

## **Pellet Watch : Global Monitoring of Persistent Organic Pollutants (POPs) using Beached Plastic Resin Pellets.**

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### **What's plastic resin pellet?**

Plastic resin pellets are small granules generally with shape of a cylinder or a disk with a diameter of a few mm. These plastic particles are industrial raw material transported to manufacturing sites where “user plastics” are made by re-melting and molding into the final products. Resin pellets can be unintentionally released to the environment, both during manufacturing and transport. The released resin pellets are carried by surface run-off, stream, and river waters eventually to the ocean. Resin pellets can also be directly introduced to the ocean through accidental spills during shipping. Because of their environmental persistence, they are distributed widely in the ocean and found on beaches and on water surfaces all over the world (Carpenter and Smith, 1972; Gregory, 1978; Moore *et al.*, 2001a; Moore *et al.*, 2001b; Derraik, 2002; Moore *et al.*, 2002). A growing production of plastic leads to a measurable increase in plastic pollution in the ocean. In 2001, we revealed the existence of various organic micropollutants (i.e., polychlorinated biphenyls : PCBs, DDE, and nonylphenol) in plastic resin pellets collected from Japanese coasts (Mato *et al.*, 2001). Since ever, we have been conducting studies on organic micropollutants in marine plastic resin pellets (Mato *et al.*, 2002; Endo *et al.*, 2005). In the present paper, we will introduce the results of our recent studies. Also ***Pellet Watch*** : Global Monitoring of Persistent Organic Pollutants (POPs) using Beached Plastic Resin Pellets will be proposed.

### **Additive-derived pollutants in marine plastic resin pellets**

Marine plastic resin pellets carry two types of organic micropollutants (i.e., additive-derived pollutants and sorbed pollutants). First type includes plastic additives (e.g., anti-oxidants) and their

degradation products. Nonylphenols (NP) were detected in plastic resin pellets collected from Japanese coasts. Endocrine disrupting potency of NP has been recently revealed (Jobling *et al.*, 1996) and distributions, sources, and environmental behavior of NP has been studied by many researchers (Ahel *et al.*, 1994; Bennet and Metcalfe, 1998; Blackburn *et al.*, 1999; Isobe *et al.*, 2001). Plastic resin pellets were collected from 9 beaches in Japan in 1998 – 2000. The pellets were classified according to their polymer types (i.e., polyethylene : PE and polypropylene : PP) using near-infrared spectroscopy (PlaScan-SH, OPT Research Inc. Tokyo, Japan). PE and PP pellets were extracted with dichloromethane (DCM) using Soxhlet extractor and the extracts were purified using silica gel column chromatography. NP was determined by gas chromatograph equipped with mass spectrometer (GC-MS). Details of analytical conditions were described in Isobe *et al.* (2001) and Mato *et al.* (2001).

Concentrations of NP in the plastic resin pellets are shown in Figure 1. NP concentrations ranged from 18 ng/g to 17000 ng/g. These concentrations were similar to those observed in bottom sediments in Tokyo Bay, one of the most polluted bay in the world. NP detected in the plastic resin pellets is likely to be derived from trisnonylphenol phosphite (TNP), polymer antioxidants, as shown in Figure 2. TNP antioxidant contains significant amounts of NP as an impurity (Gilbert *et al.*, 1986) and also TNP can be easily oxidized and hydrolyzed to be NP in ambient conditions (Murata, 1999). Nonylphenolpolyethoxylates (NPEO; Fig.2), surface active agents, are also added to plastics and are a potential source of NP in the resin pellets. Figure 1 shows a tendency of PP pellets to have a higher amount of NP than PE pellets. This is probably because PP requires more additives. Marine plastic pellets may act as significant source of NP to marine organisms. It is thought that biomagnification of NP through the food chain could not be occurring due to their hydrophilic group and metabolizable nature, although these conclusions have not been well proved. Several studies suggest that biomagnification may not play an important role to transfer NP, unlike PCBs and DDE, to animals and birds in higher trophic levels (Ahel *et al.*, 1993). In these contexts, direct ingestion of plastic pellets could be a direct and important route of NP to higher animals.

### **Sorption of organic micropollutants on marine plastic resin pellets**

Another type of micropollutants in the marine plastic resin pellets is pollutants sorbed from ambient seawater. Polychlorinated biphenyl (PCBs), dichloro-diphenyl-dichloroethylene (DDE; degradation product of organochlorine pesticide), and polycyclic aromatic hydrocarbons (PAHs) were detected in the marine plastic resin pellets collected from Japanese coasts (Mato *et al.*, 2002). Plastic resin pellets were collected from 9 beaches in Japan in 1998 – 2000. The resin pellets were classified into PE and PP by the spectroscopic method and were analyzed for PCBs, DDE, and PAHs using gas chromatograph equipped with electron capture detector (GC-ECD) and GC-MS following purification of DCM extracts by two-step silica gel column chromatography. The details of the analytical procedure were described in Endo *et al.* (2005). Concentrations of PCBs in the resin pellets are shown in Figure 3. The range of PCB concentrations in the pellets was from 5 ng/g to 892 ng/g. There observed a trend that PE pellets had higher concentrations of PCBs than PP pellets.

Because the surface of the resin pellets is highly hydrophobic, adsorption of PCBs from seawater onto the pellet surface was likely to be the mechanism of the accumulation of PCBs in the marine pellets. A field adsorption experiment, where virgin plastic pellets were deployed on the sea surface, was conducted to examine whether plastic pellets can adsorb hydrophobic pollutants from seawater

(Fig.4). The experiment was carried out in port of Tokyo Fishery University which is located in northern head of Tokyo Bay from December 20, 1998 to January 6, 1999. During this period, there were no rainy or windy days and the sea was calm. Three types of plastic materials were used for the experiment and were compared in their adsorption. Virgin PE pellets (Suntch LD M 652) and two types of PP resin pellets (Grandpolymer J104W and J705M) were obtained from polymer companies and deployed. Our prior analysis showed no significant PCBs ( $<0.01\text{ng/g}$ ) in the virgin pellets. Approximately 8 g each of the pellets were placed in stainless steel mesh baskets (30 cm diameter and 13 cm height) and the baskets were left drifting on the sea surface (Fig.4). Under these conditions, the pellets were in a single layer and could float freely in the basket in full contact with surface seawater. Three baskets were placed out for each pellet type at the beginning of the experiment, and one basket was recovered on 9 days and 16 days after the deployment. The recovered pellets were stored in glass bottles at  $-30\text{ }^{\circ}\text{C}$  in the laboratory. Just before analysis, the pellets were air-dried in the clean room for 3 days. Eight g of the dried pellets were analyzed as described above. Non-deployed virgin pellets were also analyzed along with the sample pellets and considered as Day-0 sample.

The field adsorption experiment results demonstrate that PCBs adsorb to plastic resin pellets from seawater. During the experiment, PCBs in the deployed pellets showed steady concentration increase with time (Figure 5; Matoet *al.* 2002). These increasing trends clearly indicate that ambient seawater is the source of PCBs found in the marine plastic resin pellets and that adsorption to pellet surfaces is the mechanism of enrichment. As PE and PP resin pellets are made of saturated hydrocarbon units, their surfaces are nonpolar and adsorb hydrophobic pollutants such as PCBs through hydrophobic sorption. Figure 5 also indicated an interesting result that PE pellets adsorbed more PCBs than PP pellets. This result means a higher affinity of PE pellets for hydrophobic compounds than PP pellets. The result was consistent with the tendency observed for stranded pellets on beaches, that is, PE pellets had a higher amount of PCBs than PP pellets, as shown in Figure 3. This difference is probably caused by the different surface chemistry between PP and PE.

Comparison of PCBs concentrations in marine plastic resin pellets with those in seawater suggests their high degree of accumulation (apparent adsorption coefficient :  $10^5 - 10^6$ ). The high accumulation potential suggests that plastic resin pellets serve as a carrier of toxic chemicals to marine biota (e.g., seabird) which intakes the plastic resin pellets. Further studies must be done on the transfer of the chemicals from pellets to biological tissue while they transit through the digestive system of birds and other animals. On the other hand, the high accumulation potential of the marine plastic pellets suggests the potential use of plastic pellets as a tool for monitoring of organic micro-pollutants in seawater. In the following sections, their potential for monitoring tool is examined through studying regional differences in PCB concentrations in beached resin pellets.

### **Regional difference in PCBs concentrations between countries**

Beached plastic resin pellets were collected from 11 Japanese coasts and 6 Malaysian coasts (Fig.6). Japan has used larger amounts of PCBs for industrial use than Malaysia and difference in PCBs contamination has been demonstrated between both coastal environments through various monitoring. Thus, PCBs concentrations in the beached resin pellets were compared between Japan and Malaysia. The resin pellets were classified according to their polymer types using near-infrared

spectroscopy (PlaScan-SH, OPT Research Inc. Tokyo, Japan) and PE pellets were subjected to the analysis of PCBs. Approximately 50 pellets were extracted with DCM and analyzed for PCBs as described above. As shown in Figure 7, PCB concentrations in PE pellets collected from Japanese coasts were one to two orders of magnitude higher than those from Malaysian coasts (Mato *et al.*, 2002). This difference is consistent with regional difference in PCBs concentrations in the indicator organisms (i.e., mussels). Mussel has been used as sentinel organisms to detect marine pollution because of their nature to accumulate contaminants in their tissue and cosmopolitan habitats. “Mussel Watch”, that is monitoring of marine pollution using mussel, has been applied worldwide (Goldberg, 1975; Farrington *et al.*, 1983; Tanabe *et al.*, 2000; Tsutsumi *et al.*, 2002). PCBs concentrations in mussels collected in Tokyo Bay (Yamaguchi *et al.*, 2000) were two orders of magnitude higher than those in Malaysia (Monirith *et al.*, 2000), as shown in Figure 7. The regional difference for mussel is well correlated with those for PE pellets. The correlation suggests that beached plastic pellets can be used as monitoring tool of marine pollution.

### **Piece-by-piece variability in PCB concentrations in a single beach**

However, before beached resin pellets will be applied for pollution monitoring, we should recognize that beached resin pellets have different materials (e.g. polypropylene [PP], polyethylene [PE]), surface conditions (e.g. fouled, weathered), and colors (e.g. clear, discolored, pigmented). This implies that the potential as a sorbent and the history before stranding may vary considerably among particles. Concentrations of the sorbates in resin pellets could thus vary significantly, even in pellets collected on a single beach. This piece-by-piece variability on a beach should be taken into account when concentrations of contaminants in the resin pellets from various beaches are compared and the regional variation of the concentrations is discussed. Thus, we investigated the variability of PCB concentrations among individual resin pellets from a single beach and the relationship of the concentrations to materials, weathering, fouling, and discoloration of the pellets (Endo *et al.*, 2005).

Sixty resin pellets were collected from a beach in Kasai Seaside Park, Tokyo, in July 2001 for studying the variation in PCB concentrations between pellets. The pellets were picked up along the high tide line within a range of 30 m, using solvent-rinsed stainless steel tweezers. The type of material, discoloration, fouling, and weathering of the pellets were examined using macroscopic, spectroscopic, and calorimetric methods and the individual resin pellets were characterized. The concentrations of PCBs in the individually characterized resin pellets were determined as described above.

Of the 60 resin pellets collected from Kasai Seaside Park, 35 were classified as PE and 20 as PP. These 55 pellets were analyzed individually for sorbed PCBs. The PCB concentrations ranged from below the detection limit (i.e., <28 ng/g; 24 pellets) to 2300 ng/g (Figure 8; Endo *et al.*, 2005). The pellet with the highest concentration accounted for ~50% of the sum of PCBs in the 55 pellets, and the top 3 pellets comprised ~60% of the total PCBs. The arithmetic mean of PCB concentrations of the 55 pellets was 93 ng/g, and 50 pellets had concentrations less than the mean. These results clearly indicate that the concentration of sorbates varies considerably (i.e., over 2 orders of magnitude) among the pellets collected from one beach, and that a small proportion of the pellets contains a large proportion of the chemicals. For the application of beached resin pellets as monitoring tool, this variability, especially occurrence of a few pellets with sporadically high PCBs concentration, should be taken care

carefully. Figure 8 also shows a tendency of PE pellets to have a higher amount of PCBs than PP pellets. This result is consistent with the results described above. This also suggests that PE pellets are preferable for the monitoring purpose. We also investigated the relationship of the variability in PCB concentrations to materials, weathering, fouling, and discoloration of the pellets. Discolored and/or fouled PE resin pellets had higher concentrations of PCBs than non-discolored and non-fouled ones, but no clear relationship between crystallinity and weathering with PCB concentrations was found. The piece-by-piece analysis of the beached resin pellets suggested that discolored and/or fouled PE resin pellets are suitable for the application of marine plastic resin pellets to pollution monitoring, because they had relatively higher concentration of PCBs.

### **Regional variability in PCB concentrations in plastic resin pellets stranded on 47 beaches in Japan.**

To examine the utility of marine resin pellets as monitoring indicators, we studied the regional variability in PCB concentrations in plastic resin pellets stranded on 47 beaches in Japan. The resin pellets were collected from 47 beaches (Figure 9) in 2001 and 2002 in partnership with local volunteers. The samples were collected from sandy beaches. More than 100 pellets were collected from each beach. Solvent-rinsed stainless steel tweezers were used to pick up pellets, and the samples were stored in a stainless steel container and sent to the laboratory by mail. The resin pellets were classified as PE, PP, or other by near-infrared spectroscopy (PlaScan-SH, OPT Research Inc. Tokyo, Japan). PE pellets from each beach were sorted according to discoloration (yellowing) and fouling. Pellets with being discolored and/or fouled were analyzed for PCBs. Five discolored pellets or 10 fouled pellets were pooled (grouped) and extracted as below, because at least 5 – 10 pellets were necessary to extract enough amounts of PCBs for reliable analysis of pellets collected from less polluted area than Tokyo Bay. Multiple pools (groups) from each location were analyzed so that regional PCB concentrations could be shown as a range, and any high-concentration pellets could be recognized and excluded from the mean. The pellets were extracted by maceration in n-hexane for 72 h at 20 °C. The solvent was replaced twice during the maceration, and all the extracts were combined. The clean-up procedure and instrumental analysis were as described above.

While wide ranges of PCB concentrations were recorded from the individual beaches, regional differences can be also recognized among the beaches (Endo et al., 2005). As can be seen in Figure 10, high PCB concentrations were most frequently found in the pellets from Tokyo Bay and its vicinity and Osaka Bay, whereas low PCB concentrations were found in the Tohoku and Hokkaido region, the region around the Sea of Japan, and the Pacific Ocean side of Shikoku. This regional difference in PCBs concentrations in the pellets is consistent with a spatial trend of PCB pollution in other environmental media. Tokyo Bay and Osaka Bay are surrounded by the most industrialized areas in Japan, where large amounts of PCBs were used in the past (i.e., 1960s), polluting the bays and the vicinities with PCBs. In these regions, high concentrations of PCBs are still present in bottom sediments, seawater, and marine organisms (Japan Coast Guard, 2003; Yamaguchi et al., 2000). On the other hand, the Tohoku and Hokkaido region, the Sea of Japan region, and the Pacific Ocean side of Shikoku Island are less industrialized, and lower concentrations of PCBs have been reported in sediments from these areas (Japan Coast Guard, 2003).

In addition to regionally appropriate concentrations, some sporadic high concentrations of PCBs

were found in resin pellets from remote islands such as Tanegashima, Shikinejima, and Sadogashima (locations 1, 12, and 42, respectively, in Figure 10). Their concentrations are remarkably higher than others from the same regions. This occurrence indicates that contaminant concentrations in ambient seawater are not the only factor that determines the concentration in resin pellets, and that some other factors should be also taken into account. A long residence time on the ocean surface or the presence of certain additives may have facilitated the adsorption of PCBs from the ambient seawater. In addition, during their time on the sea, the pellets may have passed close to an industrial area where they may have sorbed high amounts of PCBs, although their final stranding points were remote. Although the cause of these sporadic high concentrations is still unclear, the occurrence of resin pellets with high PCB concentrations on remote islands may pose ecotoxicological problems to marine organisms. Organisms living on the remote islands are normally exposed to low concentrations of PCBs through seawater and the food web. Resin pellets with extremely high concentrations of PCBs could become the dominant exposure route of the organisms to PCBs.

For the application of the resin pellets to pollution monitoring, the pellets with sporadic high concentrations of PCBs can be excluded through calculating median of multiple pools (groups), as shown in Figure 11. Calculating median successfully eliminated the sporadic high concentration of PCBs found at st. 1, 12, and 42. The regional pattern in PCBs concentrations in the beached resin pellets was compared with those for mussels (indicator organisms). Mussels were collected from 24 locations over Japan in 2001 – 2003 and were analyzed for PCBs. The results of our mussel samples echo this regional trend of PCBs in the beached resin pellets in Japan (Figure 11). Figure 11 indicates that the distributions of the concentrations in pellets and mussels are consistent. Through selecting 11 locations where pellets and mussels were collected within 10 km distance, correlation of PCB concentrations in the pellets with those in the mussel was examined. Figure 12 also indicates that PCBs concentrations in PE pellets (median of multiple bulks of discolored/fouled pellets) had good correlation with those in mussels. Considering that mussels well reflect the concentrations of hydrophobic organic contaminants in seawater and are therefore used widely around the world for marine pollution monitoring, we can conclude that the degree of pollution in the ambient environment is one of the most important factors that determine PCB concentrations in resin pellets.

## **Call for Beached Resin Pellets**

### ***Pellet Watch* : Global Monitoring of Persistent Organic Pollutants (POPs) using Beached Plastic Resin Pellets**

Based on above discussions, we propose the use of beached resin pellets as a tool to monitor persistent organic pollutants (e.g., PCBs and organochlorine pesticides) pollution in world coastal waters, as illustrated in Fig.13. In this project, we call for beached plastic resin pellets. Through various networks and internet, we will ask world citizens to collect beached resin pellets at nearby beaches and send them to our laboratory via air mail. More than 100 resin pellets (ideally 200 pellets) per a location are necessary for reliable monitoring. The collected pellets will be sent to our laboratory in Tokyo via Air mail. Discolored and/or fouled PE pellets will be selected by using spectroscopic method and multiple 10-piece-pools will be extracted and analyzed for organochlorine compounds (e.g., PCBs) by the method that we developed. Based on the analytical results, global POPs pollution will be mapped. The analytical results will be sent to the persons who collected the

samples via internet and released on web. Advantage of *Pellet Watch* includes extremely low cost for sampling and shipping and easiness to collect samples (i.e., no special training is required for sampling) compared with the existent monitoring media (e.g., water, sediment, biological samples such as mussel). Collect pellets on your nearby beach and send 100 pellets to Tokyo via air mail!

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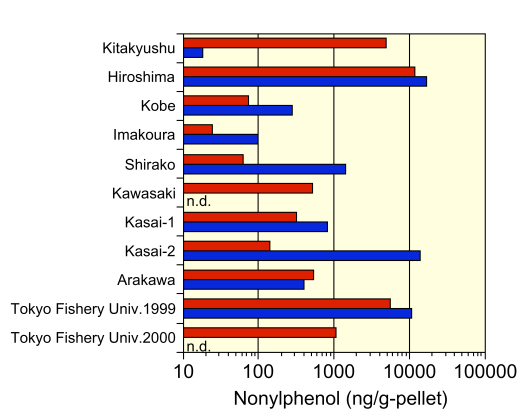


Fig.1 Nonylphenol concentrations in beached resin pellets on Japanese coasts

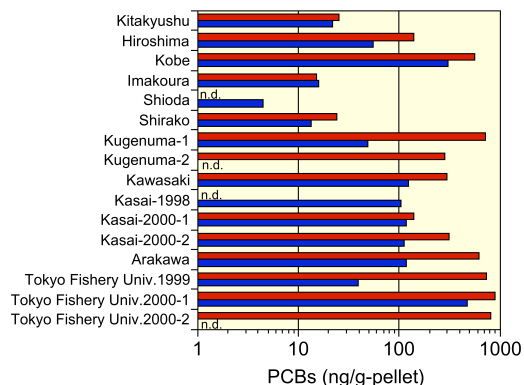


Fig.3 PCB concentrations in beached resin pellets on Japanese coasts

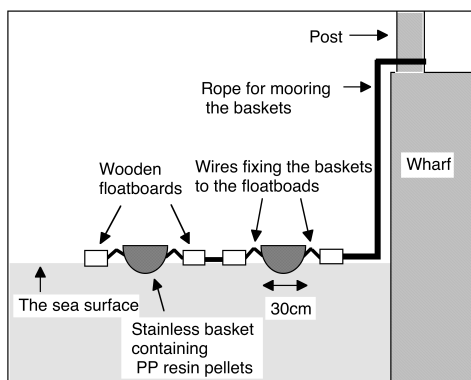


Fig.4 Schematic illustration of adsorption experiment

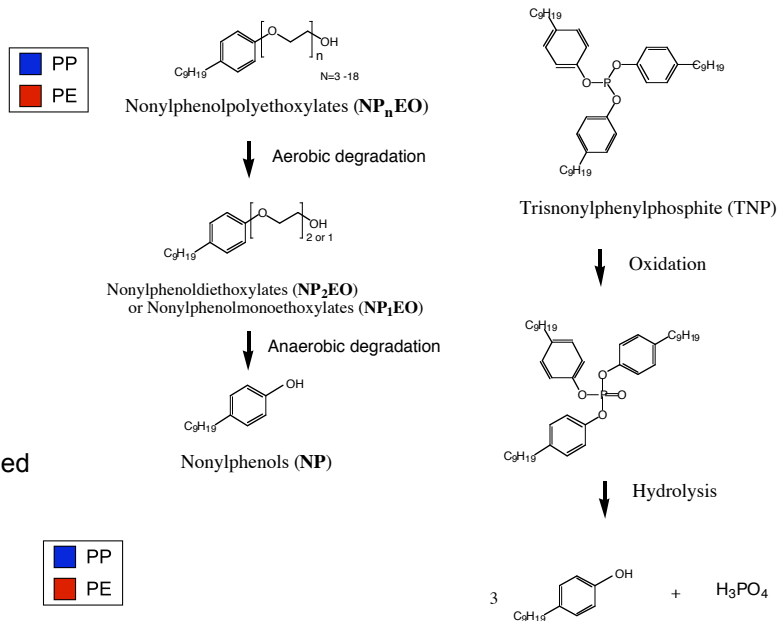


Fig. 2 Nonylphenol derived from plastic additives

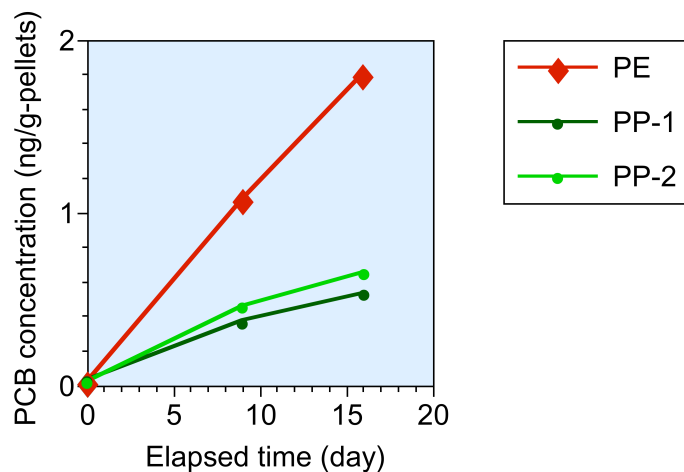


Fig.5 Concentration of PCBs in deployed plastic pellets during the field adsorption experiment



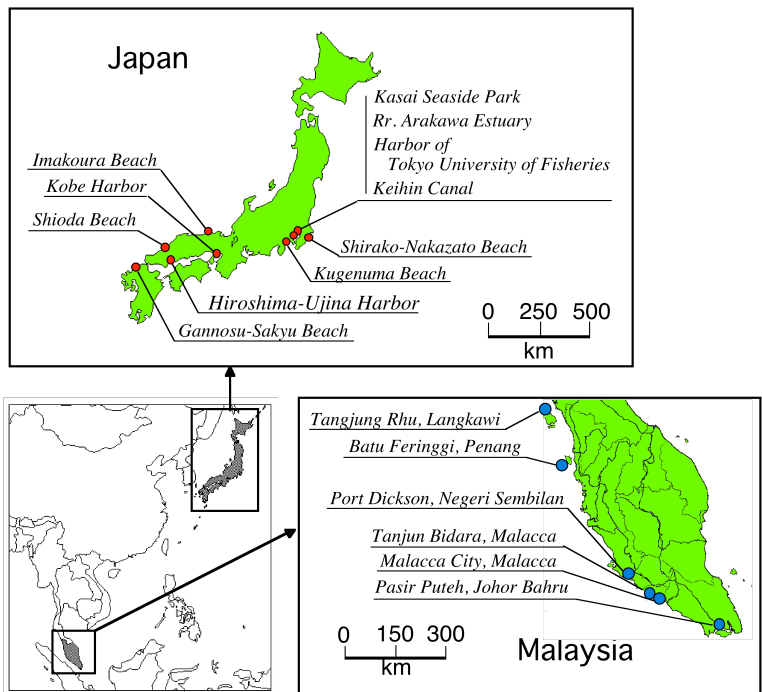


Fig.6 Sampling locations of plastic resin pellets and mussels in 1998 - 2000

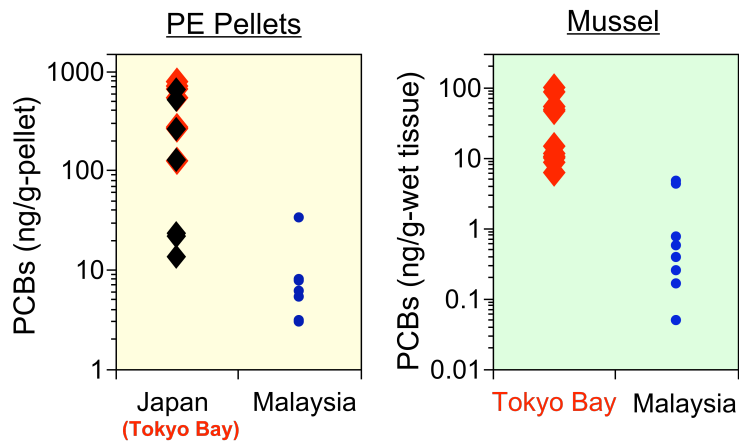


Fig.7 PCB concentrations in beached plastic resin pellets (left) and mussels (right).  
 Red diamonds : Tokyo Bay; black diamonds : the other Japanese locations than Tokyo Bay;  
 blue circles : Malaysia

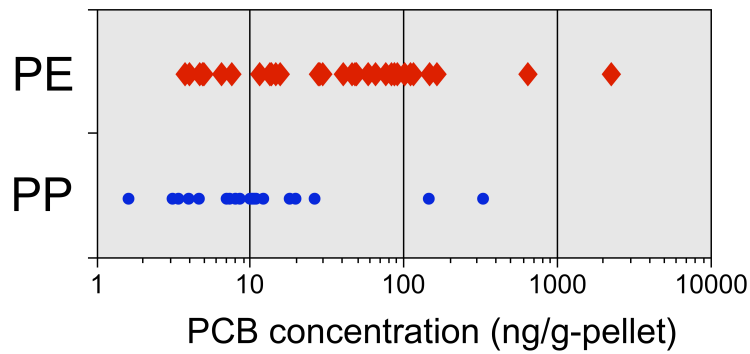


Fig.8 PCBs concentration in PE and PP resin pellets derived from piece-by-piece analysis.

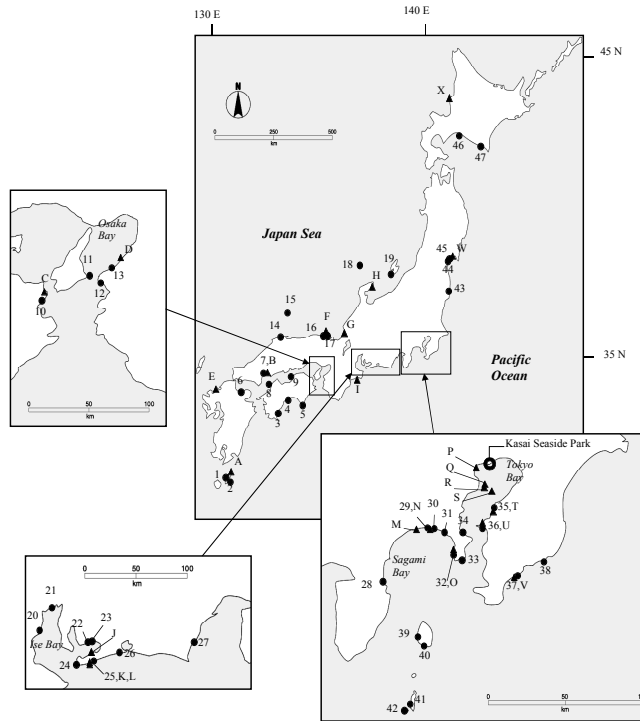


Fig.9 Sampling locations of plastic resin pellets and mussels in 2001 – 2003  
 Numbers : locations of the resin pellets; alphabets : locations for mussels

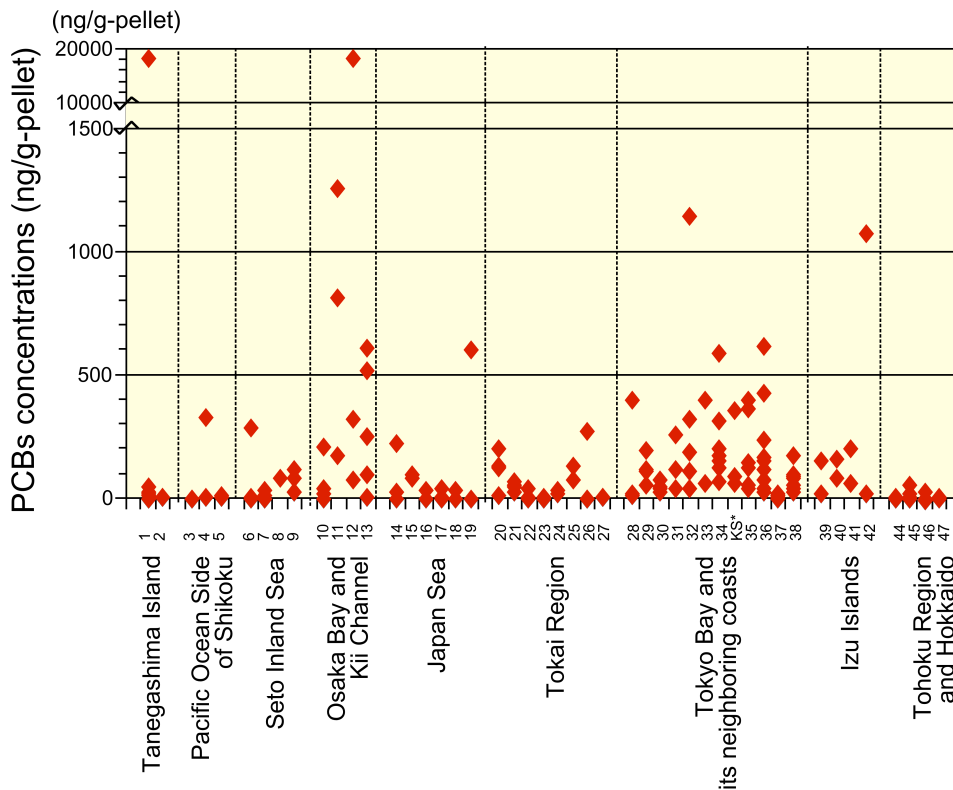


Fig.10 PCBs concentration in discolored and/or fouled PE resin pellets from 47 Japanese beaches.  
 Each pool is shown as a plot.

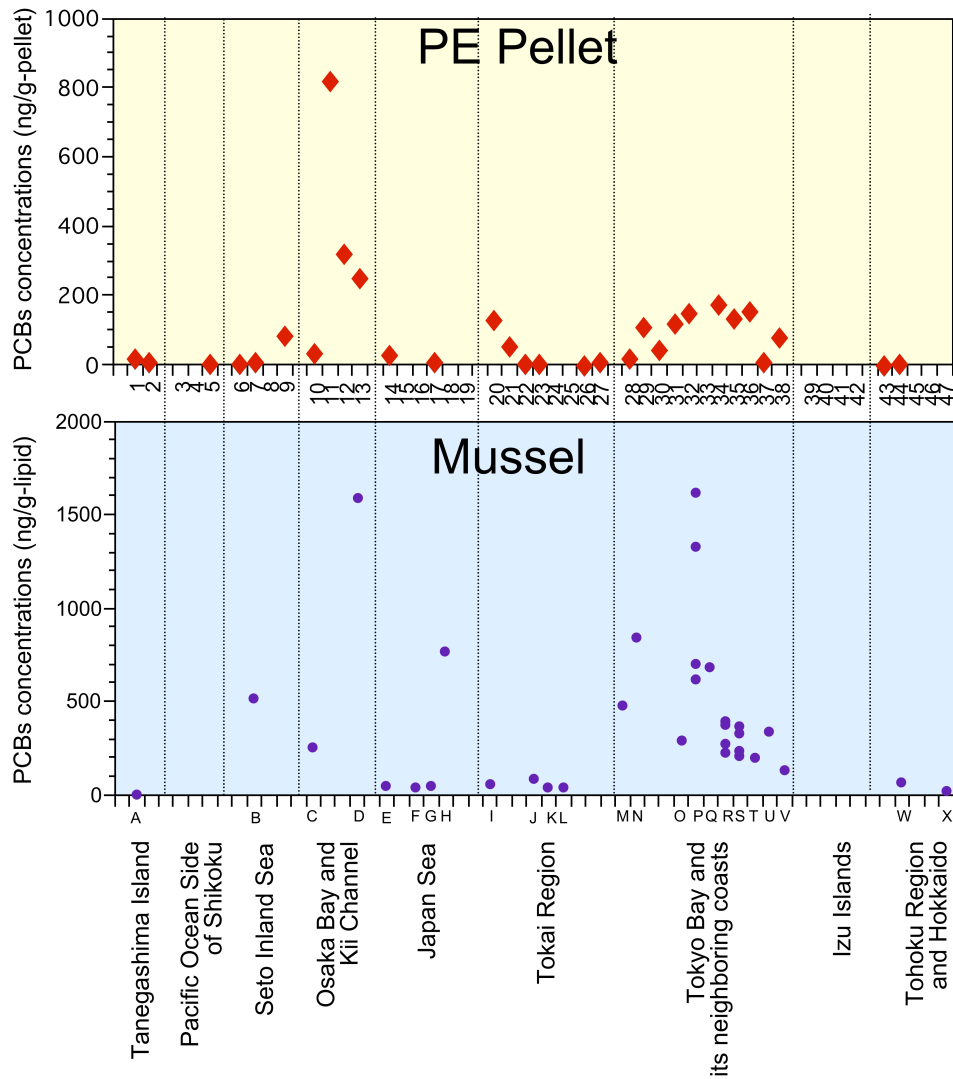


Fig.11 PCBs concentration in discolored and/or fouled PE resin pellets from 47 Japanese beaches and in mussels from 24 Japanese coasts. Median concentrations are plotted for PE pellets.

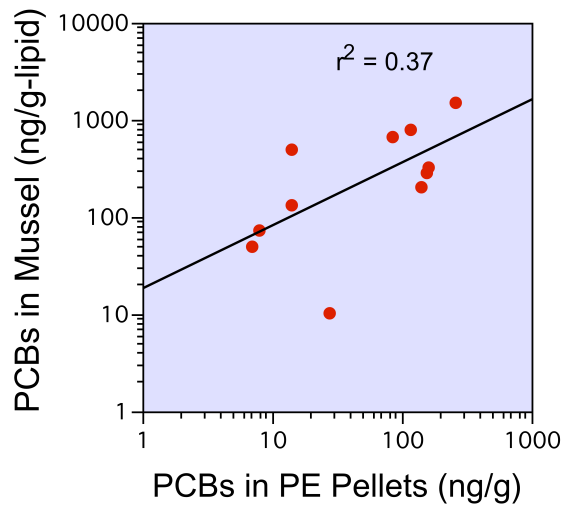
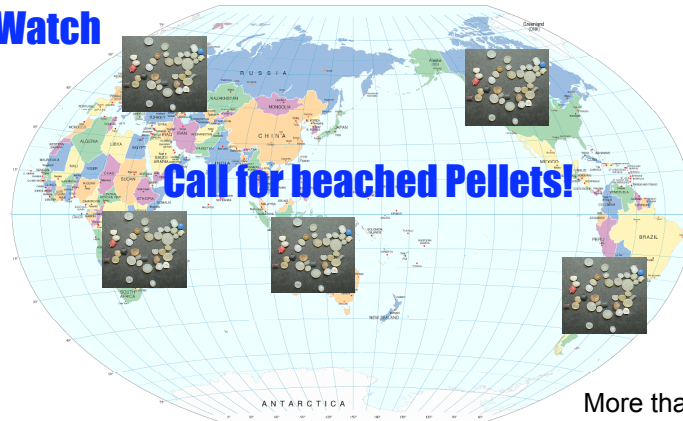


Fig.12 Relationship between median concentrations of PCBs in discolored and/or fouled PE resin pellets and PCBs concentrations in mussels.

# Global Monitoring of Persistent Organic Pollutants (POPs) Using Beached Plastic Resin Pellets

## Pellet Watch



More than 100 pieces  
(~ 200 pieces)  
per one location



Laboratory of Organic Geochemistry, Dr. Hideshige Takada,  
Tokyo University of Agriculture and Technology,  
Fuchu, Tokyo 183-8509, Japan



More than 100 pieces (~200 pieces)  
per one location

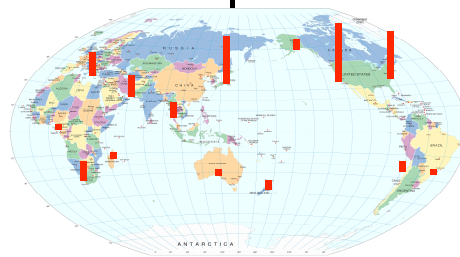
### Sorting

PE, discolored or fouled

### Analysis for POPs (PCBs, organochlorine pesticides)

by GC-ECD, GC-MS  
more than 3 pools of 10 pellets  
to exclude sporadic high concentration

### Mapping POPs pollution



Example of expected map of  
PCBs in beached plastic pellets

- Sending the data via Internet to the collectors
- Releasing the results on web

Fig.13 *Pellet Watch* project: Call for beached pellets