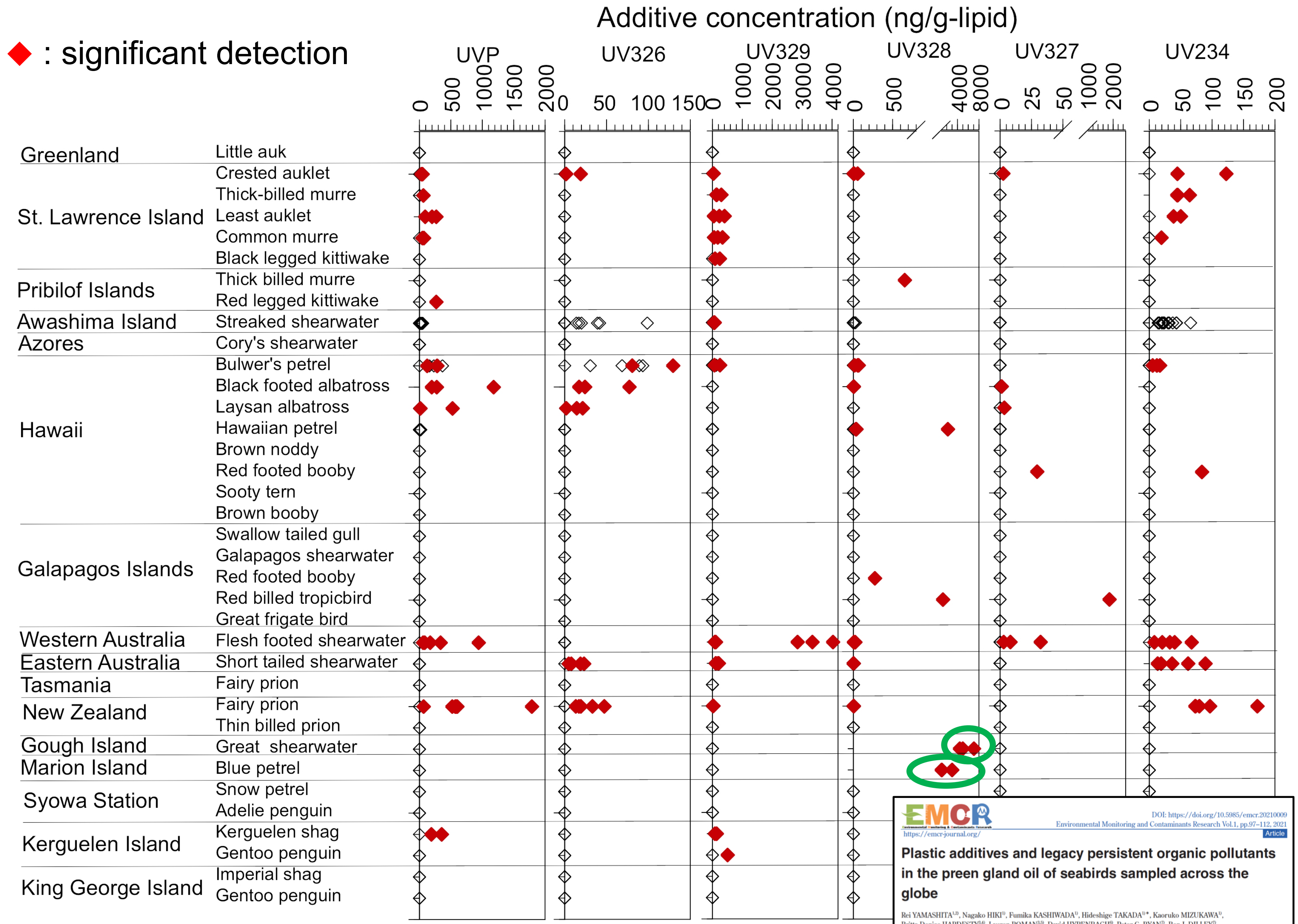
Extremely high concentrations of UV stabilizer were detected in preen gland oil from seabirds from remote islands



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Field observations demonstrated the bioaccumulation of plastic additives in seabirds preen gland oil (46 % of individuals examined)

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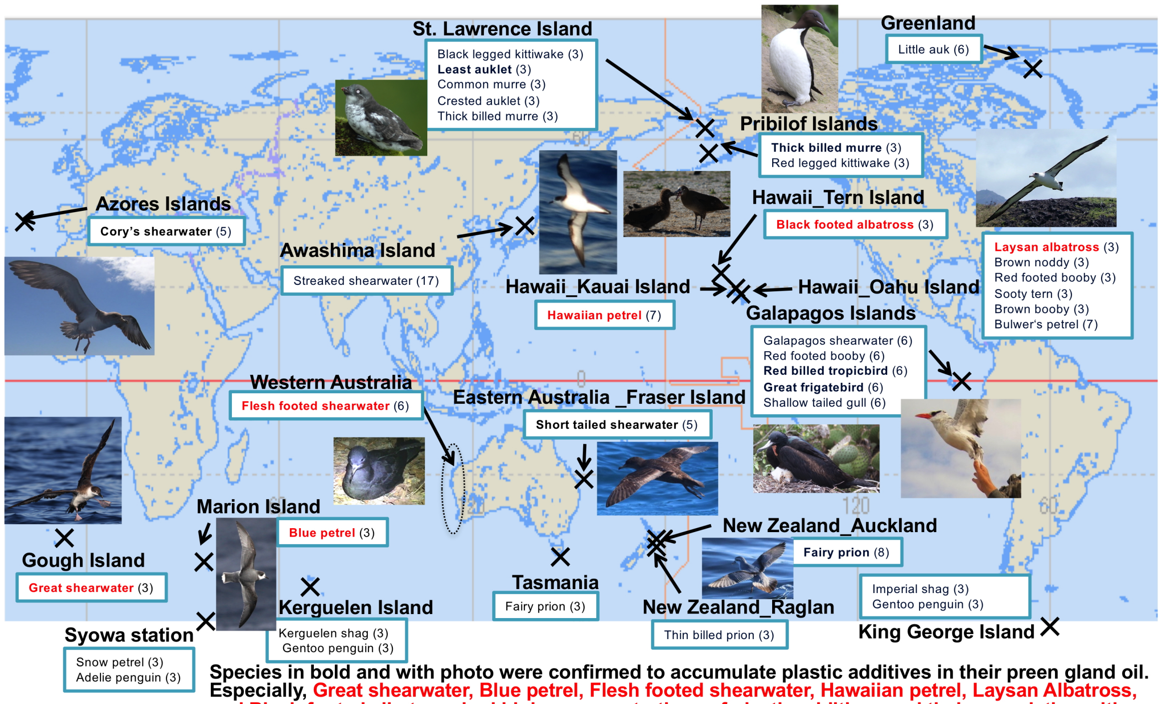
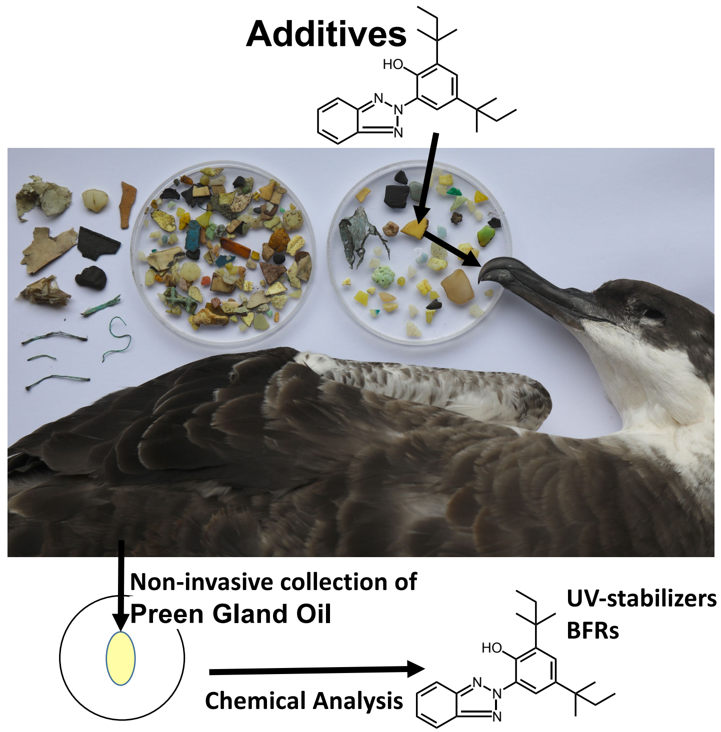
World seabirds accumulating plastic additives



Species in bold and with photo were confirmed to accumulate plastic additives in their preen gland oil. Especially, Great shearwater, Blue petrel, Flesh footed shearwater, Hawaiian petrel, Laysan Albatross, and Black-footed albatross had higher concentrations of plastic additives and their association with plastic ingestion were indicated.

All Photos by
David Hyrenbach (Hawaiian petrel, Black-footed albatrosses, Laysan albatrosses), Juan Pablo Muñoz-Pérez (Red billed tropicbird), Christopher K. Pham (Cory's shearwater), Lauren Roman (Flesh footed shearwater, Short tailed shearwater, Fairy prion), Peter G. Ryan (Great shearwater, Blue petrel), Akinori Takahashi (Least Auklet), Carlos A. Valle (Great frigate bird), Takashi Yamamoto (Thick-billed murre).



Bioaccumulation and metabolism of polybrominated diphenyl ethers (PBDEs) in coenobitid hermit crabs from marine litter-polluted beaches in remote islands

Nana Tanaka[a], Naohiko Takada[a], Mami Takahashi[a], Bee Geok Yeo[a], Yuki Oya[b], Izumi Watanabe[b], Yoshihisa Fujita[c], Hideshige Takada[a], Kaoruko Mizukawa[a,*]

[a] Laboratory of Organic Geochemistry, Tokyo University of Agriculture and Technology, 3-5-8, Saiwaicho, Fuchu, Tokyo 183-8509, Japan
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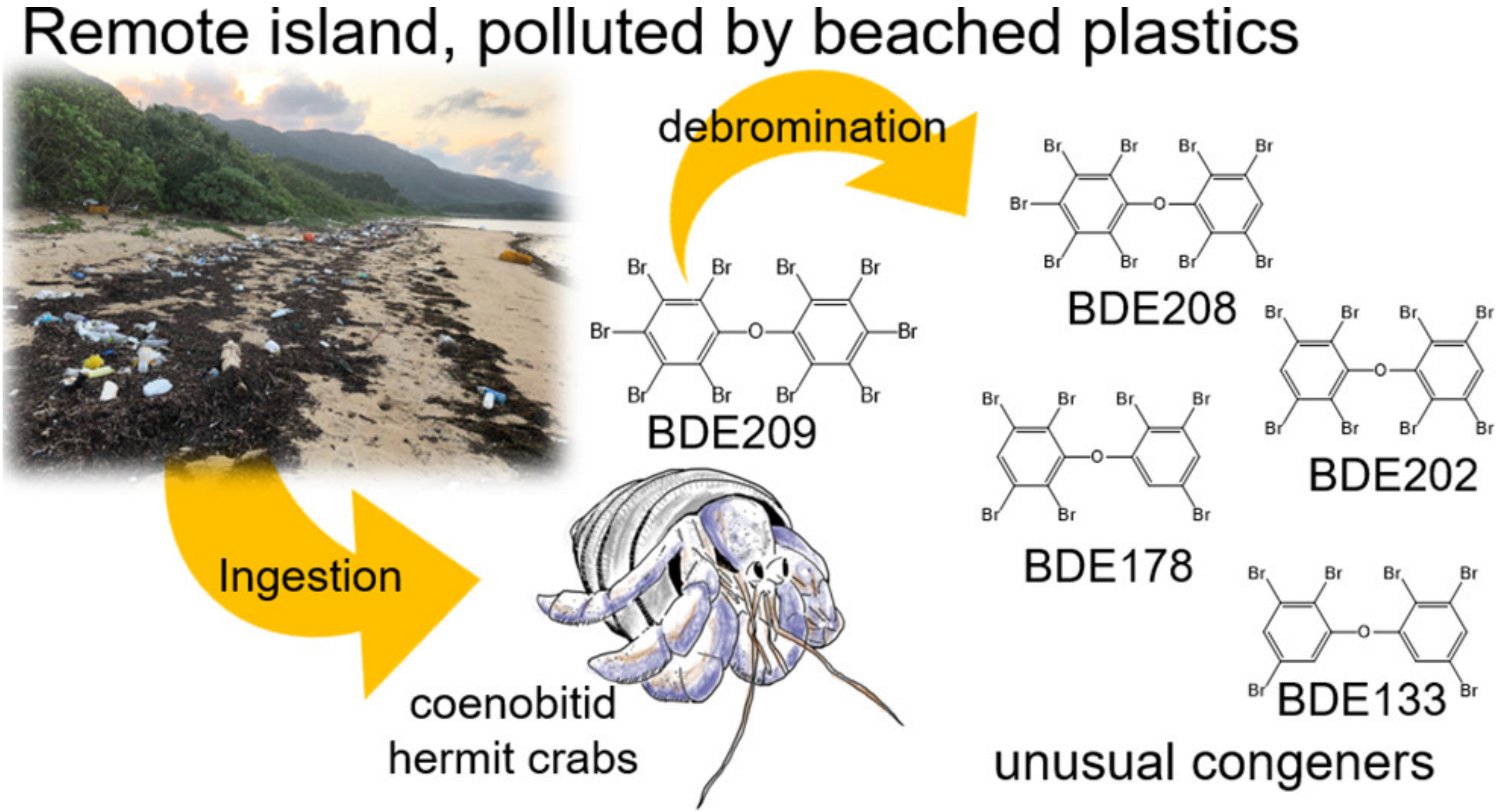
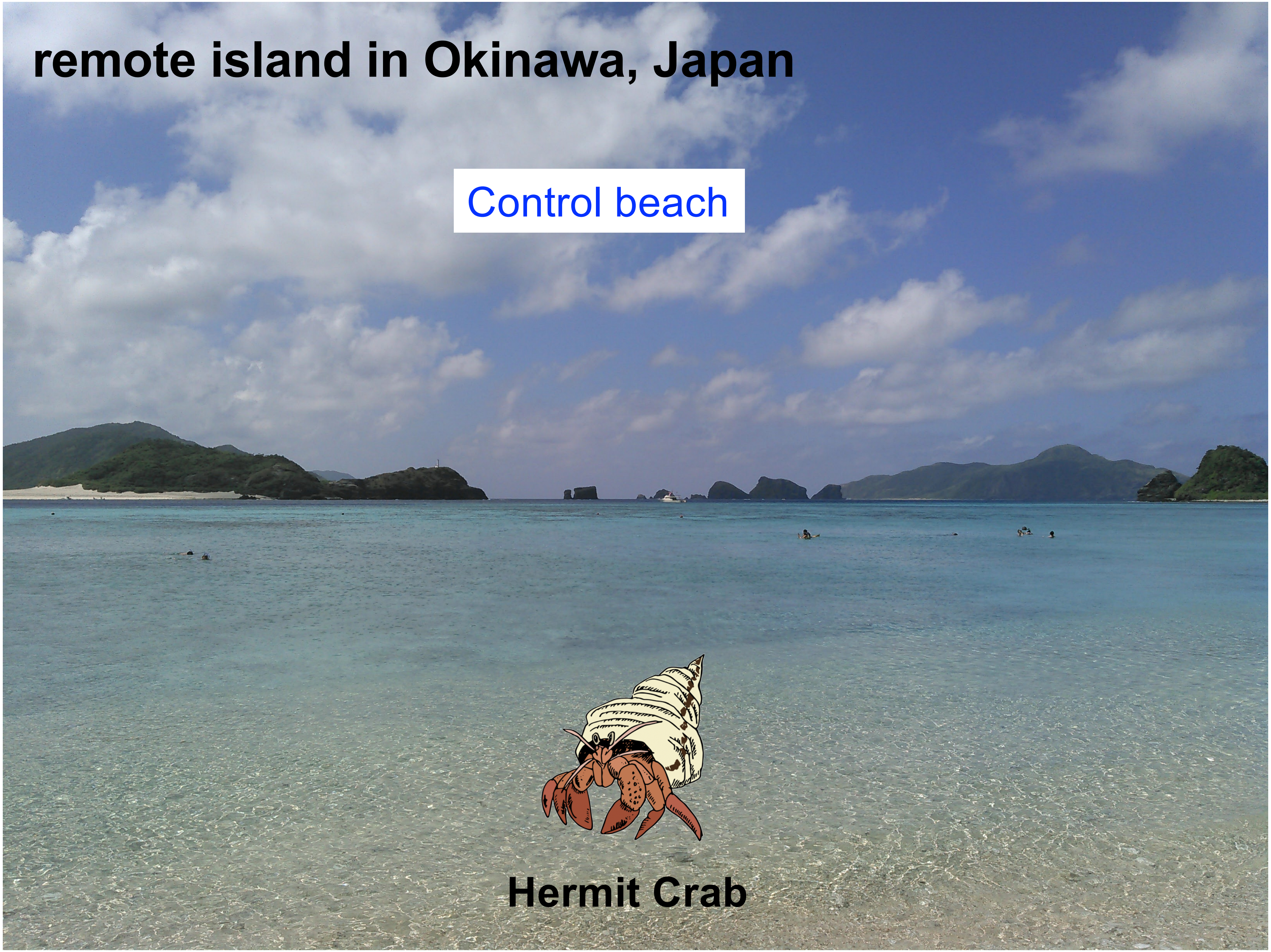
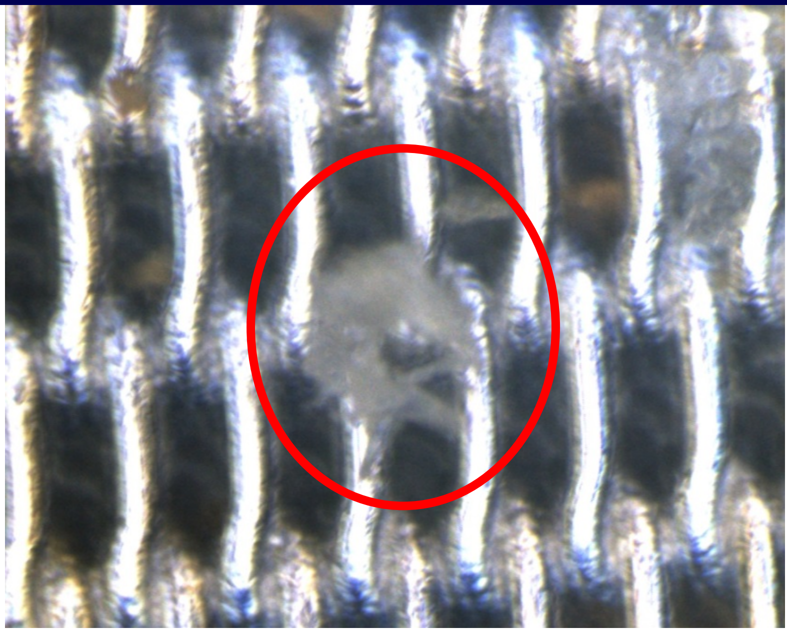
[c] Okinawa Prefectural University of Arts, 1-4, Shuri-Tounokura, Naha, Okinawa 903-8602, Japan

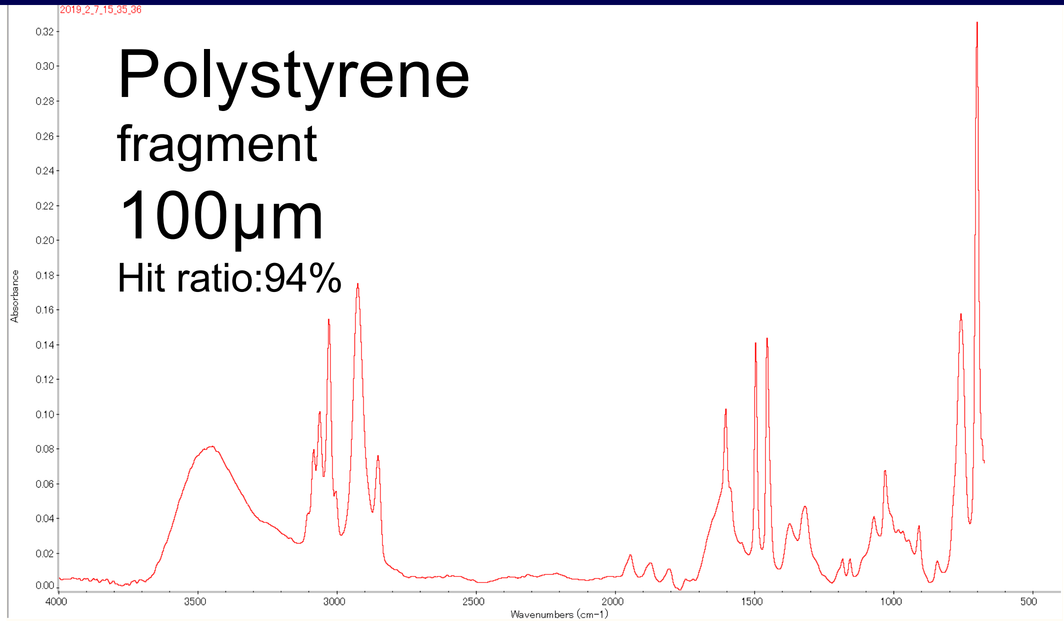


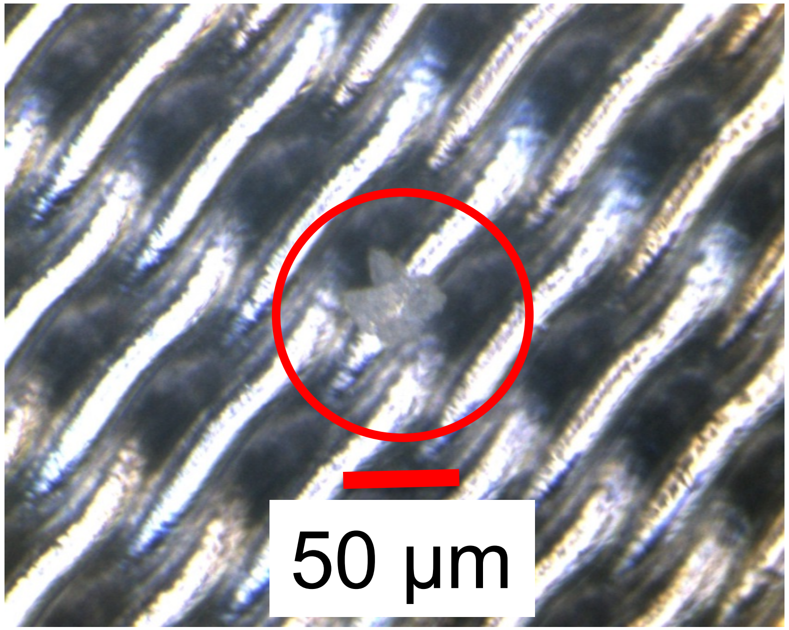

remote island in Okinawa, Japan
Control beach
Hermit Crab

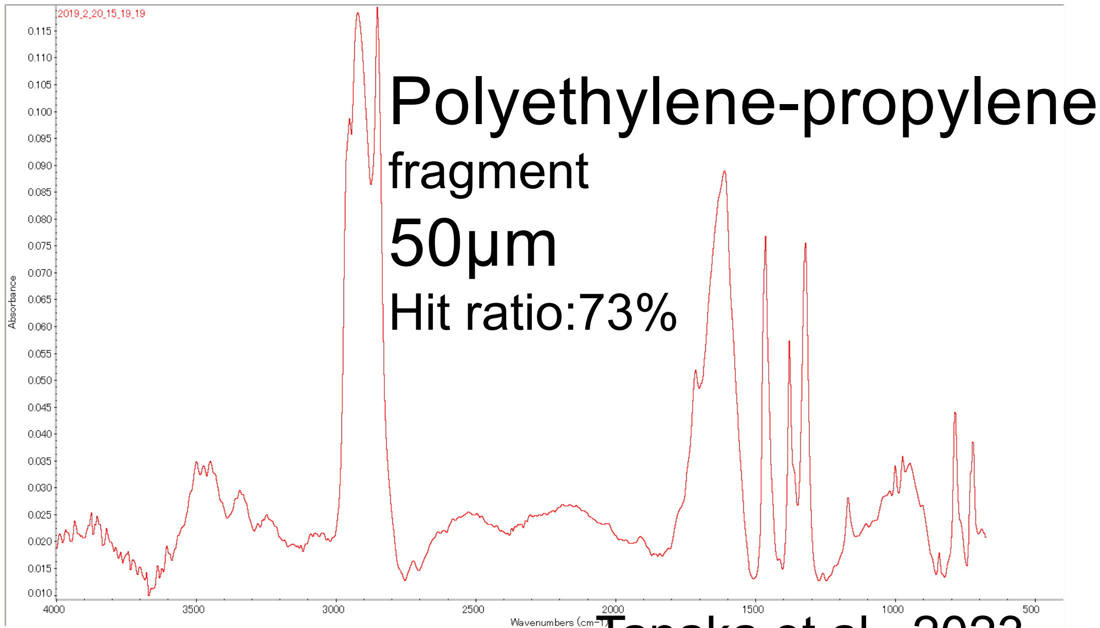

remote island in Okinawa, Japan
Plastic contaminated beach
Hermit Crab

Images and FTIR spectrum of microplastics found in stomach of Hermit Crab



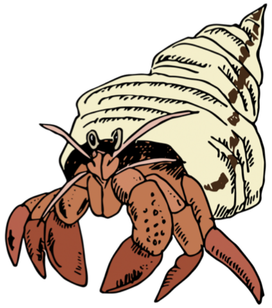






Tanaka et al., 2023

Microplastics in digestive tract of Hermit Crab



0 – 13 pieces/g-wet

Control
beach

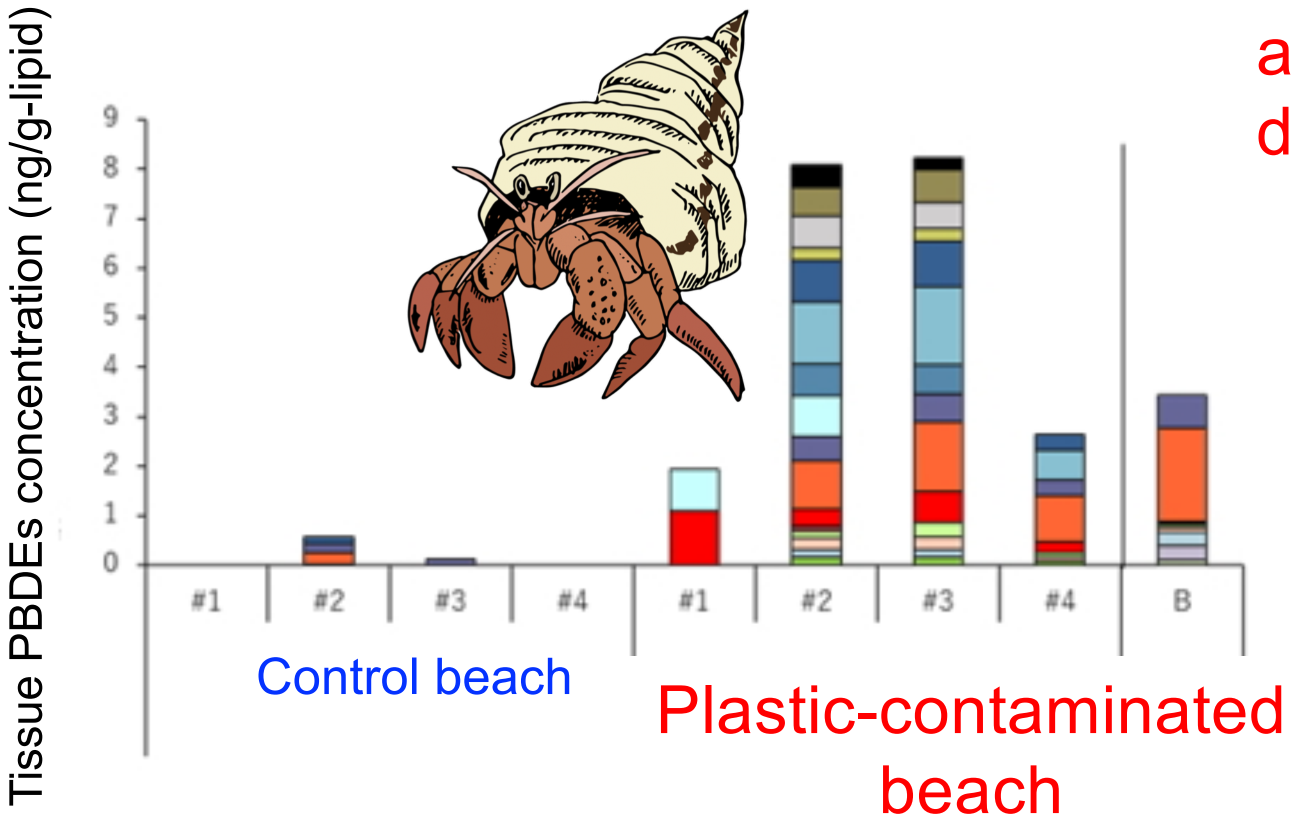
293 - 482 pieces/g-wet

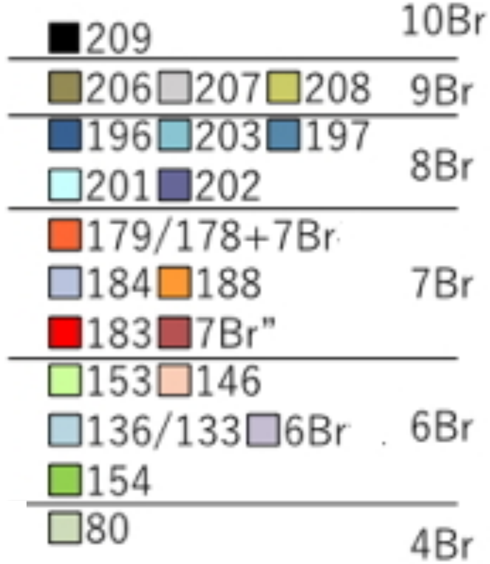
Plastic
contaminated
beach

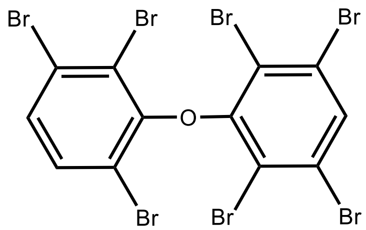
Tanaka et al., 2023

Field observation of transfer of plastic additives (**brominated flame retardants**) to hepatopancreas of **hermit crab**

Metabolites of the additives were dominant.







Tanaka et al., 2023

Laboratory exposure experiment of microbeads containing BDE209 to Hermit Crab

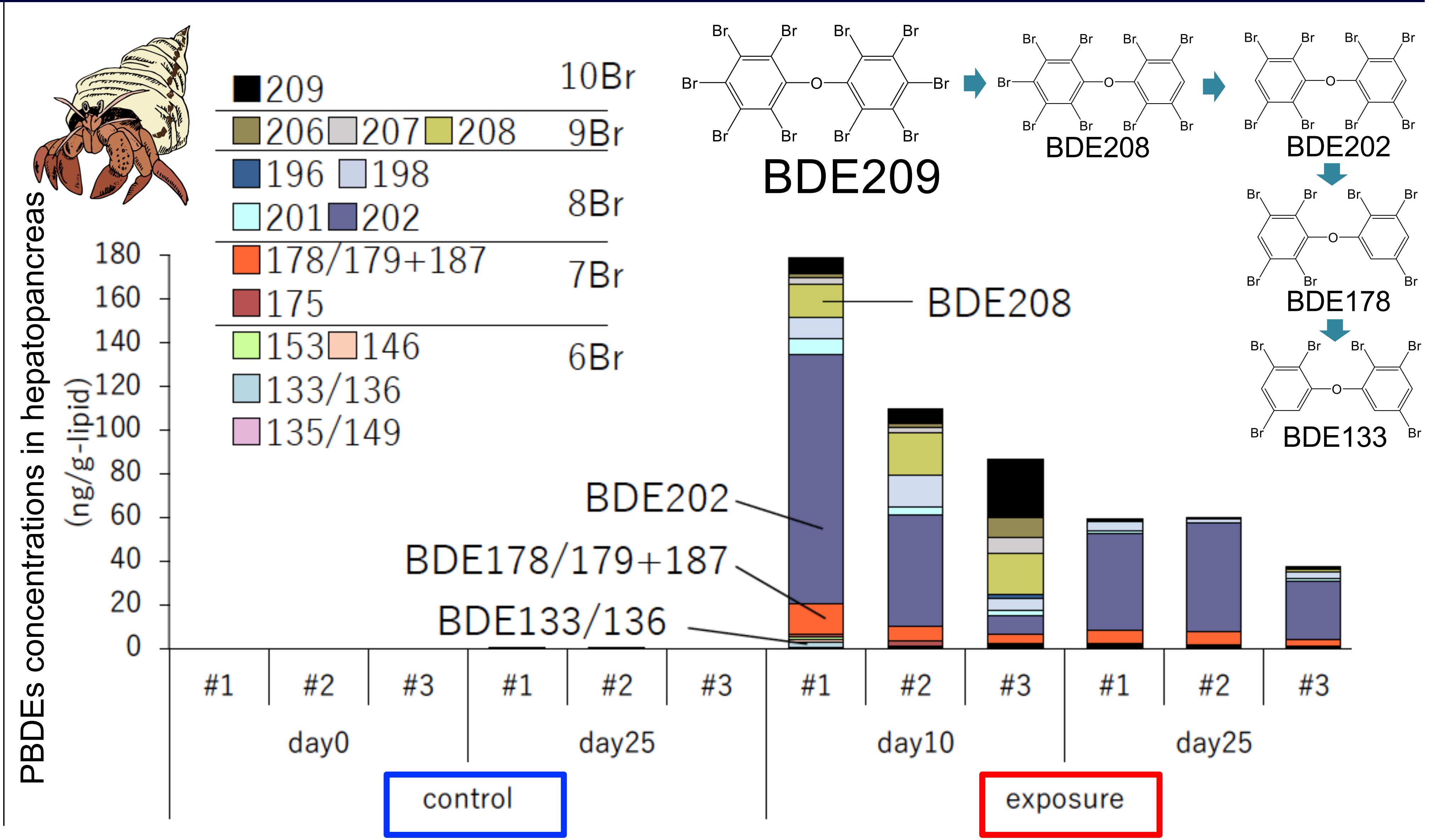
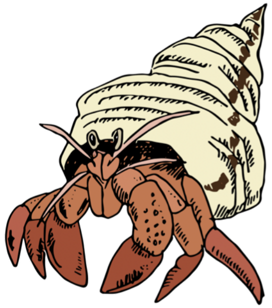


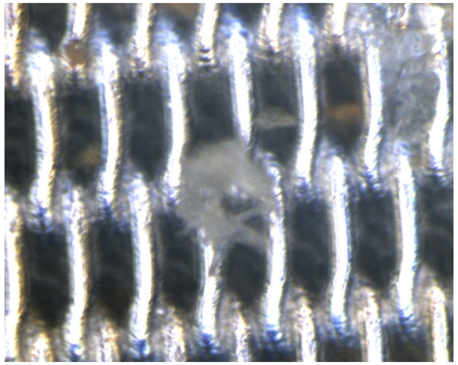
Fig. PBDEs concentrations in hepatopancreas of hermit crab exposed with microbeads containing BDE209

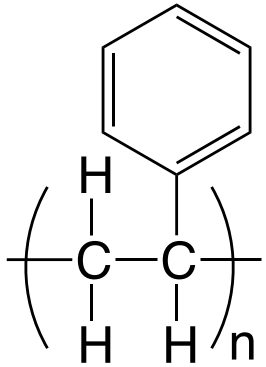
Tanaka et al., 2023

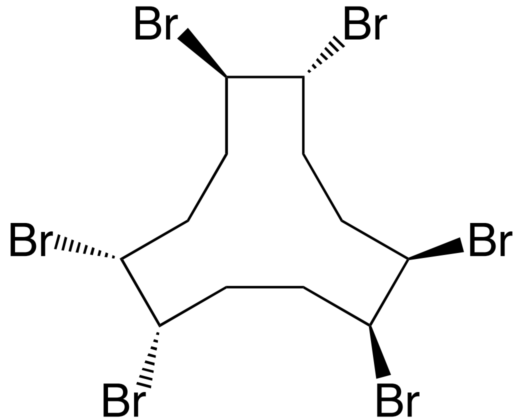
Pollution of plastics and related compounds from stranded litter to terrestrial hermit crabs in remote islands

Kaoruko Mizukawa, et al. (2023)
(Tokyo University of Agriculture and Technology)



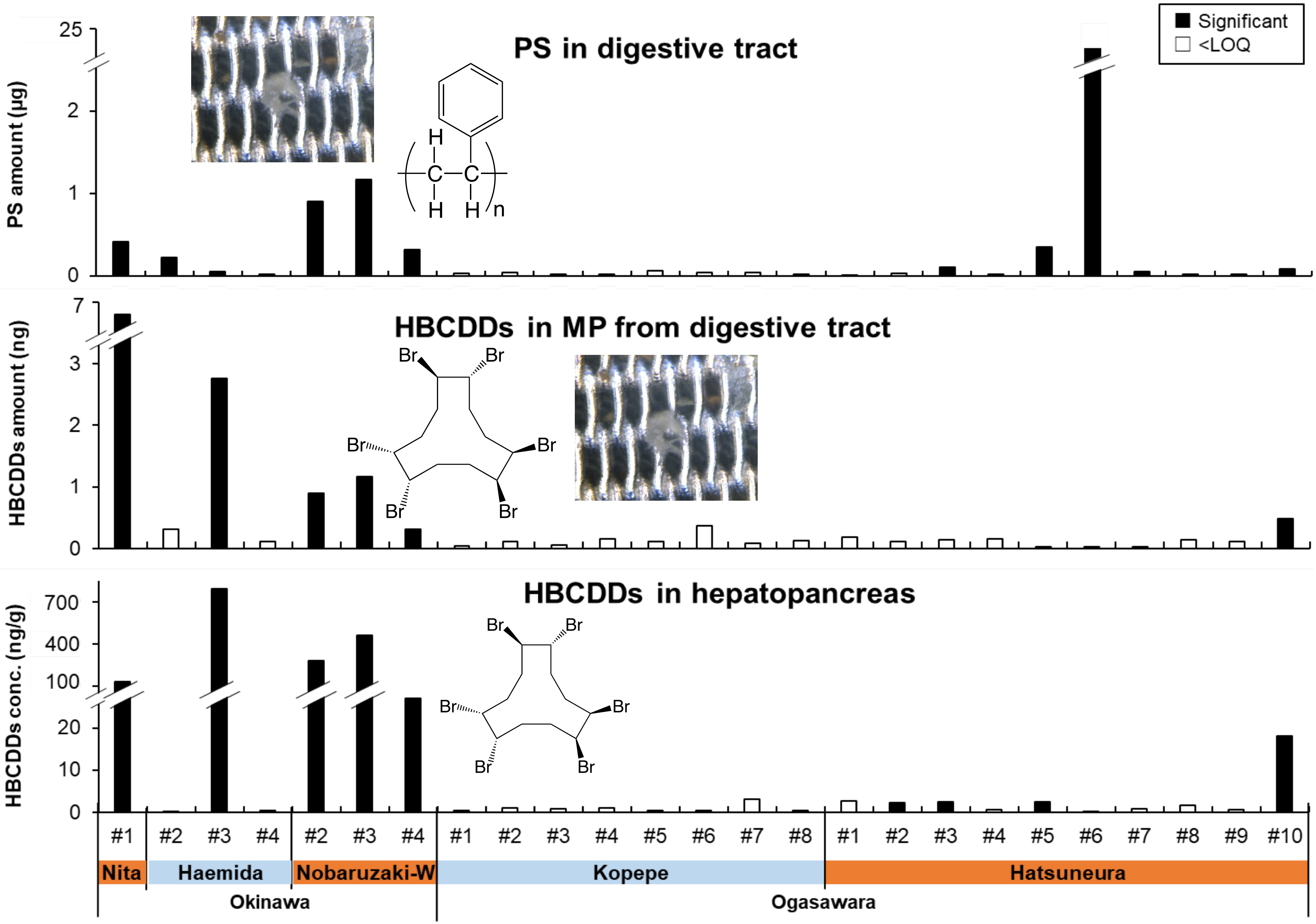






HBCDDs

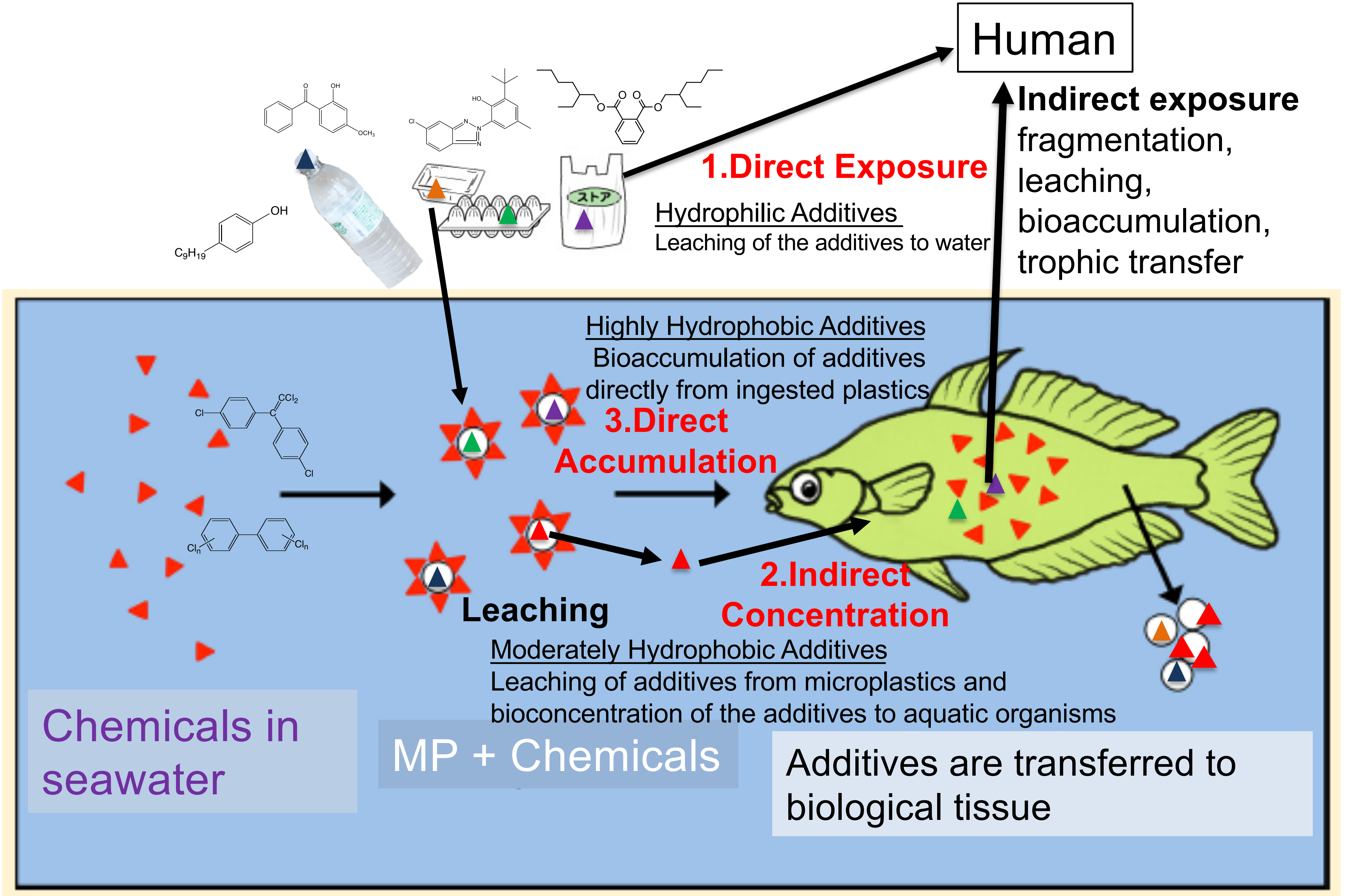
to be presented on Annual meeting of Japan Society for Environmental Chemistry (JEC)



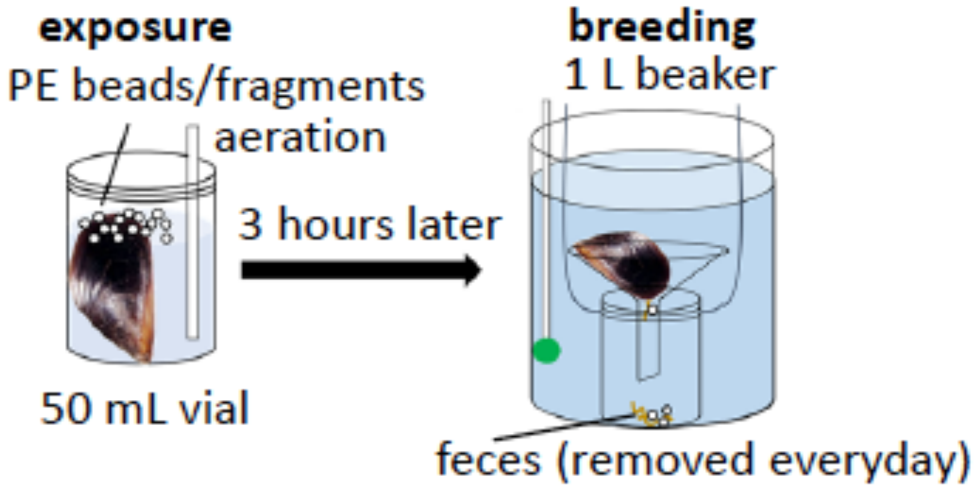
HBCDs in Hermit Crab

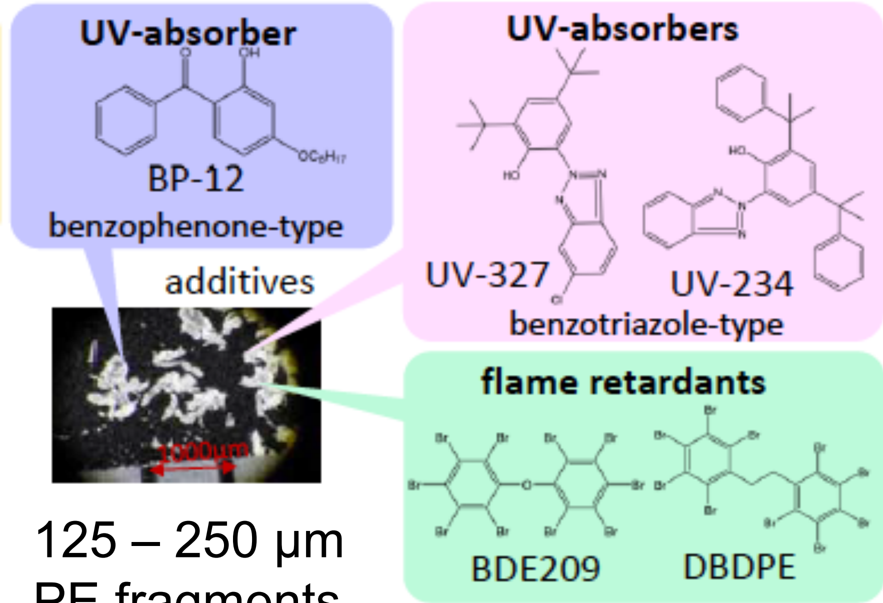
Mizukawa et al., 2023

All the classes of additives can be finally exposed to human through 3 pathways



Accumulation of Benzotriazole UV stabilizers following exposure of microplastic fragments containing the additives to mussel





125 – 250 µm
PE fragments

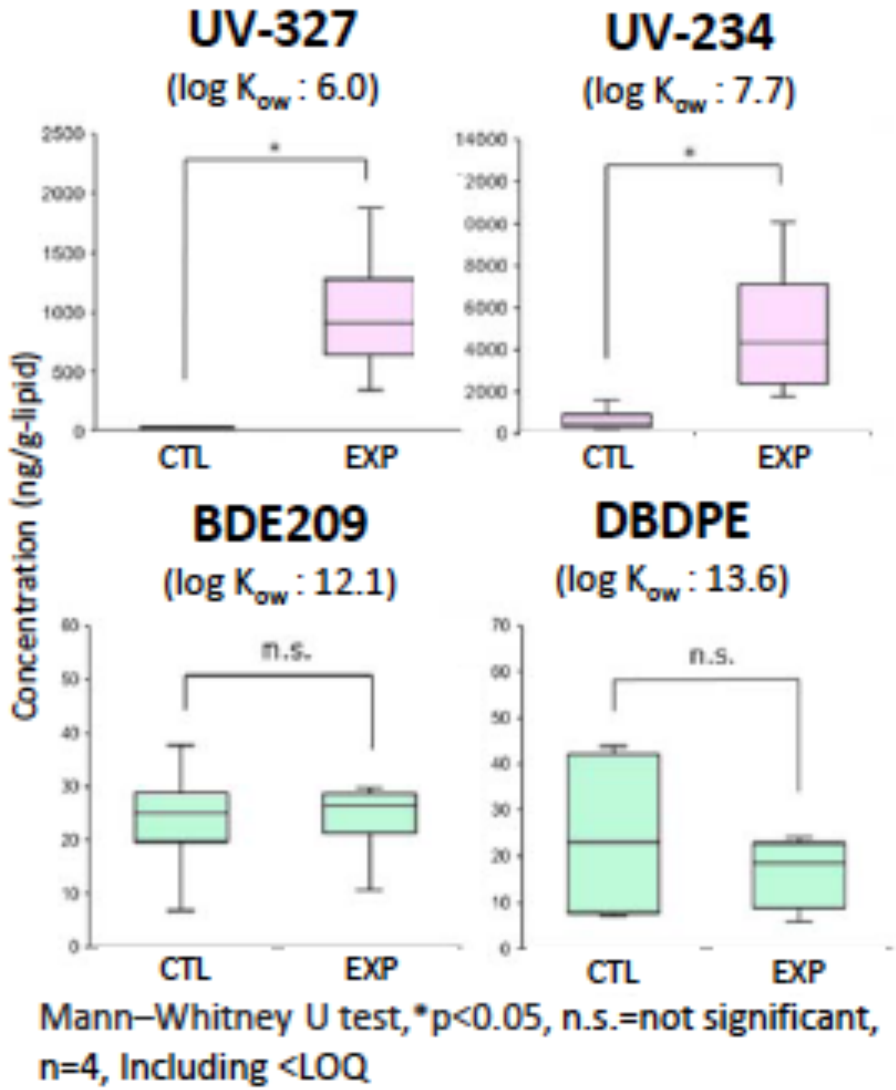
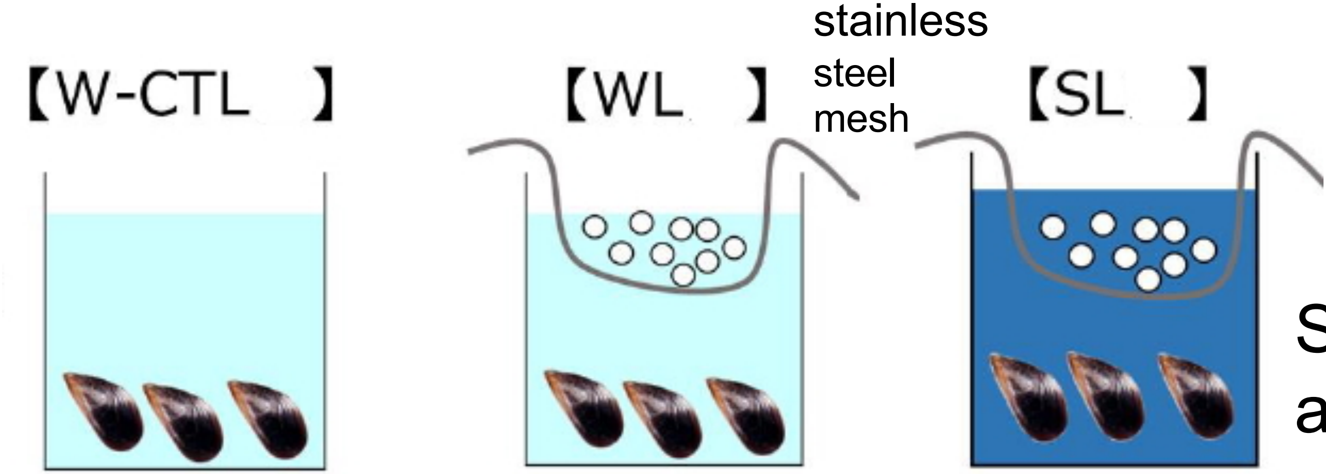


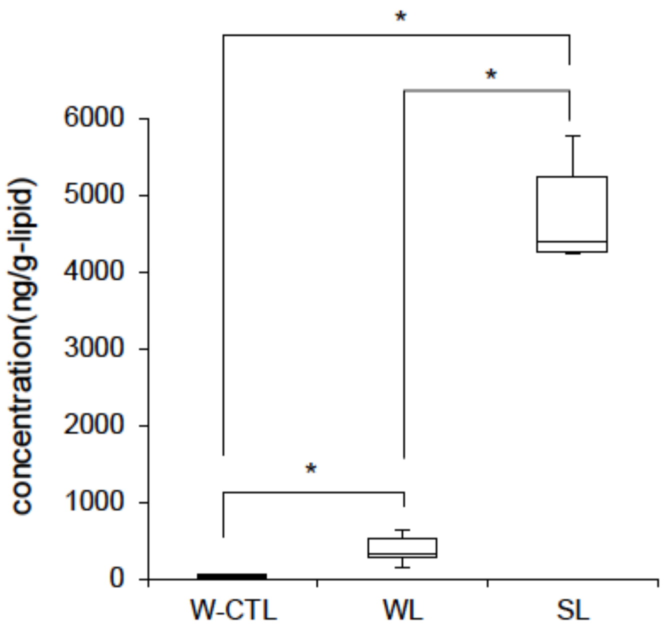
Fig. Concentrations of the additives in gonad of mussels

Mizukawa, K., Takano, T., Sakurai, R., Ota, M., Nakaoka, M., Kinjo, A., Inoue, K., Takada, H., 2022. Dietary exposure experiments on the transfer of chemical pollutants from microplastics to bivalves. In: International Online Workshop on Microplastics Issues, online.

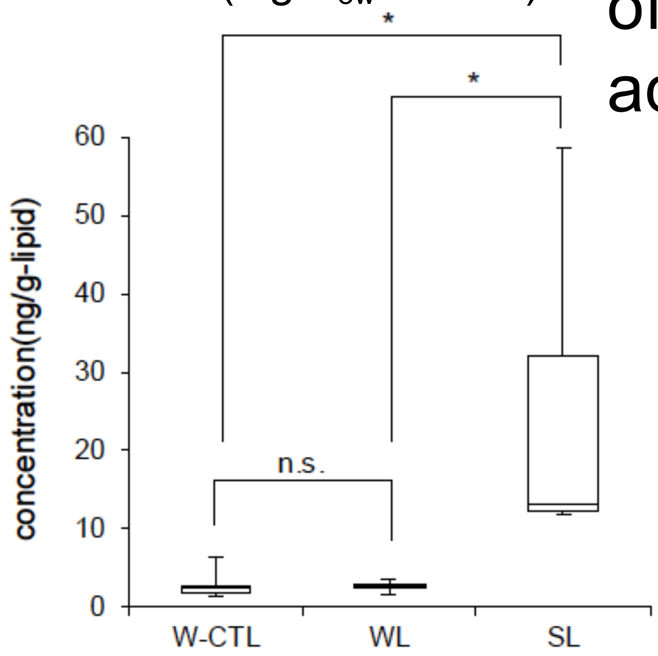
Indirect concentration of additives through leaching and bioconcentration occurred for moderately hydrophobic additives



【UV327】
(log K_{ow} : 6.91)



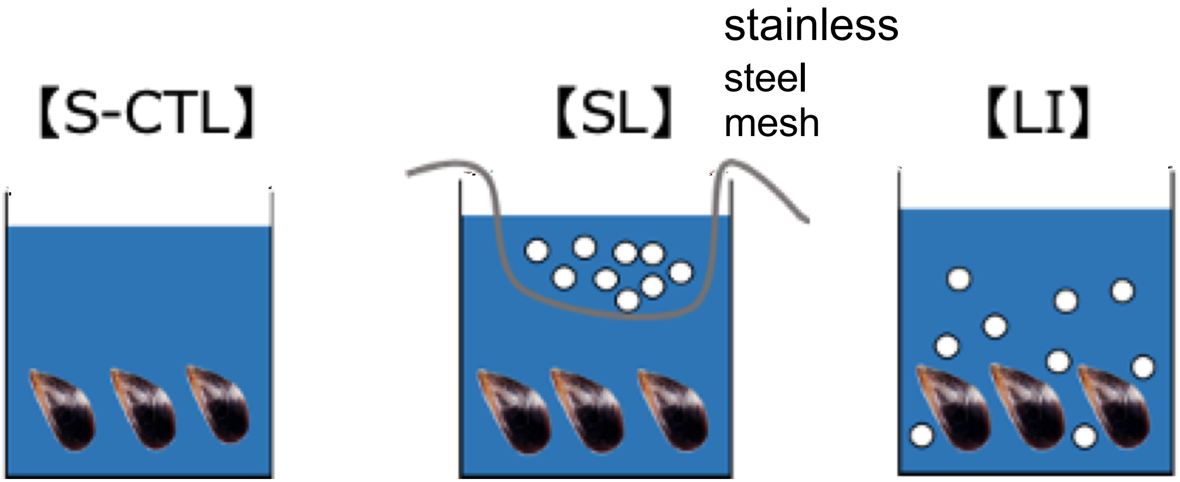
【BDE209】
(log K_{ow} : 12.11)

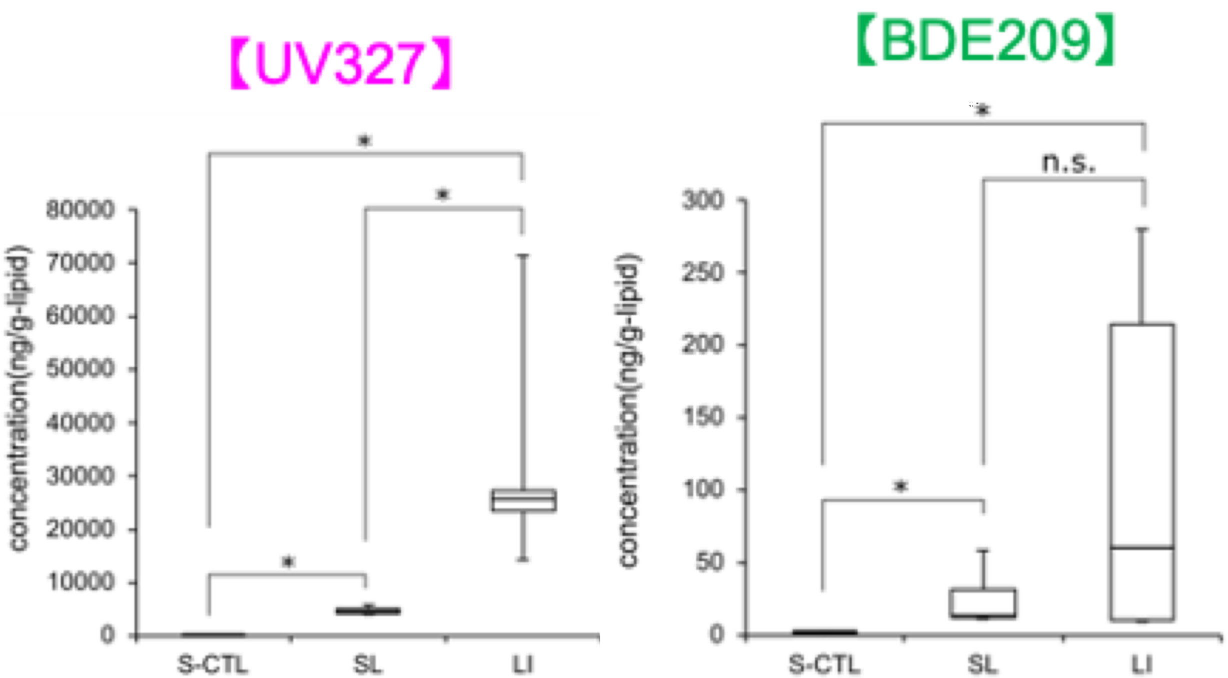


Surfactant accumulated leaching and indirect concentration of hydrophobic additives

Mizukawa et al., 2022

Direct bioaccumulation is more important for highly hydrophobic additives

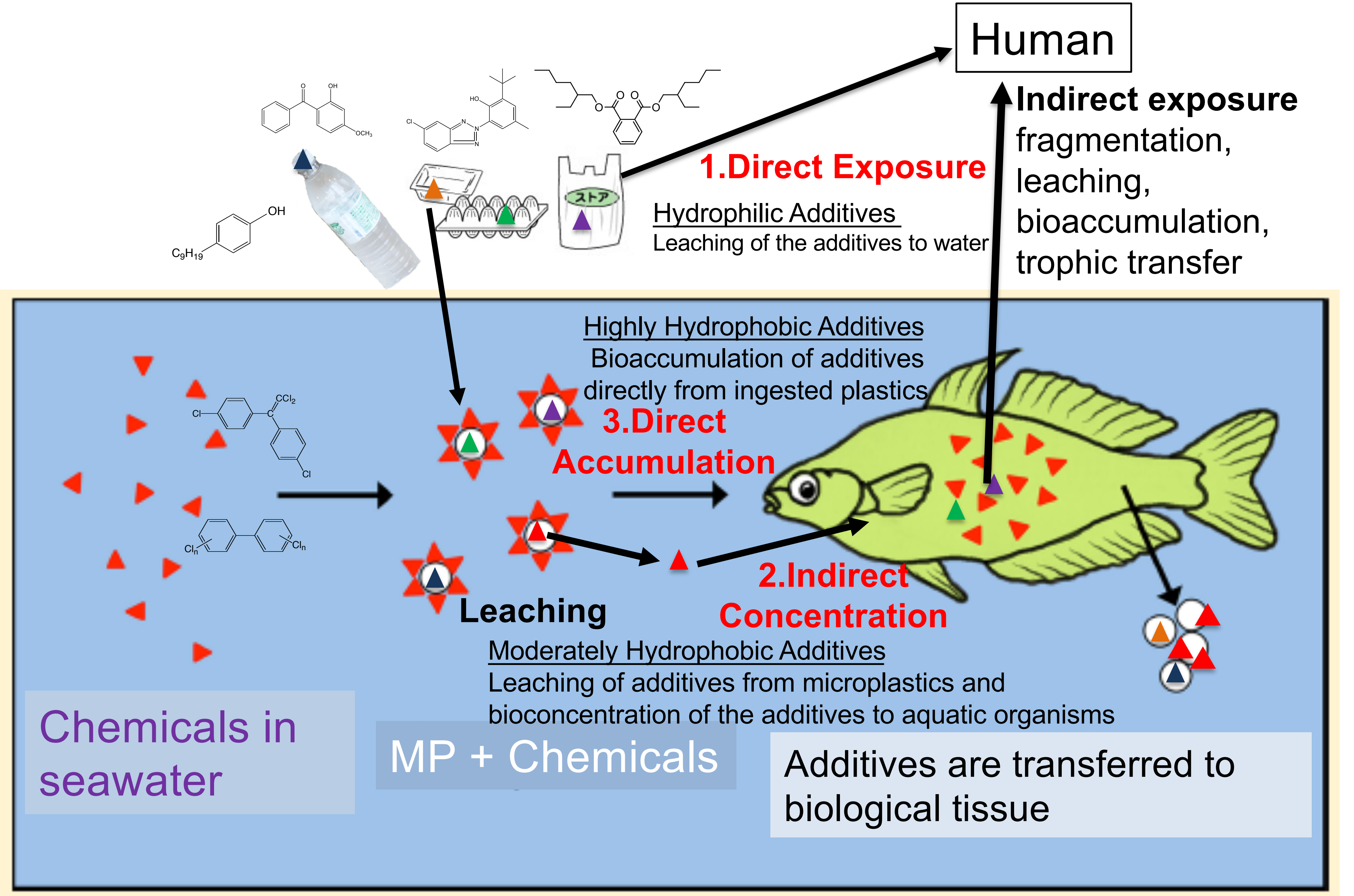




Surfactant accumulated leaching and
indirect concentration of hydrophobic additives

Mizukawa et al., 2022

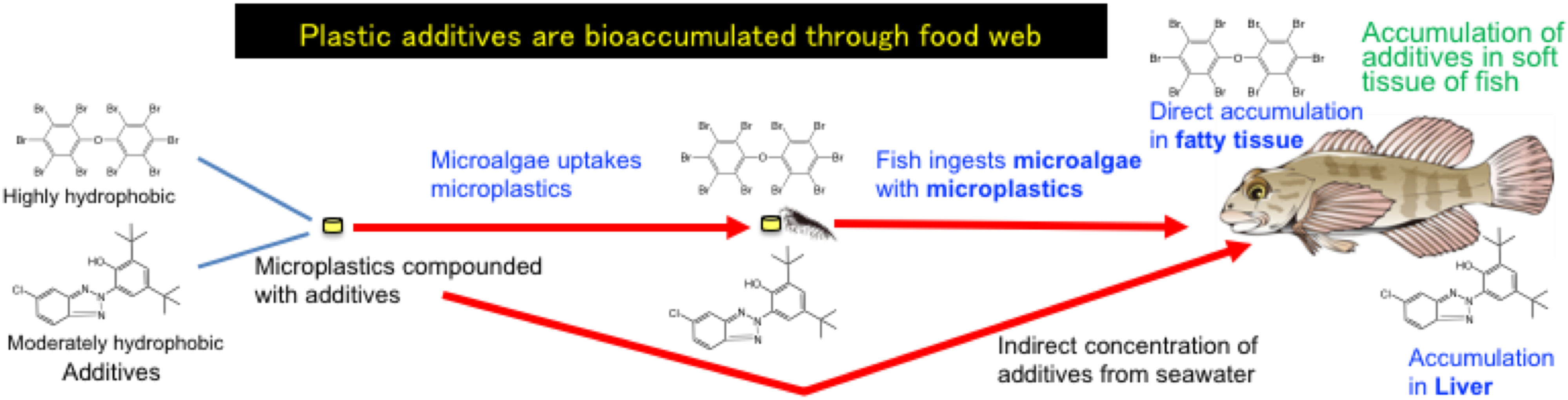
All the classes of additives can be finally exposed to human through 3 pathways



The significance of trophic transfer of microplastics in the accumulation of plastic additives in fish: An experimental study using brominated flame retardants and UV stabilizers

Takaaki Hasegawa [a,1], Kaoruko Mizukawa [b], Bee Geok Yeo [b], Tomonori Sekioka [c], Hideshige Takada [b], Masahiro Nakaoka [d,*]

[a] *Graduate School of Environmental Science, Hokkaido University, Akkeshi, Hokkaido 088-1113, Japan*
[b] *Laboratory of Organic Geochemistry, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183-8509, Japan*
[c] *Faculty of Fisheries Sciences, Hokkaido University, Hakodate, Hokkaido 041-8611, Japan*
[d] *Akkeshi Marine Station, Field Science Center for Northern Biosphere, Hokkaido University, Akkeshi, Hokkaido 088-1113, Japan*



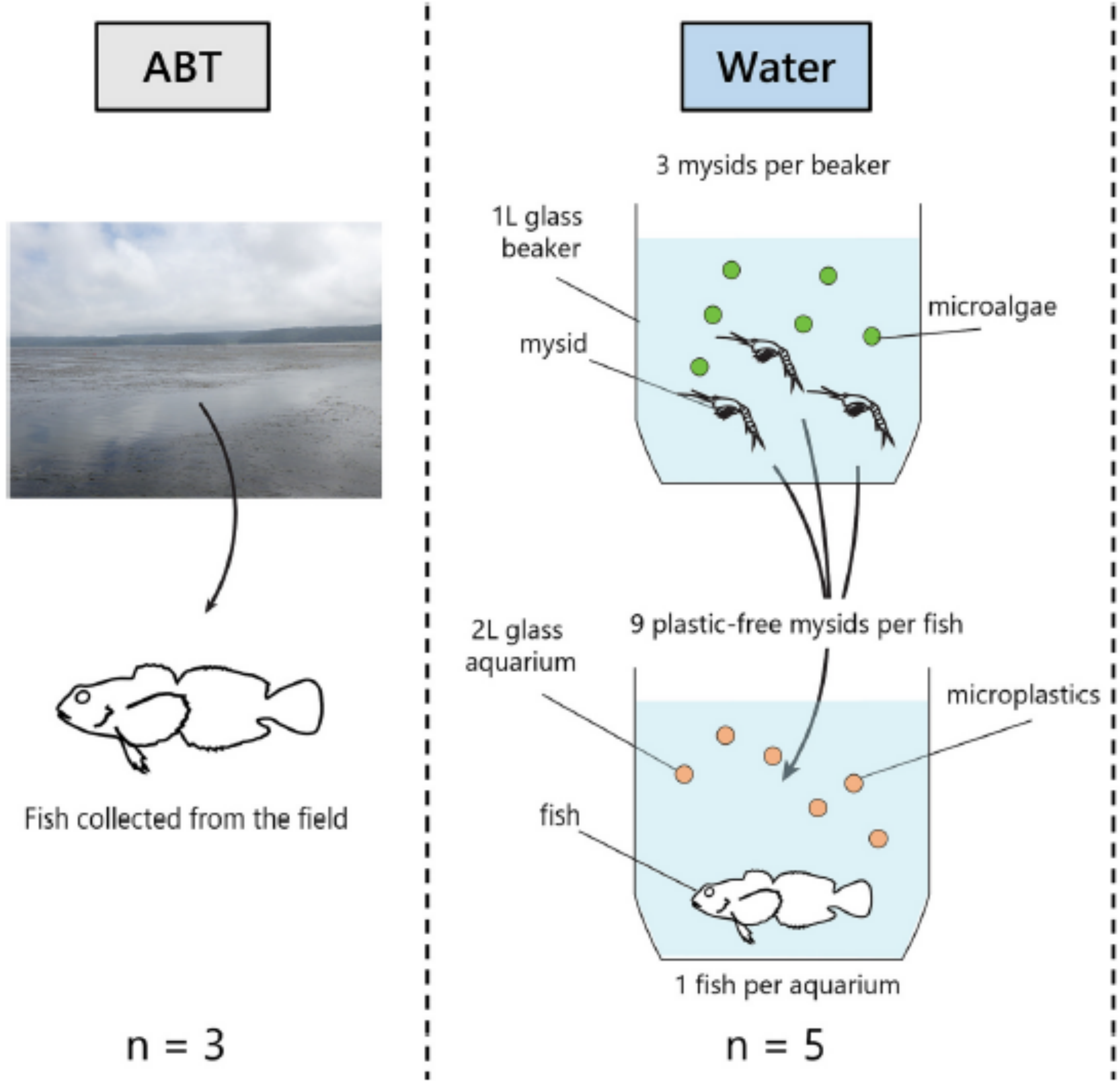


Fig. 1. Experimental design of the trophic transfer experiment. Fish (ABT) were collected from the field and analyzed to determine baseline concentrations of the target additives. Fish (Water) were fed plastic-free mysids and exposed to microplastics compounded with the additives suspended in the water column for 10 days. Fish (Mysid) were fed mysids that had been pre-exposed to microplastics compounded with the additives, but the aquarium water did not contain a microplastic suspension. n in the figure denotes the sample size for each treatment group.

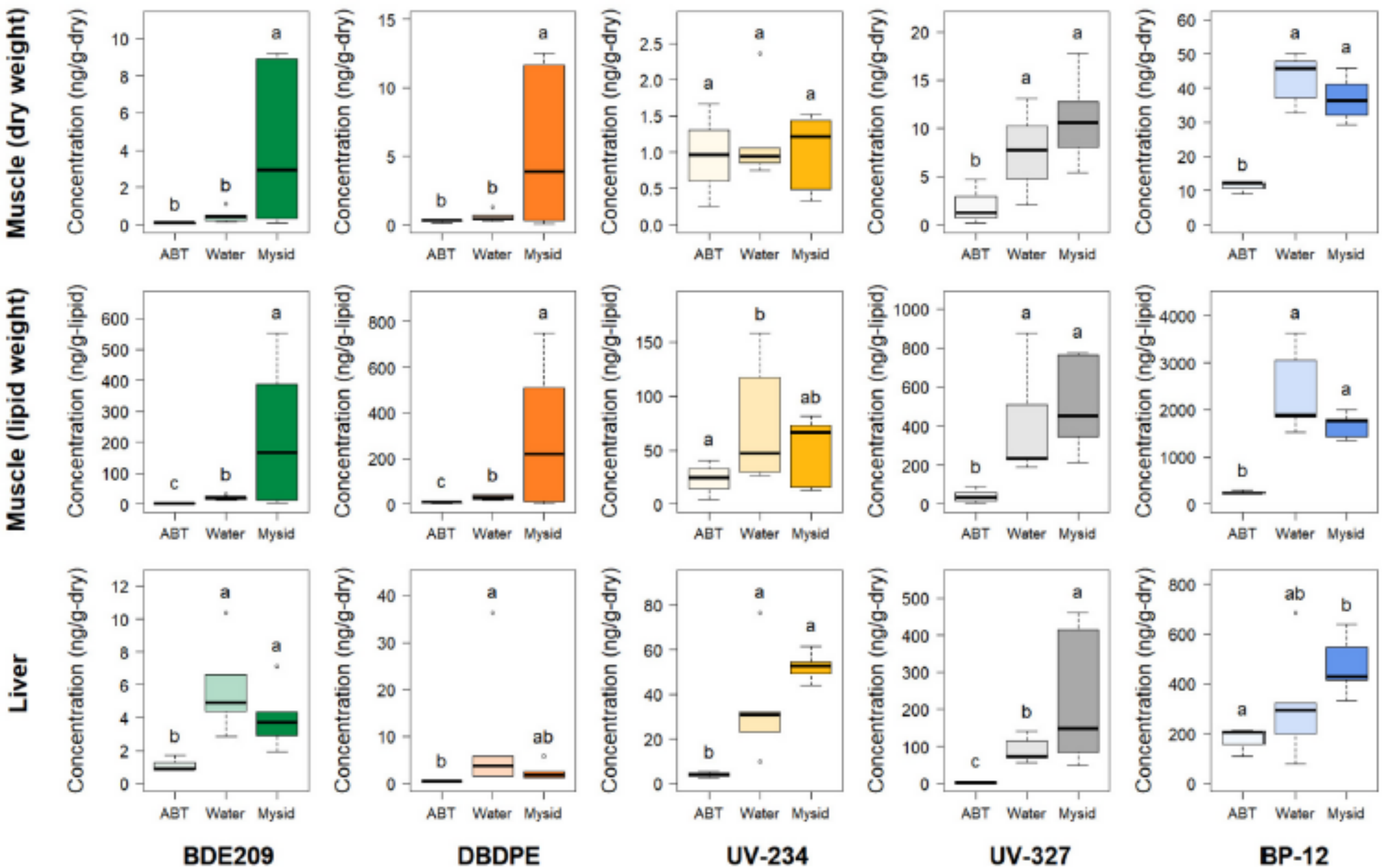
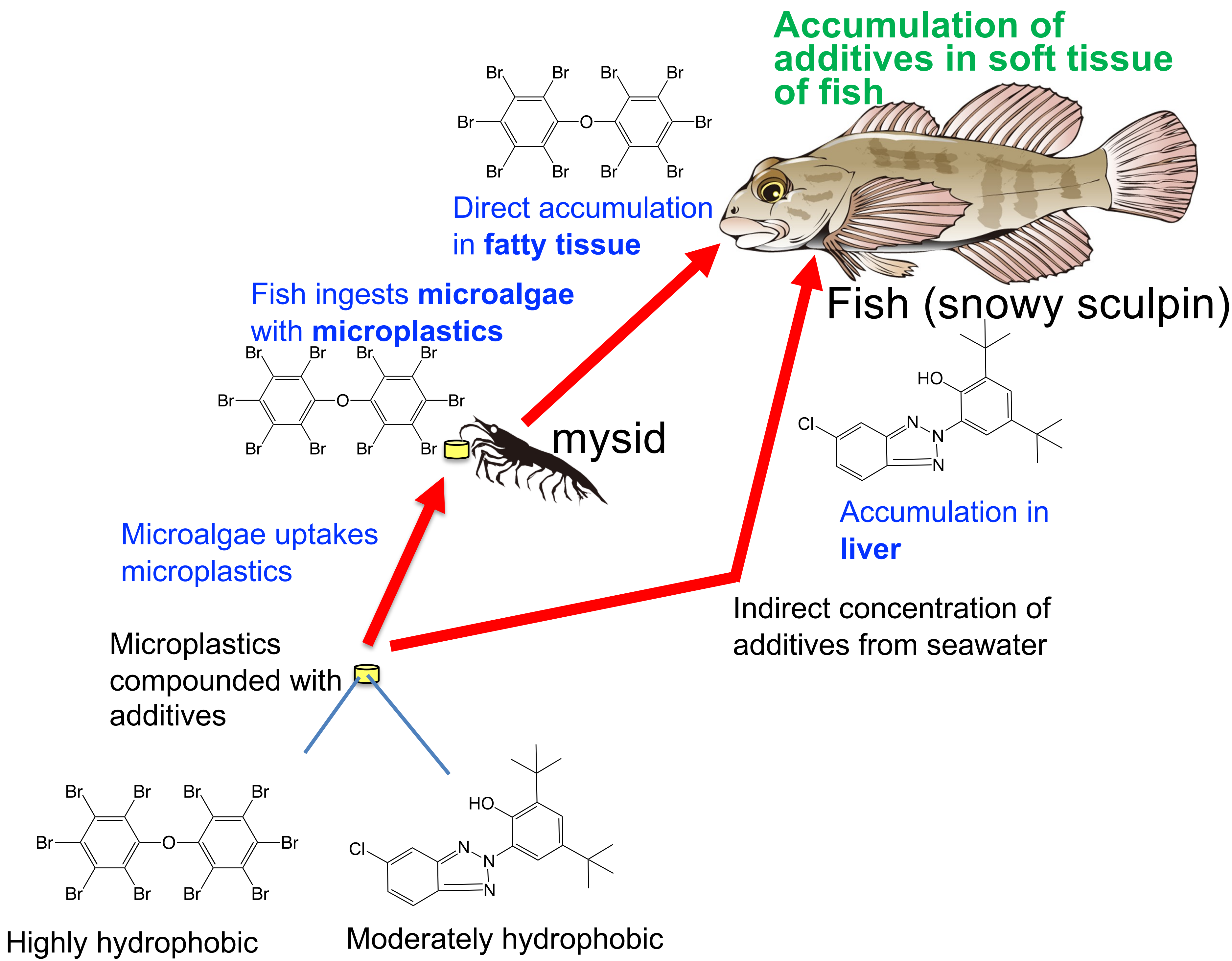


Fig. 3. Concentrations of the five additives in muscle (dry- and lipid-weight basis) and liver (dry-weight basis only) from fish immediately after collection from the ambient environment (ABT), fish exposed to microplastics suspended in the water column (Water), and fish fed mysids previously exposed to microplastics (Mysid). Concentrations were calculated by dividing the amount of the additive (ng) in the tissue by the dry weight (also lipid weight for muscle) of the tissue (g). Within each box and whisker plot, the solid horizontal line is the median, the bottom and top are the 25th to 75th percentiles, and the whiskers show the 10th and 90th percentiles. Values with the same letter are not significantly different ($P < 0.01$, generalized linear model with post hoc Tukey's honestly significant difference test).



Accumulation of additives in soft tissue of fish
Direct accumulation in **fatty tissue**
Fish (snowy sculpin)
Fish ingests **microalgae** with **microplastics**
mysid
Accumulation in **liver**
Microalgae uptakes microplastics
Indirect concentration of additives from seawater
Microplastics compounded with additives
Highly hydrophobic
Moderately hydrophobic

Conclusion:

Analysis of PP pellets demonstrated that **mm-size plastics transport BUVSs for hundreds to thousands kilo meters** without drastic degradation nor leaching.
>>International regulation is necessary.

Laboratory exposure experiments and field observations indicate that **hydrophobic additives** are not inert and **can be bioaccumulated in marine organisms including mussel, hermit crab, fish, and seabirds**.

Fragmentation, breaking down of microplastics, oily digestive fluid, and **surfactants** facilitate bioaccumulation of the additives.

For **highly hydrophobic additives** such as BDE209, **direct accumulation from ingested plastics** would be more important.

For **moderately hydrophobic additives** such as UV328 and UV327, **leaching to water and bioconcentration via water** could be important, too.

Implication and Recommendation:

>>International regulation

Microplastics are not biodegraded and easy to be resuspended and can be **repeatedly ingested by the biota until their burial.** Long-term accumulative exposure of additives to marine organisms and finally to human could be larger than we estimated based on single ingestion.

>>Total exposure of additives to human should be assessed by measurement of additives in all the potential sources and understating of individual processes.

On the other hand, **various plastic additives are detected in human body** and **some symptom of endocrine disruption has been already detected**.

>>As a precautionary approach, reduction of plastic usage has been recommended.

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THE MINDEROO-MONACO COMMISSION ON PLASTICS AND HUMAN HEALTH

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AROUB K. YOUSUF
SARAH DUNLOP

*Author affiliations can be found in the back matter of this article

Conclusions: It is now clear that current patterns of plastic production, use, and disposal are not sustainable and are responsible for significant harms to human health, the environment, and the economy as well as for deep societal injustices.

The main driver of these worsening harms is an almost exponential and still accelerating increase in global plastic production. Plastics' harms are further magnified by low rates of recovery and recycling and by the long persistence of plastic waste in the environment.

The thousands of chemicals in plastics—monomers, additives, processing agents, and non-intentionally added substances—include amongst their number known human carcinogens, endocrine disruptors, neurotoxicants, and persistent organic pollutants. These chemicals are responsible for many of plastics' known harms to human and planetary health. The chemicals leach out of plastics, enter the environment, cause pollution, and result in human exposure and disease. All efforts to reduce plastics' hazards must address the hazards of plastic-associated chemicals.

Recommendations: To protect human and planetary health, especially the health of vulnerable and at-risk populations, and put the world on track to end plastic pollution by 2040, this Commission supports urgent adoption by the world's nations of a strong and comprehensive Global Plastics Treaty in accord with the mandate set forth in the March 2022 resolution of the United Nations Environment Assembly (UNEA).

International measures such as a Global Plastics Treaty are needed to curb plastic production and pollution, because the harms to human health and the environment caused by plastics, plastic-associated chemicals and plastic waste transcend national boundaries, are planetary in their scale, and have disproportionate impacts on the health and well-being of people in the world's poorest nations. Effective implementation of the Global Plastics Treaty will require that international action be coordinated and complemented by interventions at the national, regional, and local levels.

This Commission urges that a cap on global plastic production with targets, timetables, and national contributions be a central provision of the Global Plastics Treaty. We recommend inclusion of the following additional provisions:

Acknowledgement

We express our sincere appreciation to International Pollution Elimination Network (IPEN) and IPW volunteers for their collection of plastic samples, Dr. Denise Britta Hardesty, Dr. Juan Pablo Muñoz, Dr. Carlos A. Valle, Dr. Peter Ryan, Dr. David Hyrenbach, and Dr. Yutaka Watanuki for their providing the preen gland oil samples. The present study was supported by a Grant-in-Aid from the Ministry of Education and Culture of Japan (Projects No. 16H01768 and No. 17J05875) and the Environment Research and Technology Development Fund (SII-2-2)