Geochemical Perspectives Letters

Toxic anthropogenic pollutants reach the deepest ocean on Earth

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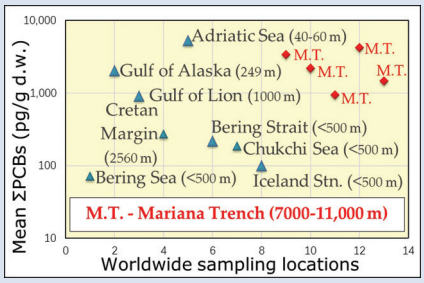
Abstract



Persistent organic pollutants (PCBs and PBDEs) were analysed in sediment core samples (0-2 cm) from the southern Mariana Trench at water depths of 7000-11000 m. ∑PCBs concentrations ranged from 931 to 4195 pg/g, far higher than those recorded before in marine sediments from shallower depths. Toxic Equivalence (TEQ) of dl-PCBs ranged from 0.650 – 14.9 pg/g, which is higher than most marine surficial sediments at <500-2500 m ocean depth, recovered from semi-industrial to industrial areas. However, $\sum_8$PBDEs values (averaging ~136 pg/g) were lower than those in surficial sediments from shelf areas recorded in past studies. Evidently, anthropogenic pollutants have reached the deepest realm on Earth, and the Mariana Trench acts as a repository for POPs amplification. The high concentration of PCBs is an eye-opener, which is directly affecting our deep sea ecosystems, considering their pervasiveness and persistence in trench sediments.

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Letter

The deep ocean arguably acts as the largest potential sink for discarded pollutants (Dachs *et al.*, 2002). However, the hadal zone (~6000 to 11000 m deep), which represents the deepest ocean on Earth, has largely been unexplored due to its remoteness. Of all the toxic anthropogenic pollutants damaging the oceans, persistent organic pollutants (POPs) are of particular concern due to their robust residence time in the environment (Kukučka *et al.*, 2015), global transport through atmospheric and oceanic currents (Wania and Mackay, 1996), and their ability to bioaccumulate in marine foodwebs (Lohmann *et al.*, 2007), resulting in organism endocrine disruption (Rhind, 2012) and other adverse health effects. Studies revealing presence of POPs in surface to deep marine sediments are plentiful (*e.g.*, Iwata *et al.*, 1994; Ma *et al.*, 2015; Combi *et al.*, 2016; Neira *et al.*, 2018), but none of them probed beyond the continental shelf area, leaving the real "depth" of the oceans practically unexplored. In a latest study, Jamieson *et al.* (2017) reported high concentrations of POPs in endemic amphipod fauna from the Mariana and Kermadec Trenches. The signature of these pollutants through bioaccumulation could, however, be markedly different from that residing in the sediments.

The Mariana Trench is located in the western Pacific Ocean, where the Pacific Plate subducts beneath the Mariana and Philippine Sea Plates at convergence rates of 4–8 cm/yr. The southern Mariana Trench encloses the deepest point on the Earth's surface-the Challenger Deep, which is 11034 m down, and 2 km deeper than the average depth along the axis of the Mariana Trench (Fujioka *et al.*, 2002).

We detected polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in five sediment samples recovered from the southern Mariana Trench at ocean depths of 6980 m (C1-I-M-S015B02), 8638 m (C1-I-M-S078B10), 9373 m (TS03-S106GT02 (S) and TS03-S106GT02 (D) for top and bottom surface of the core respectively), and 10908 m (TS03-S090LANDER11), (Fig. 1), during TS03 Hadal Trench Cruise carried out *via* R/V Tansuo Yihao on June, 2017 (See Table S-1 for sampling location details).

The prominent finding was that, 36 PCB and 10 PBDE congeners (Figs. 2 and 3 respectively) were detected. Lower-chlorinated congeners such as CB-8, CB-37, CB-52 and CB-60 were most abundant (refer to Table S-2), and their concentrations ranged from 1460 to 3300 pg/g. For PBDEs, concentrations ranged from 245 (TS03-S106GT102 (S)) to 591 pg/g (TS03-S090LANDER11) (Table S-3). Low and medium weight PBDEs, such as BDE-47 and BDE-153 were more common in the samples. However, higher weight BDEs 207 and 208 were detected in C1-I-M-S015B02 and TS03-S090LANDER11, with high concentrations of 266 and 280 pg/g respectively.

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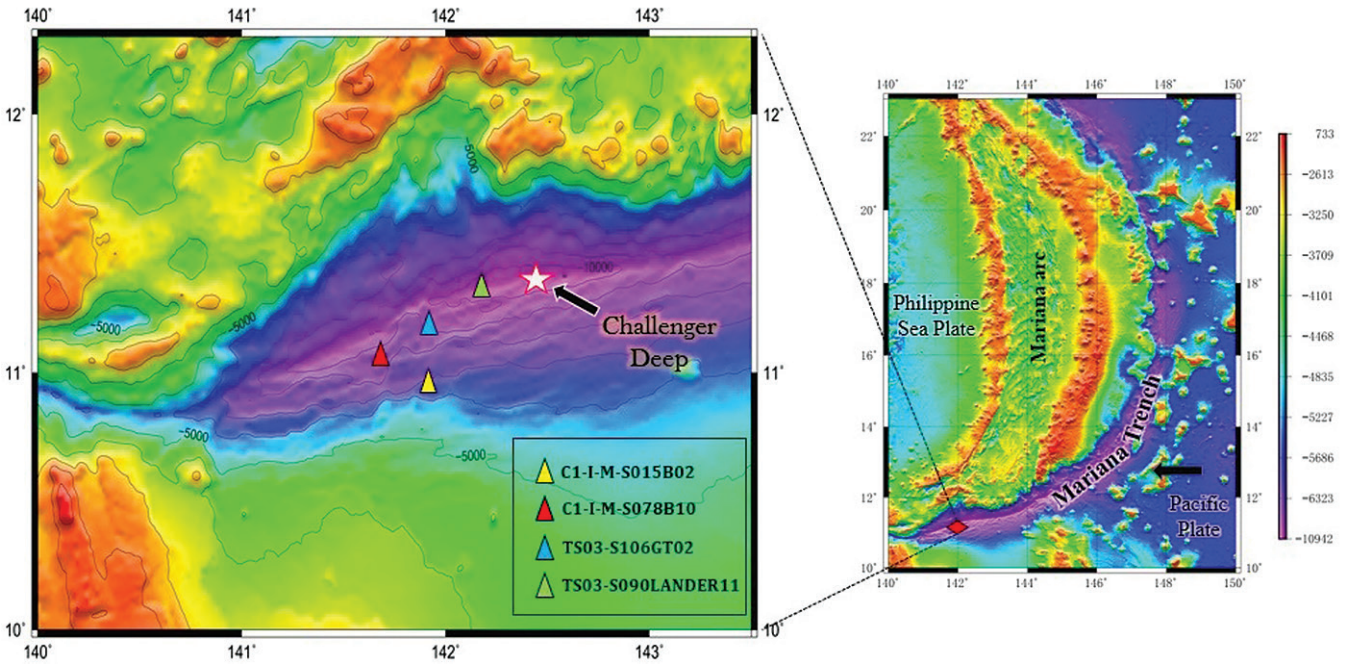

Figure 1 Sampling location map of Mariana Trench sediments.

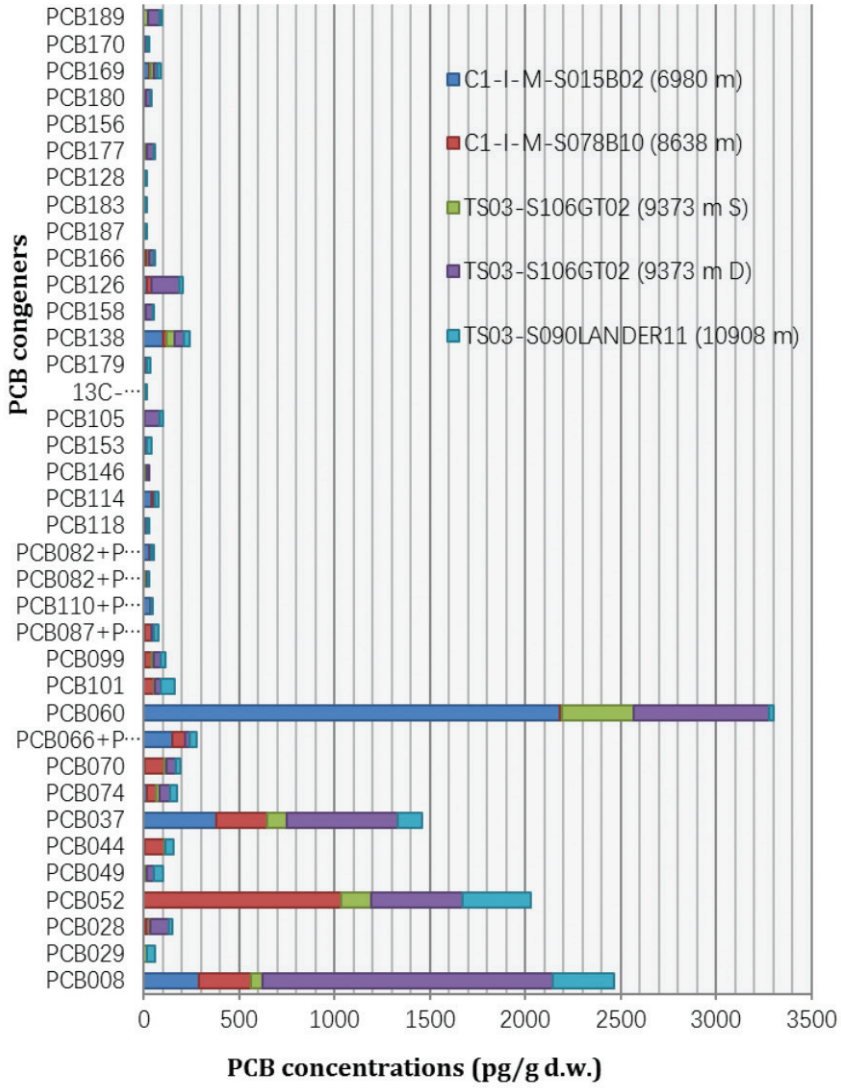

Figure 2 Concentrations of PCB congeners (pg/g d.w.) for different sampling locations at various water depths, as represented by coloured stacked bars. Refer to Table S-2 for details.

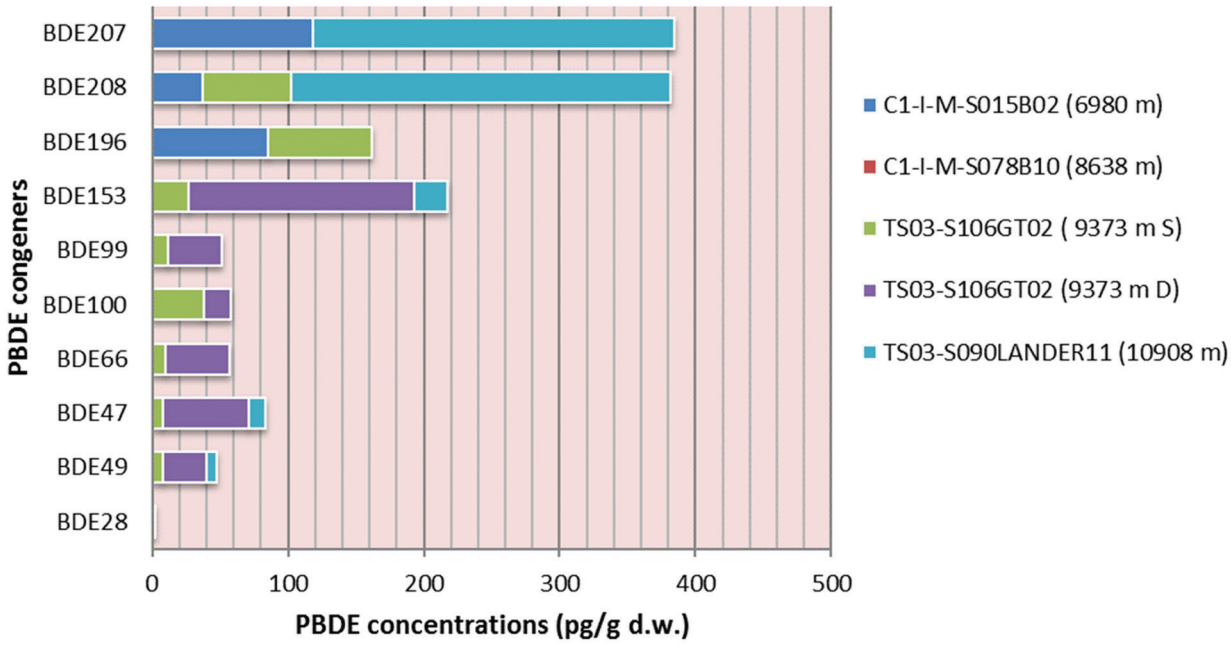

Figure 3 Concentrations of PBDE congeners (pg/g d.w.) for different sampling locations at various water depths, as represented by coloured stacked bars. Refer to Table S-3 for details.

∑PCBs (pg/g) concentrations ranged from 931-4195 pg/g dry weight in all five sediment samples. The Mariana sediments showed higher levels of low- and medium-chlorinated PCB congeners. It is noteworthy that no clear trend of ∑PCB variation with water depth was observed (Fig. S-1).

The seven "indicator PCBs" (Table S-4) chosen following the International Council for the Exploration of the Sea (ICES) convention contributed to 22 % of all PCBs. They showed highest Σ_7PCBs values at depths of 8638 m and 9373 m. CB-52 was abundant in most of the samples, except in C1-I-M-S015B02, where it was not detected. These PCBs are indicative of lipophilic contaminants, but their concentrations can vary with the contaminated source (Kim *et al.*, 2004). High concentrations of low-chlorinated CBs, such as CB-52 may be from processes using chlorine oxidation; further, high-chlorinated CBs could be decayed to lighter ones by bacterial decomposition (Kim *et al.*, 2004). Presence of mid-chlorinated CBs, such as Penta-CB (CB-101) and Hexa-CBs (CB-138 and CB-153) could be related to partitioning of these compounds to particulate organic matter. High-chlorinated PCBs, such as CB-180 possibly came from terrestrial pollution in the form of industrial sewage, or from marine traffic (Hong *et al.*, 2005). Although no pattern in concentration *vs.* water depth was evident, deeper trench samples (C1-I-M-S078B10, TS03-S106GT02 (S), TS03-S106GT02 (D), and TS03-S090LANDER11) exhibited higher Σ_7PCBs (average 642.2 pg/g) than C1-I-M-S015B02 (129 pg/g) values.

Toxic potency of PCBs was assessed by measuring Toxic Equivalence (TEQ) of twelve dioxin-like (dl-) CB congeners (CB-77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 166, and 189) in all the sediment samples (normalised by multiplying their measured concentrations by the appropriate WHO-TEFs) (Table S-5). The respective TEQ_{PCBs} express these analyte concentrations as a single number, which is equivalent to toxicity derived exclusively from 2,3,7,8-TCDD. Mariana dl-PCB-TEQ concentrations ranged from 0.650 – 14.9 pg/g (Table S-6), with the highest TEQ_{PCBs} recorded at 9373D. Our results are higher than TEQ_{PCBs} values from past studies with sediments from shallow water depths, *e.g.*, a semi-industrial area, Asaluyeh Harbor, Iran (0.001-3.4) (Arfaeinia *et al.*, 2017); marine sediments from Mediterranean Sea, Catalonia, Spain (0.03-24.8) (Eljarrat *et al.*, 2001); or Han River, Korea (0.0118-0.626) (Kim *et al.*, 2009). Surface marine sediments in shallow water are susceptible to organic pollution due to their proximity to industrial areas, as well as from atmospheric interactions. It is however, surprising to witness such contaminants have reached the deepest parts on Earth.

Unlike PCBs, concentrations of PBDEs were lower in the Trench samples. To maintain consistency (since PBDEs are presented as mixtures of congeners), and form a comparable dataset, a total of 8 PBDEs (BDE-28, 47, 99, 100, 153, highlighted in blue in Table S-3; BDE-54, 183, and 209 were not detected) were chosen (Zhang *et al.*, 2016), which commonly occur in environmental samples. Their concentrations ranged from 36-289 pg/g. Mid-brominated congener BDE-153 was the most abundant, with concentrations ranging from <LOD-166.0 pg/g d.w. (mean 43.3 pg/g). Penta-BDE mixture components BDE-47 and 99 possibly originated terrestrially from polyurethane foams (Yogui and Sericano, 2009), and were found at concentrations of <LOD 63.3 pg/g d.w. (mean 16.7 pg/g) and <LOD 40.0 pg/g d.w. (mean 10.3 pg/g) respectively. The PBDE congener distribution of Mariana sediments resembles those reported elsewhere in surface marine sediments from Europe (de Wit, 2002) and in Qingdao coastal sea sediments (Pan *et al.*, 2007), China where BDEs 47, 99, and 153 were the most frequently reported congeners. Our results indicate that penta-BDEs could possibly be transported with water in the soluble and particle phases and degradation and fractionation of higher-brominated congeners (such as PBDE 209) may occur during long range transportation. Σ_8PBDEs values (range <LOD 0.289 ng/g, mean 0.082 ng/g) (Table S-3) for Mariana Trench sediments are lower than those in shallow water surface marine sediments recorded in past studies. Looking at the research carried in San Francisco Bay sediments, the Σ_8PBDEs in each sample area (Suisun Bay, San Pablo Bay, Central Bay, and South Bay) range from 2.46-5.14 ng/g (Klosterhaus *et al.*, 2012). In China, samples collected from offshore sediment of northern South China Sea, Σ_8PBDEs is 0.93 ng/g and from East China Sea it is below 1 ng/g (Liu *et al.*, 2015). The salient finding is that PBDEs were detected in the deepest Trench sediments with concentrations lower than, or nearing those of, coastal surface sediments adjacent to industrial areas.

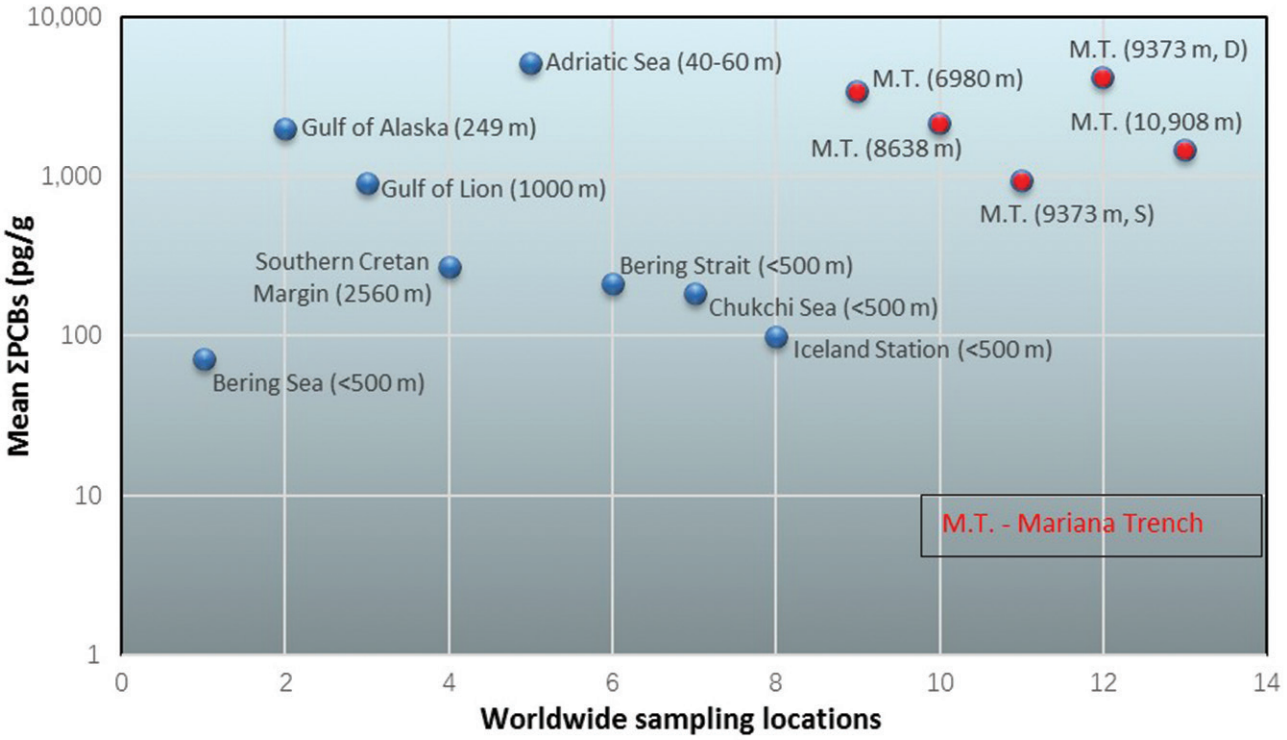

Figure 4 Comparison of ∑PCBs concentrations in the Mariana Trench sediments with other worldwide marine surface water sediments. Figures in brackets indicate water depth.

We compared the level of ∑PCB concentrations in the Mariana Trench sediments to those reported in past studies at shallower depths (Fig. 4). The overall ∑PCB values from our study (range: 931-4195 pg/g, mean 2424 pg/g) are overwhelmingly higher than those from shallow marine sediments studied in the past (Table S-7); and higher than the world baseline levels for ∑PCBs arising from atmospheric transport found in clean coastal sediments, which is placed at <1 ng/g d.w. (Phillips, 1986). However, the PCB concentrations in the sediments are markedly lower than those found in endemic amphipod fauna through bioaccumulation from ~7000-10000 m in the Mariana (Jamieson *et al.*, 2017), since these organisms have been known to rapidly locate and consume any particulate organic matter (POM) from surface-derived carrion falls. Further, pollutants can accumulate in the wax esters of capacious guts of larger amphipods, which are used as energy reserves in times of prolonged food deprivation (Lee *et al.*, 2006).

Hadal communities, such as Hirondellidae tend to accumulate along the trench axis, where gravity driven down-slope transport of sediments results in a nutrient-rich environment (Ichino *et al.*, 2015). However deep sea lysianassoid amphipods are also recorded from shallower trench depths of nutrient poor environments, thought to be advantageous due to limited competitive interactions (Blankenship and Levin, 2007). Post-mortal discharge of pollutants bioaccumulated by these endemic species and stored in the wax esters, or ingestion and faecal release may account for the high concentration of PCBs and PBDEs in the Mariana sediments. In addition, remote trenches encounter seafloor landslides or earthquakes. The funnel-like shape and high fluid dynamics within the Trench favour accumulation of pollutants along the trench axis (Turnewitsch *et al.*, 2014), associated with these geotectonic events. However, apart from slightly higher organic carbon content in the Challenger Deep (0.3-0.4 %) (Glud *et al.*, 2013), hadal sediments are more depleted in total organic carbon (TOC), often reaching as low as 0.22 % (Luo *et al.*, 2017). Therefore, the dominance of organic matter-associated POPs in deep sea sediments over surface marine sediments seems paradoxical. We suspect that clay minerals may play a major role in the adsorption of contaminant particles in hadal sediments. Due to the lack of available studies, we conducted a preliminary mineralogical investigation of three sediment samples from deepest points by X-ray diffraction (XRD). Our results indicate clay assemblages (illite, nontronite, clinochlore, and gismondine) are abundant in the deepest sediments (Fig. S-2), with their total relative concentrations ranging from 51.8 to 86.5 % in the three samples (Table S-8) A number of possible mechanisms of sorption of POPs with clay minerals are explained in previous studies (*e.g.*, Li *et al.*, 2015). For example, hydrogen bonding is demonstrated to bind polar groups of contaminants and basal oxygen atoms or adsorbed water of clay minerals (Wang *et al.*, 2011). Binding of POPs in surface or interstitial layers of clays could act towards the agglomeration of pollutants in the hadal sediments. Further, long range transport of these pollutants could be linked to allogenic clays from terrestrial or shallow water origins.

The present discovery serves as evidence as to how far man-made pollutants have reached; and any such contamination will have ecological and toxic effects, long-term or ephemeral, depending on the scale of impact. Possibly, there no longer exists "pure land" that can completely be isolated from human activities in the Earth's ocean. High concentrations of pervasive pollutants in the trench sediments also imply that the Trench is a repository for POPs amplification, which occurs regardless of the source of these toxic anthropogenic pollutants. Certainly, high concentrations of POPs in the deepest ocean sediments are directly affecting hadal ecosystems, considering their persistent nature. The immediate challenge, therefore, is to assess the impact of anthropogenic pollutants reaching or residing in the deepest ocean.

More detailed spatial and ecotoxicological studies on trench sediments will better our understanding of effects of POPs and other pervasive pollutants such as microplastics and litter in such environment.

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Additional Information

Supplementary Information accompanies this letter at http://www.geochemicalperspectivesletters.org/article1814.

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References

Arfaeinia, H., Asadgol, Z., Ahmadi, E., Seifi, M., Moradi, M., Dobaradaran, S. (2017) Characteristics, distribution and sources of polychlorinated biphenyls (PCBs) in coastal sediments from the heavily industrialized area of Asalouyeh, Iran. *Water Science and Technology* 76, 3340-3350.

Blankenship, L.E., Levin, L.A. (2007) Extreme food webs: foraging strategies and diets of scavenging amphipods from the ocean's deepest 5 km. *Limnology Oceanography* 52, 1685-1697.

Combi, T., Miserocchi, S., Langone, L., Guerra, R. (2016) Polychlorinated biphenyls (PCBs) in sediments from the western Adriatic Sea: Sources, historical trends and inventories. *Science of the Total Environment* 562, 580-587.

Dachs, J., Lohmann, R., Ockenden, W.A., Mejanelle, L., Eisenreich, S.J., Jones, K.C. (2002) Oceanic biogeochemical controls on global dynamics of persistent organic pollutants. *Environmental Science and Technology* 36, 4229-4237.

de Wit, C.Y., (2002) An overview of brominated flame retardants in the environment. *Chemosphere* 46, 583-624.

Eljarrat, E., Caixach, J., Rivera, J. (2001) Toxic potency of non- and mono-ortho PCBs, PCDDs, PCDFs, and PAHs in Northwest Mediterranean sediments (Catalonia, Spain). *Environmental Science and Technology* 35, 3589-3594.

Fujioka, K., Okino, K., Kanamatsu, T., Ohara, Y. (2002) Morphology and origin of the Challenger Deep in the Southern Mariana Trench. *Geophysical Research Letters* 29, doi: 10.1029/2001GL013595.

Glud, R.N., Wenzhöfer, F., Middelboe, M., Oguri, K., Turnewitch, R., Canfield, D.E., Kitazato, H. (2013) High rates of microbial carbon turnover in sediments in the deepest oceanic trench on Earth. *Nature Geoscience* 6, 284-288.

Hong, S.H., Yim, U.H., Shim, W.J., Oh, J.R. (2005) Congener-specific survey for polychlorinated biphenyls in sediments of industrialized bays in Korea: regional characteristics and pollution sources. *Environmental Science and Technology* 39, 7380-7388.

Ichino, M.C., Clark, M.R., Drazen, J.C., Jamieson, A., Jones, D.O.B., Martin, A.P., Rowden, A.A., Shank, T.M., Yancey, P.H., Ruhl, H.A. (2015) The distribution of benthic biomass in hadal trenches: a modeling approach to investigate the effect of vertical and lateral organic matter transport to the seafloor. *Deep-Sea Research I* 100, 21-33.

Iwata, H., Tanabe, S., Aramoto, M., Sakai, N., Tatsukawa, R. (1994) Persistent organochlorine residues in sediments from the Chukchi Sea, Bering Sea and Gulf of Alaska. *Marine Pollution Bulletin* 28, 746-753.

Jamieson, A.J., Malkocs, T., Piertney, S.B., Fujii, T., Zhang, Z. (2017) Bioaccumulation of persistent organic pollutants in the deepest ocean fauna. *Nature Ecology and Evolution* 1, 0051.

Kim, K.S., Lee, S.C., Kim, K.H., Shim, W.J., Hong, S.H., Choi, K.H., Yoon, J.H., Kim, J.G. (2009) Survey on organochlorine pesticides, PCDD/Fs, dioxin-like PCBs and HCB in sediments from the Han river, Korea. *Chemosphere* 75, 580-587.

Kim, M.K., Kim, S., Yun, S., Lee, M., Cho, B., Park, J., Son, S, Kim, O. (2004) Comparison of seven indicator PCBs and three coplanar PCBs in beef, pork, and chicken fat. *Chemosphere* 54, 1533-1538.

Klosterhaus, S.L., Stapleton, H.M., La Guardia, M.J., Greig, D.J. (2012) Brominated and chlorinated flame retardants in San Francisco Bay sediments and wildlife. *Environmental International* 47, 56-65.

Kukučka, P., Audy, O., Kohoutek, J., Holt, E., Kalábová, T., Holoubek, I., Klánová, J. (2015) Source identification, spatio-temporal distribution and ecological risk of persistent organic pollutants in sediments from the upper Danube catchment. *Chemosphere* 138, 777-783.

Lee, R.F., Hagen, W., Kattner, G. (2006) Lipid storage in marine zooplankton. *Marine Ecology Progress Series* 307, 273-306.

Li, Z., Fitzgerald, N.M., Albert, Z., Schnabl, A., Jiang, W.-T. (2015) Contrasting mechanisms of metoprolol uptake on kaolinite and talc. *Chemical Engineering Journal* 272, 48-57.

Liu, L., Li, H., Wang, Z., Liu, R., Zhang, Y., Lin, K. (2015) Insights into spatially and temporally co-occurring polybrominated diphenyl ethers in sediments of the East China Sea, *Chemosphere* 123, 55-63.

Lohmann, R., Breivik, K., Dachs, J., Muir, D. (2007) Global fate of POPs: current and future research directions. *Environmental Pollution* 150, 150-165.

Luo, M., Gieskes, J., Chen, L., Shu, X., Chen, D. (2017) Provenances, distribution, and accumulation of organic matter in the southern Mariana Trench rim and slope: Implication of carbon cycle and burial in hadal trenches. *Marine Geology* 386, 98-106.

Ma, Y., Halsall, C.J., Crosse, J.D., Graf, C., Cai, M., He, J., Gao, G., Jones, K. (2015) Persistent organic pollutants in ocean sediments from the North Pacific to the Arctic Ocean. *Journal of Geophysical Research: Oceans* 120, 2723-2735.

Neira, C., Vales, M., Mendoza, G., Hoh, E., Levin, L.A. (2018) Polychlorinated biphenyls (PCBs) in recreational marina sediments of San Diego Bay, southern California. *Marine Pollution Bulletin* 126, 204-214.

Pan, J., Yang, Y-L., Xu, Q., Chen, D-Z., Xi, D-L. (2007) PCBs, PCNs and PBDEs in sediments and mussels from Qingdao coastal sea in the frame of current circulations and influence of sewage sludge. *Chemosphere* 66, 1971-1982.

Phillips, D.J. H. (1986) Use of organisms to quantify PCBs in marine and estuarine environments. In: Waid, J.S. (Ed.) *PCBs and the Environment Vol. II.* CRC Press, Boca Raton, FL., 127-181.

Rhind, S.M. (2012) Anthropogenic pollutants- an insidious threat to animal health and productivity? *Acta Veterinaria Scandinavica* 54, S2.

Turnewitsch, R., Falahat, S., Stehlikova, J., Oguri, K., Glud, R.N., Middelboe, M., Kitazato, H., Wenzhoefer, F., Ando, K., Fujio, S. (2014) Recent sediment dynamics in hadal trenches: evidence for the influence of higher-frequency (tidal, near-inertial) fluid dynamics. *Deep-Sea Research I Oceanographic Research Papers* 90, 125-138.

Wang, C.J., Li, Z., Jiang, W.T. (2011) Adsorption of ciprofloxacin on 2:1 dioctahedral clay minerals. *Applied Clay Science* 53, 723–728.

Wania, F., Mackay, D. (1996) Tracking the distribution of persistent organic pollutants. *Environmental Science and Technology* 30, 390A-396A.

Yogui, G.T., Sericano, J.L. (2009) Polybrominated diphenyl ether flame retardants in the U.S. Marine environment: a review. *Environment International* 35, 655-666.

Zhang, Y., Wang, W., Song, J., Ren, Z., Yuan, H., Yan, H., Zhang, J., Pei, Z., He, Z. (2016) Environmental characteristics of polybrominated diphenyl ethers in marine system, with emphasis on marine organisms and sediments. *BioMed Research International* 2016, Article ID 1317232, 16 pp, doi: 10.1155/2016/1317232.

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Supplementary Information

The Supplementary Information includes:

- Sampling locations
- Methods
- Tables S-1 to S-8
- Figures S-1 and S-2
- Supplementary Information References

Sampling locations

Table S-1 Sampling details of Mariana Trench sediments.

Sample Name	Location	Water Depth (m)	Core Length (cm)	Coring method	ΣPCBs (pg/g)
C1-I-M-S015B02	10°59.38'N; 141°57.87'E	6980	50	**Box** 	3373
C1-I-M-S078B10	11°13.706'N; 141°41.376'E	8638	50	**Box** 	2157
TS03-S106GT02 (S) TS03-S106GT02 (D)	11°13.5384'N; 142°00.4764'E	9373	216	**Gravity** 	931 4195
TS03-S090LANDER11	11°19.4914'N; 142°11.4527'E	10908	25	**Lander system** 	1462

Note: * TS03-S106GT02 (S) and TS03-S106GT02 (D) denotes the top 2 cm (S) and the bottom 2 cm (D) of the same core.

Methods

Extraction and clean-up

Sediments for analysis were scooped up carefully from the 0-2 cm surface of the cores, avoiding any contamination that may result from contact with the walls. Samples were wrapped in pre-burned Al-foil and transferred to 4 °C, without any exposure to plastics. Sediment samples were homogenised and freeze-dried. The extraction/clean-up procedure for the sediment samples has been described previously (Gong *et al*., 2007). A 10 g sediment subsample was spiked with 2,4,5,6-tetrachloro-m-xylene (TCmX) and decachlorobiphenyl (PCB209), as surrogate standards, and Soxhlet extracted with dichloromethane (DCM) for 24 hours. Anhydrous Na_2SO_4 and activated copper were added for dehydration and desulphurisation, respectively. The extracts were concentrated and solvent-exchanged to hexane by rotary evaporator. The hexane solution was purified by an alumina/silica (1:2, v/v) gel column. The column was eluted with 30 mL of DCM/hexane (2:3, v/v). The final solvent was concentrated to 0.2 mL under a gentle nitrogen stream, and spiked with penta-chloronitrobenzene (PCNB) (purchased from o2si Smart Solutions, SC, United States) as an internal standard for instrument analysis.

Instrumental analysis

Analysis of the PCBs was conducted using an Agilent 7890A gas chromatograph with a 63Ni electron capture detector (GC-μECD) at China University of Geosciences, Wuhan, having a DB-5 capillary column (30.0 m×320 μm×0.25 μm film thickness). Nitrogen was used as carrier gas at 2.5 mL/min under constant-flow mode. Injector and detector temperatures were maintained at 290 °C

and 300 °C, respectively. 2.0 μL of each sample was injected into the GC-μECD for analysis. The oven program was as follows: start at 100 °C (with an equilibration time of 1 min), then to 200 °C at a rate of 4 °C/min, then to 230 °C at 2 °C/min, and finally to 280 °C at 8 °C/min, held at 280 °C for 15 min. Individual PCBs were identified by comparison of their retention times and quantified using internal standard.

For PBDEs, Agilent 6980/5973 Gas Chromatograph-Mass Spectrometer (GC-MS) was used at Jiaxing Tongji Environmental Research Institute, Zhejiang, China. Analytes were separated using DB-5 (0.25 mm i.d. × 15 m; 0.15 um film thickness). The GC conditions were as follows: 1 uL splitless injection at 280 ˚C, He as carrier gas at constant flow rate of 1.0 mL/min, with an oven temperature program of 110 ˚C (1 min hold), 10 ˚C/min to 290 ˚C (20 min hold). Single ion monitoring (m/z of 79 and 81) was used as the scanning mode, with a negative ionisation potential (NCI) detector.

Quality control

Analytical procedures for PCBs were monitored using strict quality control measures, as follows: TCmX and PCB209 were spiked as surrogate standards to judge procedural performance. The surrogate recoveries for TCmX and PCB 209 were 80 % and 114 % respectively. The target compounds were identified by the comparison of their retention times, and quantified by a six-point internal standard calibration curve obtained from the PCB standard reagents with concentration of 200, 150, 100, 50, 20 and 10 μg/L. A procedural blank sample was run to check for interferences and cross contamination. No significant peaks overlapping the PCB standards appeared in the chromatograms of the procedural blanks. No target PCBs were detected in the solvent blank. The results from blank samples were used to establish the limit of detection (LOD). The LOD of each analyte was calculated as the average plus three standard deviations of its blank levels. The limit of detection (LOD) of PCBs ranged from 0.0001-0.0399 ng/g. Actual values of the PCB congeners were calculated by subtracting the respective LOD from the obtained values.

Supplementary Tables

Table S-2 Concentrations of PCBs (pg/g d.w.) in Mariana Trench sediments across all depths.

PCBs	C1-I-M-S015B02	C1-I-M-S078B10	TS03-S106GT02 (S)	TS03-S106GT02 (D)	TS03-S090LANDER11	∑iPCB
CB-8	290	272	63.4	1520	320	2470
CB-29	0	0	19.9	0	39.9	59.8
CB-28	9.00	21.2	6.10	93.8	20.9	151
CB-52	0	1040	155	480	358	2030
CB-49	0.90	5.50	12.2	34.4	470	100
CB-44	3.60	104	4.30	4.70	37.0	154
CB-37	380	268	99.7	583	125	1460
CB-74	20.0	45.8	20.4	54.5	32.2	172.9
CB-70	5	103	15.0	46.9	23.8	194
CB-66+CB-95	150	67.3	2.60	19.9	40.8	281
CB-60	2180	14.1	373	707	28.3	3300
CB-101	0	52.9	7.60	29.6	74.2	164
CB-99	7.00	36.2	8.10	37.3	28.5	117
CB-87+CB-81	4.00	36.2	1.50	13.4	20.2	75.3
CB-110+CB-77	33.0	0	1.20	6.70	7.70	48.6
CB-82+CB-151	0	13.5	5.00	3.80	9.50	31.8
CB-82+CB-152	28.0	0	2.60	11.8	10.5	52.9
CB-118	13.0	0	0.800	5.70	13.0	32.5
CB-114	42.0	14.2	0.600	1.90	16.7	75.4

CB-146	13.0	0	1.80	14.3	0	29.1
CB-153	2.00	0	1.40	13.4	24.1	40.9
CB-105	2.00	0	1.10	79.5	17.0	99.6
CB-179	2.00	0	1.80	15.4	14.3	33.5
CB-138	103	18.1	41.0	49.4	28.1	240
CB-158	2.00	7.70	2.00	35.2	6.7	53.6
CB-126	20.2	19.3	1.40	145	17.5	203
CB-166	13.0	12.4	2.70	27.7	7.10	62.9
CB-187	2.00	0	5.00	4.30	7.10	18.4
CB-183	2.00	0	5.00	3.90	9.00	19.9
CB-128	4.00	0	1.20	7.70	7.00	19.9
CB-177	7.00	0	12.3	35.3	5.70	60.3
CB-156	0	0	0	0	0	0
CB-180	2.00	0	11.5	19.3	8.00	40.8
CB-169	30.0	8.70	17.1	13.5	21.4	90.7
CB-170	2.00	0	3.60	12.2	9.90	27.7
CB-189	2.00	0	21.3	60.7	13.6	97.6
ΣPCBs	3373	2157	931	4195	1462	12118

Table S-3 Concentrations of PBDEs (pg/g d.w.) in Mariana Trench sediments across all depths.

PBDEs	C1-I-M-S012B02	C1-I-M-S078B10	TS03-S106GT02 (S)	TS03-S106GT02 (D)	TS03-S090LANDER11
BDE-28			1.80		
BDE-49			7.50	32.7	7.70
BDE-47			7.50	63.3	12.6
BDE-66			9.80	47.3	
BDE-100			37.9	19.8	
BDE-99			11.4	40.0	
BDE-153			26.6	166	24.1
BDE-196	84.8		77.0		
BDE-208	36.9		65.0		280
BDE-207	118				266
∑PBDEs	240		245	369	591
∑$_8$PBDEs	0	0	85.2	289	36.7

Note: PBDEs highlighted in grey indicate the most commonly occurring eight congeners in the environment. While five (BDE-28, 47, 99, 100, and 153) were detected, BDE-54, 183, 209 were not detected. ∑$_8$PBDEs indicates the summation of the PCBs highlighted in blue.

Table S-4 ICES Convention inventory of 7 PCBs.

PCBs (pg/g)	C1-I-M-S015B02	C1-I-M-S078B10	TS03-S106GT02 (S)	TS03-S106GT02 (D)	TS03-S090LANDER11
PCB028	9.0000	21.2000	6.1000	93.8000	20.9000
PCB052	0.00	1036.9000	154.7000	479.8000	358.2000
PCB101	0.000	52.9000	7.6000	29.6000	74.2000
PCB118	13.000	0.00	0.8000	5.7000	13.0000
PCB138	103.000	18.1000	41.0000	49.4000	28.1000
PCB153	2.000	0.00	1.4000	13.4000	24.1000
PCB180	2.000	0.00	11.5000	19.3000	8.0000
Σ_7PCBs	129.0000	1129.1000	223.1000	691.0000	526.5000
Mean Σ_7PCBs	18.4286	161.3000	31.8714	98.7143	75.2143

Table S-5 TEF values of dl-like PCBs.

PCBs (pg/g)	TEF
PCB110+PCB077	0.0001
PCB087+PCB081	0.0003
PCB126	0.1
PCB169	0.03
PCB105	0.00003
PCB114	0.00003
PCB118	0.00003
PCB123	0.00003
PCB156	0.00003
PCB157	0.00003
PCB166	0.00003
PCB189	0.00003

Table S-6 Toxic Equivalence (TEQ) of Mariana Trench sediment samples.

Samples	TEQ (∑PCBi*TEF)
C1-I-M-S015B02	2.92666
C1-I-M-S078B10	2.202598
TS03-S106GT02 (S)	0.654365
TS03-S106GT02 (D)	14.874955
TS03-S090LANDER11	2.400852

Table S-7 Comparison of PCB concentrations of Mariana sediments with other marine sediments.

Location	Mean ΣPCBs (pg/g d.w.)	Ocean Depth (m)	Number of congeners	References
Bering Sea	71	<500	46	Hong *et al*., 2012
Gulf of Alaska	2,000	249	36	Iwata *et al*., 1994
Gulf of Lion, Mediterranean	900	1,000		Burns and Villeneuve, 1987
Southern Cretan Margin	270	2,560	33	Mandalakis *et al*., 2014
Adriatic Sea	5,200	40-60	28	Combi *et al*., 2016
Bering Strait	214	<500	46	Ma *et al*., 2015
Chukchi Sea	185	<500	46	Ma *et al*., 2015
Iceland Station, Arctic Ocean	99	<500	46	Ma *et al*., 2015
Mariana Trench	3373	6,980	36	This study
Mariana Trench	2157	8638	36	This study
Mariana Trench	931	9373 (S)	36	This study
Mariana Trench	4,195	9373 (D)	36	This study
Mariana Trench	1,462	10,908	36	This study

Table S-8 Relative percentage concentration of clays (illite, nontronite, clinochlore, and gismondine) and other minerals detected from three Mariana Trench sediment samples, by XRD analysis.

Samples	Quartz	Albite	Illite	Nontronite	Clinochlore	Gismondine	Halite
TS03-S090LANDER11	7.0 %	17.3 %	5.4 %	32.4 %	13.3 %	17.9 %	6.8 %
TS03-S106GT02 (S)	4.9 %	14.9 %	6.3 %	41.5 %	17.5 %	14.9 %	
TS03-S106GT02 (D)	6.3 %	37.3 %	11.6 %	23.3 %	16.9 %		4.7 %

Supplementary Figures

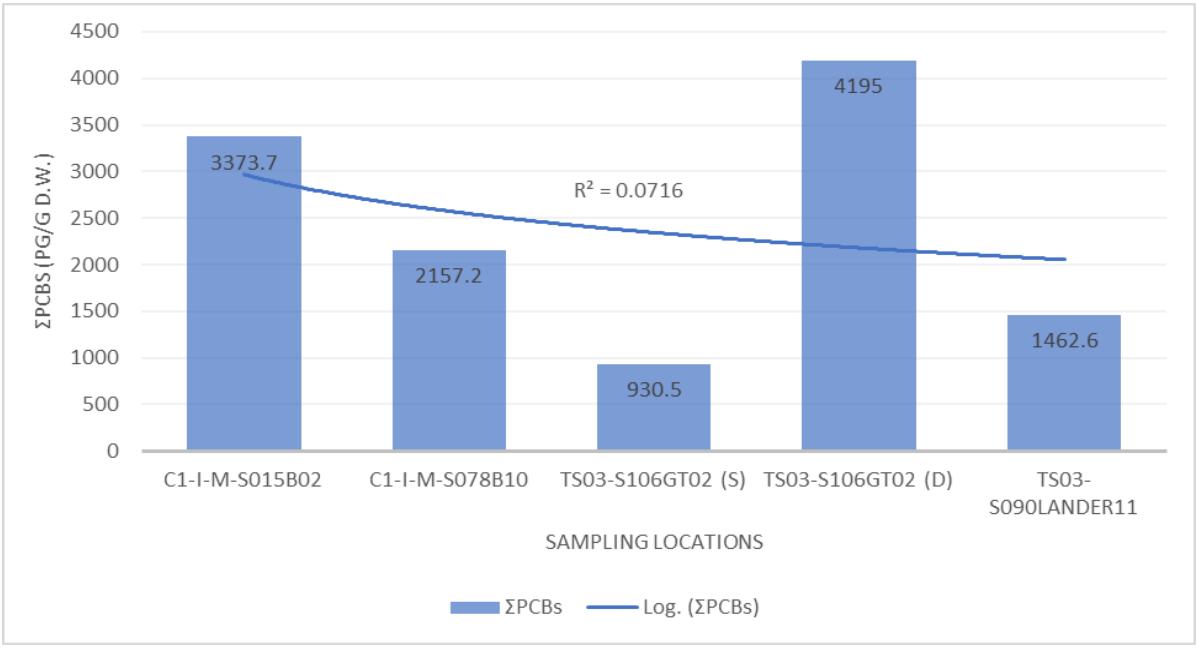

Figure S-1 ΣPCBs along with logarithmic trendline for the five sediment samples across different water depths in Mariana Trench.

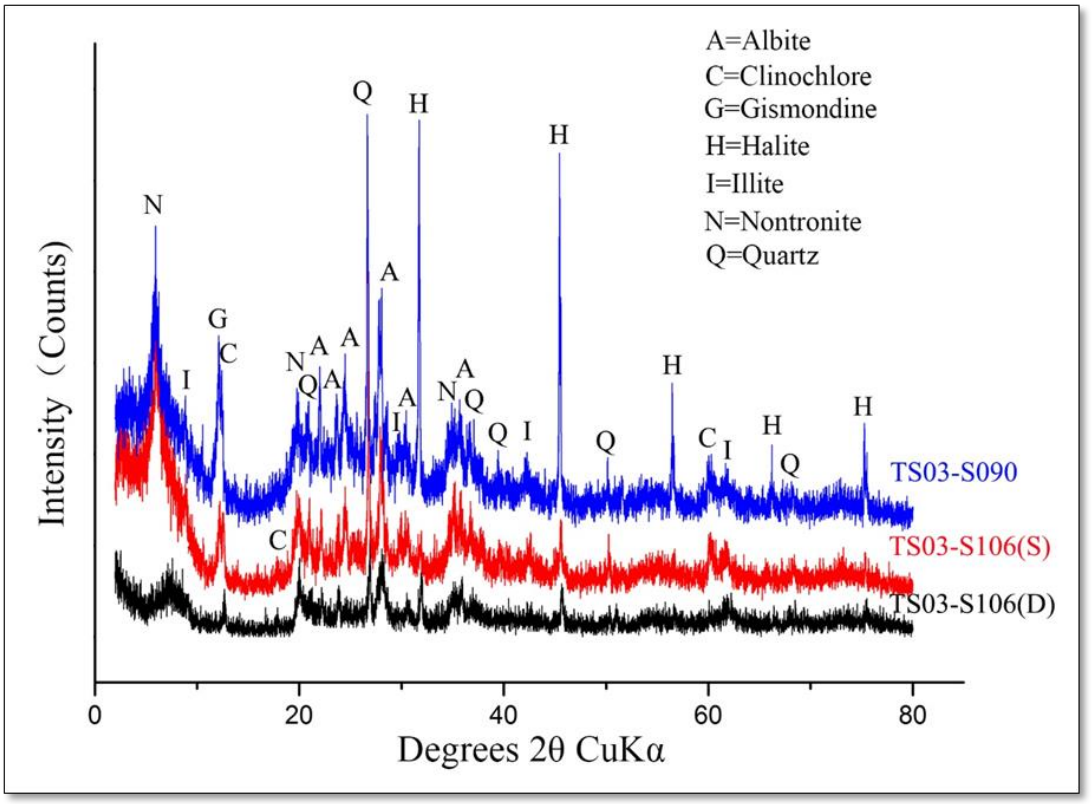

Figure S-2 XRD analysis of Mariana Trench sediments samples TS03-S090, TS03-S106(S), and TS03-S106(D). Clinochlore, nontronite, illite, and gismondine are clay minerals, while albite, and quartz represent non-clay minerals. Halite originates from sea-water, and is not a component from the Trench sediments.

Supplementary Information References

Burns, K.A., Villeneuve, J.P. (1987) Chlorinated hydrocarbons in the open Mediterranean ecosystem and implications for mass balance calculations. *Marine Chemistry* 20, 337-359.

Combi, T., Miserocchi, S., Langone, L., Guerra, R. (2016) Polychlorinated biphenyls (PCBs) in sediments from the western Adriatic Sea: Sources, historical trends and inventories. *Science of the Total Environment* 562, 580-587.

Gong, X., Qi, S., Wang, Y., Julia, E.B., Lv, C. (2007) Historical contamination and sources of organochlorine pesticides in sediment cores from Quanzhou Bay, Southeast China. *Marine Pollution Bulletin* 54, 1434-1440.

Hong, Q.Q., Wang, Y., Luo, X.J., Chen, S.J., Chen, J.G., Cai, M.H., Cai, M.G., Mai, B.X. (2012), Occurrence of polychlorinated biphenyls (PCBs) together with sediment properties in the surface sediments of the Bering Sea, Chukchi Sea and Canada Basin. *Chemosphere* 88, 1340-1345.

Iwata, H., Tanabe, S., Aramoto, M., Sakai, N., Tatsukawa, R. (1994) Persistent organochlorine residues in sediments from the Chukchi Sea, Bering Sea and Gulf of Alaska. *Marine Pollution Bulletin* 28, 746-753.

Ma, Y., Halsall, J., Crosse, J.D., Graf, C., Cai, M., He, J., Gao, G., Jones, K. (2015) Persistent organic pollutants in oceanic sediments from the North Pacific to the Arctic Ocean. *Journal of Geophysical Research: Oceans* 120, 2735-2743.

Mandalakis, M., Polymenakou, P.N., Tselepides, A., Lampadariou, N. (2014) Distribution of aliphatic hydrocarbons, polycyclic aromatic hydrocarbons and organochlorinated pollutants in deep-sea sediments of the southern Cretan margin, Eastern Mediterranean Sea: a baseline assessment. *Chemosphere* 106, 28-35.