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Contamination Levels and Human Dietary Exposure of Polychlorinated Biphenyls (PCBs) in Seafood from Korea

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Polychlorinated biphenyls (PCBs) residues were measured in 70 marine organisms (42 species) from Korean coastal waters. The PCBs residues in the organisms varied from 0.02 to 102 ng/g dry weight. The levels in various marine organisms from Korea were lower than those found in other countries. Comparing organism groups, the highest PCBs residues were recorded in crustaceans, followed by fish, bivalves, cephalopods, gastropods and seaweeds. PCB 153, 110 and 187 were the most frequently and abundantly found in the all organisms. The result indicates that these PCBs congeners can be used as indicator PCBs to monitor PCBs contamination in various marine organisms from Korean coastal waters. The dietary intake of PCBs through the seafood consumption was 0.81 ng/kg body weight/day. The relative contributions of each organism group to the total dietary intake were, in descending order, 50.3% for fish, crustaceans (39.0%), cephalopods (4.9%), seaweeds (1.1%) and gastropods (0.2%). Lifetime cancer risk and target hazard quotient (THQ) due to the seafood consumption in the Korean population were evaluated by the equation of the US EPA risk assessment. Results indicate that the dietary intake of PCBs through the consumption of seafood from Korea seems to be safe for human health with negligible cancer and non-cancer risk.

Key words: PCBs, seafood, dietary intake, lifetime cancer risk, target hazardous quotient (THQ), human health

1. Introduction

A large number of studies have demonstrated that many toxic organic contaminants are present in the marine environment, the highest concentrations being often detected in estuaries and coastal zones.[1] Among a large number of anthropogenic chemicals, polychlorinated biphenyls (PCBs) are widespread contaminants in aquatic environments that can accumulate in aquatic organisms through the food chain[2-3]. The United Nations Environment Programme (UNEP) have designated the PCBs as persistent organic pollutants (POPs), because of their characters such as highly toxicity, bio-accumulation in living organisms, and long-range transport[4].

Since PCBs were first manufactured commercially in 1929, they have been used in many countries as dielectrical fluids, non-flammable plasticizers, because of their high chemical stability and low flammability. The attractive properties of PCBs to industry have also resulted in their environmental persistence and toxicological problems for marine organisms and humans[5-6].

An extensive survey has been launched in the National Fisheries Research and Development Institute (NFRDI), to determine the presence of POPs in the marine environments of Korea. In particular, the residues of PCDDs/DFs, organochlorine pollutants (OCPs), polycyclic aromatic hydrocarbons (PAHs), hexachlorobenzene (HCB) and tributyltin (TBT) in various marine organisms have been determined. The results on dietary intakes and human health risks of PCDDs/DFs[7], PAHs[8], HCB[9] and TBT[10] have been reported.

Seafood is one of major sources of protein for people in the world and is an important part of the Korean diet. Many studies that have estimated human exposure to toxic chemicals have concluded that over 90% of the

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intake is from contaminated food[11-14]. The other exposure pathways only contribute negligible levels[15-16]. Limited data are available concerning the levels of PCBs in seafood from Korea, in spite of the great public concern about the presence of these chemicals in food. The objectives of this study were to estimate dietary intake of PCBs due to the seafood consumption from Korea, and to assess human health risks by their consumption.

2. Materials and Methods

2.1. Sample collection

Seventy marine organisms (42 species) were sampled from local fish markets in several Korean coastal regions from January 2001 to October 2003. These organisms are commonly consumed species that are commercially important in Korea. The selected marine species were classified into six groups: fish (31 samples, 24 species), crustaceans (7 samples, 5 species), bivalves (15 samples, 6 species), gastropods (2 samples, 2 species), cephalopods (4 samples, 2 species), and seaweed (11 samples, 3 species). Biological information on the marine organisms analysed is summarized in Table 1.

2.2. Sample preparation and analysis

Marine organism samples were stored in a cooler box with ice or dry ice and immediately transported to the laboratory. The muscles of the fish and cephalopods were homogenized with an ultra-disperser. The shells of the crustaceans, bivalves and gastropods were removed and the whole soft tissues were pooled and homogenized. The whole body of seaweed samples were homogenized. All organism samples were stored at -20°C and then freeze-dried.

Freeze-dried organism samples were extracted (approximately 10 g) using accelerated solvent

Table 1. Biological information on various marine organisms collected from Korean coastal waters

Common name (species)	Moisture (mean, %)	Lipid[a] (mean, %)
Fish (n=31) Korean flounder (*Glyptocephalus stelleri*), Roundnose flounder (*Eopsetta grigorjewi*), Olive flounder (*Paralichthys olivaceus*), Alaska pollock (*Theragra chalcogramma*), Sailfin sandfish (*Arctoscopus japonicus*), Pacific saury (*Cololabis saira*), Herring (*Clupea pallasii*), Hairtail (*Trichiurus lepturus*), Mackerel (*Scomber japonicus*), Anchovy (*Engraulis japonica*), Eelgrass (*Conger myriaster*), Sea bass (*Lateolabrax japonicus*), Red tongue sole (*Cynoglossus joyneri*), Sharp toothed eel (*Muraenesox cinereus*), Butterfish (*Pampus argenteus*), Brown croaker (*Miichthys miiuy*), Spanish mackerel (*Scomberomorus niphonius*), Rockfish (*Sebastes schlegeli*), Marbled rockfish (*Sebastiscus marmoratus*), Whitesaddled reef fish (*Chromis notata*), Korean pomfret (*Pampus echinogaster*), Armorclad rockfish (*Sebastes hubbsi*), Greenling (*Hexagrammos otakii*), Grey mullet (*Mugil cephalus*)	62-84 (76)	1.1-48 (14)
Crustaceans (n=7) Hair crab (*Erimacrus isenbecki*), Snow crab (*Chionoecestes opilio*), Red snow crab (*Chionoecestes japonicus*), Fleshy prawn (*Penaeus chinensis*), Blue crab (*Portunus trituberculatus*)	58-79 (70)	2.4-17 (10)
Bivalves (n=15) Mussel (*Mytilus coruscus*), Pacific oyster (*Crassostrea gigas*), Korean scallop (*Chlamys farreri*), Japanese cockle (*Fulvia mutica*), Razor clam (*Solen strictus*), Manila clam (*Ruditapes philippinarum*)	46-84 (68)	2.0-10 (5.1)
Gastropods (n=2) Abalone (*Haliotis discus hannai*), Top shell (*Batillus cornutus*)	65-78 (71)	2.1-4.0 (3.1)
Cephalopods (n=4) Cuttlefish (*Todarodes pacificus*), Whiparm octopus (*Octopus minor*)	63-79 (73)	3.1-4.7 (4.1)
Seaweeds (n=11) Seersucker (*Costaria costata*), Sea mustard (*Undaria pinnatifida*), Sea-lattuce (*Lva latuca*)	73-87 (79)	0.29-0.86 (0.5)

[a]Lipid in the dry weight.

Table 2. Limits of detection (LOD, ng/g dry weight) of each PCB congener in marine organisms in this study

PCB congeners	IUPAC numbers	Limit of detection
2,4'-Dichlorobiphenyl	PCB 8	0.016
2,2',5-Trichlorobiphenyl	PCB 18	0.021
2,4,4'-Trichlorobiphenyl	PCB 28	0.029
2,4,5-Trichlorobiphenyl	PCB 29	0.028
2,2',3,5'-Tetrachlorophenyl	PCB 44	0.025
2,2',5,5'-Tetrachlorobiphenyl	PCB 52	0.032
2,2',3,4,5'-Pentachlorobiphenyl	PCB 87	0.026
2,2',4,5,5'-Pentachlorobiphenyl	PCB 101	0.033
2,3,3',4',6-Pentachlorobiphenyl	PCB 110	0.030
2,3',4,4',5-Pentachlorobiphenyl	PCB 118	0.053
2,2',3,3',4,4'-Hexachlorobiphenyl	PCB 128	0.033
2,2',3,4,4',5'-Hexachlorobiphenyl	PCB 138	0.030
2,2',4,4',5,5'-Hexachlorobiphenyl	PCB 153	0.035
2,2',3,3',4,4',5-Heptachlorobiphenyl	PCB 170	0.058
2,2',3,4,4',5,5'-Heptachlorobiphenyl	PCB 180	0.033
2,2,3,4',5,5',6-Heptachlorobiphenyl	PCB 187	0.034
2,2',3,3',4,4',5,5'-Octachlorobiphenyl	PCB 194	0.030
2,2',3,3',4,4',5,6-Octachlorobiphenyl	PCB 195	0.025
2,3,3',4,4',5,5',6-Octachlorobiphenyl	PCB 205	0.036
2,2',3,3',4,4',5,5',6-Nonachlorobiphenyl	PCB 206	0.036
Decachlorophenyl	PCB 209	0.036

extraction (ASE) system under the conditions of the extraction of 10 minutes at 30 W. The extraction solvent was *n*-hexane (Suprapur, Merck, Germany) and acetone (Suprapur, Merck, Germany) (1:1). The extracts were then evaporated to a small volume using a rotary evaporator. Extracted samples were cleaned on a multi-layer silica gel (100-200 mesh, Sigma-Aldrich, USA) column containing NaOH-silica gel and H_2SO_4-silica gel with 150 mL of *n*-hexane. The eluted fractions were concentrated to less than 1 mL and analysed for PCBs.

PCBs quantifications were performed using Hewlett Packard 6890 gas chromatography (GC) equipped with electron capture detector (ECD). A capillary column DB-XLB (60 m length, 0.32 mm inner diameter, 0.25 μm film thickness) was used for separation of PCBs. The oven temperature was programmed from 100°C (1 min) to 265°C at 1.2 °C/min. The injection and detector temperatures were maintained at 250°C and 300°C, respectively. Nitrogen was used as carrier gas and make-up gas.

In order to assess the quality of determinations using the routine experimental procedure and instrument conditions, the certified mussels (*Mytilus edulis*) homogenate (2974, NIST, USA) as a Standard Reference Material (SRM) were analyzed. The recovery results of PCBs ranged from 70-80%. Procedural blanks were processed in the same manner as the real samples, and the response levels were below 10% for each analyte. In this study, the 21 PCB congeners (IUPAC numbers 8, 18, 28, 29, 44, 52, 87, 101, 110, 118, 128, 138, 153, 170, 180, 187, 194, 195, 205, 206, 209) were analyzed and the limits of detection (LOD) for individual PCB congeners are presented in Table 2. In this study, the concentrations of non-detected congeners of the chemicals are expressed as zero.

2.3. Calculation of dietary intake

The average body weight of a Korean adult was set as 60 kg. The data on average daily dietary intake were obtained from national food survey results by the Ministry of Health and Welfare (MOHW).[17] Total average daily ingestion of food in Korea was 1,315 g. Seafood, including both natural and processed seafood, comprised 73.1 g or 5.6% of the total food ingested. The ingestion values were calculated as the sum of all the individual natural species sampled. Daily dietary intake of PCBs by the consumption of seafood was calculated by multiplying the concentrations of chemicals in the organisms (ng/g wet weight) by the available ingestion of individual marine organisms (average daily intake in g/day).

3. Results and Discussion

3.1. PCBs residues in marine organisms

PCBs were detected in all of the marine organisms analysed in this study, ranging from 0.02 to 102 ng/g dry weight. The moisture and lipid contents (dry weight basis) in the organisms were in the ranges of 46-87% and 0.3-48%, respectively (Table 1). Fish in particular showed the widest range of lipid content.

In decreasing order, the concentrations of PCBs in the six organism groups were crustaceans, fish,

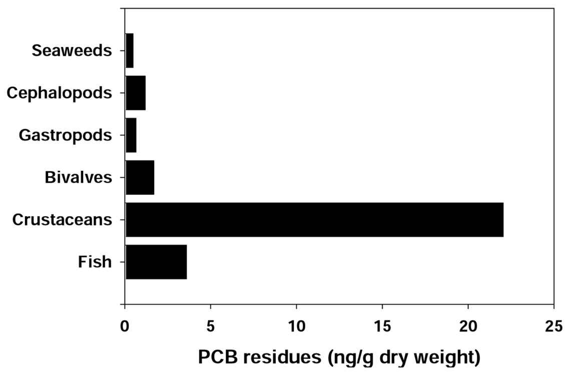

Fig. 1. Comparison of average concentrations of total PCBs in various marine organisms from Korean coastal waters.

bivalves, cephalopods, gastropods and seaweeds (Fig. 1). The PCBs concentrations in fish varied from 0.02 to 12.4 ng/g dry weight with a mean 3.6 ng/g dry weight. The highest PCBs residue in fish was found in grey mullet (*Mugil cephalus*) from the Incheon coast. Eelgrass (*Conger myriaster*) from the Yeosu coast and Pacific saury (*Cololabis saira*) from the Busan coast had relatively high levels of PCBs . The residues of PCBs in crustaceans ranged from 1.42 to 102 ng/g dry weight with a mean 22.0 ng/g dry weight. Red snow crabs (*Chionoecestes japonicus*) from the Pohang coast had the highest PCBs levels (102 ng/g dry weight) of all the marine organism surveyed in this study. Snow crabs (*Chionoecestes opilio*) from the Pohang coast also had higher levels of these contaminants than other crustacean species. The levels of PCBs in bivalves ranged from 0.15 to 4.1 ng/g dry weight (mean value 1.7 ng/g dry weight). Korean scallops (*Chlamys farreri*) from the Sokcho coast and Japanese cockle (*Fulvia mutica*) from the Yeosu coast contained relatively high levels. In gastropods, the levels of PCBs ranged from 0.71 to 2.13 ng/g dry weight (mean value 1.2 ng/g dry weight). PCBs concentrations in cephalopods were 0.71-2.13 ng/g dry weight. Seaweed residues of PCBs showed the lowest values among six organism groups surveyed. The contamination tendency of PCBs in various marine organisms from Korea is similar to that of PCDDs/DFs in the same organism samples.[7] This means that the main source and/or bioaccumulation character of PCBs in marine organisms is similar to those of PCDDs/DFs.

PCBs residues in various marine organisms from Korean coastal waters were compared with those in various marine organisms from other countries (Table 3). Although the number of PCBs and organism species analysed in other studies may differ, the 21 congeners of PCBs have found in most samples regardless of organism species. The PCB concentrations in crustaceans in this study ranged from 0.41-22.4 ng/g wet weight. These values were comparable to PCBs levels detected in Sri Lanka[18)] and White Sea[21)]. The PCBs levels (0.01-1.43 ng/g wet weight) in bivalves from Korean coastal waters were lower than those from Italy[22)], Biscay Bay in Spain[19)] and Adriatic Sea[20)]. The PCB residues in fish in this study were comparable to those from Hong Kong[23)]. However, PCB residues from other reports were greater than those in this study. In particular, PCBs concentrations in fish from Sri Lanka[18)], Douro estuary in Portugal[24)], Po delta in Italy[24)] and San Francisco Bay in USA[27)] were about 10-150 times greater than those reported in this study. Consequently, PCBs residues in marine organisms from Korean coastal waters were low level with respect to those from other countries.

3.2. Occurrence of PCB congeners

All of the marine organism samples contained PCB congeners at different occurrence. In this study, the organism group with high occurrence for PCB congeners analysed was crustaceans, in particular snow crabs (*C. opilio*), red snow crabs (*C. japonicus*), and hair crabs (*Erimacrus isenbecki*). In fish, benthic organism such as oliver flounder (*Paralichthys olivaceus*) roundnose flounder (*Eopsetta grigorjewi*) showed a relatively high occurrence of PCB congeners compared with other species. Seaweed group was characterized by the lowest occurrence of these congeners with the lowest residue of the contaminants.

Among PCBs congeners, the penta- and hexa-CBs such as PCB 29, 87, 110, 118, 153, 138 and 187 were the most frequently and abundantly found in all of the marine organisms examined (Fig. 2). In particular, PCB

Table 3. Comparison of PCBs concentrations (ng/g wet weight) in marine organisms measured in this study with those from different locations and/or other countries

Locations	Species	Concentrations
Korean coasts (this study)	Fish (Grey mullet, Mackerel, Pacific saury, Herring, Alaska pollack)	0.004-1.99
Korean coasts (this study)	Crustaceans (Crab, Shrimp)	0.41-22.4
Korean coasts (this study)	Bivalves (Mussel, Pacific oyster, Clam)	0.04-1.43
Sri Lanka[18)]	Crab (Blue crab, Mud crab)	0.90-10.0
Biscay Bay, Spain[19)]	Crab	58.4-137
Adriatic Sea[20)]	Crustacean (Red shrimp, Norway lobster, Pink shrimp, Shrimp)	0.35-1.70
White Sea[21)]	Crab	8.5-19.2
Italy[22)]	Clam	1.6-15.4
Biscay Bay, Spain[19)]	Bivalve (Mussel, Oyster)	4.0-77.9
Adriatic Sea[20)]	Bivalve (Mussel, Clam)	1.24-18.5
Sri Lanka[18)]	Fish (Mullet, Cat fish, Green cromide, Juvenile, Silver batfish, Flounder)	8.0-160
Hong Kong[23)]	Fish (Grouper, Gold line sea bream, Macau sole)	0.01-0.58
Douro estuary, Portugal[24)]	Fish (Flounder, Mullet)	23.3-344
Finland[25)]	Fish (Baltic herring, Sprat)	31-77
Biscay Bay, Spain[19)]	Fish (Mullet)	18.5-84.5
Po delta, Italy[26)]	Fish (Eel)	211-265
San Francisco Bay, USA[27)]	Fish (California halibut, Jacksmelt, Striped bass, White croaker, White sturgeon)	22-191
Salton Sea, USA[28)]	Fish (Corvina, Tilapia)	10.9-85.2
White Sea[21)]	Fish (White sea cod)	6.80-25.7
Adriatic Sea[20)]	Fish (Anchovy, Mackerel, Red mullet)	16.4-80.6
Adriatic Sea[29)]	Fish (Mackerel, Anchovy, Red mullet, Cod, Sole, Frog-fish)	1.21-88.2

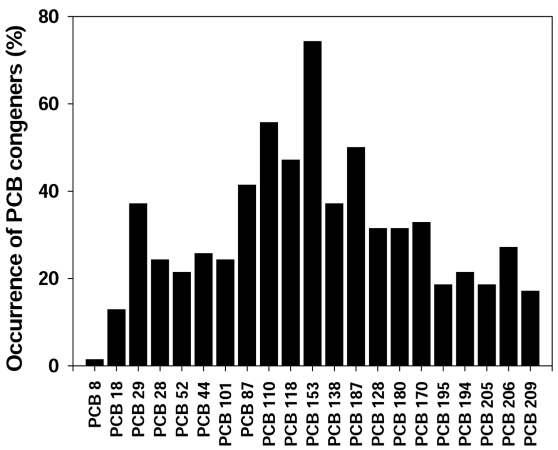

Fig. 2. Occurrence of individual PCB congener in various organisms from Korean coastal waters.

153, 110 and 187 were the dominant congeners in this study, showing over 50% of total occurrences. This finding could be interpreted by two reasons. The first reason is that a large amount of these PCB congeners have been widely used and introduced into the marine environment through various routes. The other reason is that the penta- and hexa-CBs is more bioaccumulative congeners to fatty tissue in the living organisms than other PCB congeners. Actually, these PCB congeners were shown to be the dominant congeners in various marine environmental matrices by previous papers[21,30-31)]. The result suggests that PCB 153, 110 and 187 can be used as the indicator PCBs in various marine organisms in monitoring contamination levels and bioaccumulation extent of PCBs in the marine environment of Korea.

3.3. Dietary intake of PCBs

The daily dietary intakes of PCBs through the seafood consumption by the Korean population were presented in Table 4. The dietary intake of PCBs by the seafood consumption in Korea was estimated to be 0.81 ng/kg body weight/day. This value is lower than the dietary intakes through the seafood consumption of Italy (3.69 ng/kg body weight/day)[32)] and the Netherlands (5.6 ng/kg body weight/day)[33)]. In this study, the daily dietary intake of PCBs by the fish

Table 4. Estimated dietary intake (ng/kg body weight/day) of PCBs through the seafood consumption by the Korean population

	Intake amount (g/day)	Dietary intake (ng/kg body weight/day)	Contribution (%)
Fish	30.5	0.41	50.4
Crustaceans	3.3	0.32	39.0
Bivalves	4.4	0.037	4.5
Gastropods	0.5	0.001	0.13
Cephalopods	7.8	0.04	4.9
Seaweeds	6.5	0.009	1.1
Sum	53.0	0.81	100

consumption was 0.41 ng/kg body weight/day. The dietary intake of PCBs by the consumption of crustaceans was 0.32 ng/kg body weight/day. Bivalve's daily intake of PCBs was 0.037 ng/kg body weight/day. The dietary intake of PCBs by cephalopods consumption was 0.04 ng/kg body weight/day. Gastropod and seaweed groups showed relatively low contribution to the intakes of PCBs.

The relative contribution of individual organism group to the estimated dietary intake of PCBs was also given in Table 4. The fish group, comprising 50.3% of the total dietary intake, had the highest contribution, followed by crustaceans (39.0%), cephalopods (4.9%), seaweeds (1.1%) and gastropods (0.2%). Although the residues of PCBs in fish was low, fish were the highest contributor to the dietary intake. This is due to the higher consumption of this group compared with the other organism groups.

3.3. Human health risk assessment

In general, there are two methods of human health risk assessment from ingesting toxic organic contaminants. One is carcinogenic risk and the other is non-carcinogenic risk. PCBs can be considered as a contaminants with cancer and non-cancer effects. The potential health risks of the consumption of seafood contaminated with PCBs were evaluated for the Korean adult population using the risk assessment method of the United States Environmental Protection Agency (US EPA)[34)]. For carcinogenic effects, the risk is expressed as a cancer risk. The cancer risk below 1×10^{-6} means the adverse effects are negligible. The equation for estimating exposure of carcinogenic pollutants by marine organisms consumption is as follows:

$$\text{Cancer Risk} = \frac{IFR \times C \times ED \times EF \times CSFo}{BW \times AT}$$

where IFR is the food ingestion rate (g/day); C is the concentration of chemicals in food (mg/g wet weight); ED is the exposure duration (adults=30 years); EF is the exposure frequency (350 days/year); CSFo is the oral cancer slope factor (2.0 (mg/kg/day)-1)[35)], the data for individual chemicals were obtained from integrated risk information system (IRIS) reported by US EPA (2004))[35)]; BW is the human body weight (adults=60 kg); AT is the average time for carcinogens (365 day/year×number of exposure years, assuming 70 years). From this equation, a cancer risk by the seafood consumption was 6.70×10^{-7} and the value is below 1×10^{-6} of cancer risk guideline. The result indicates that the level of exposure is not likely to cause any adverse effect in the Korean population during their lifetime.

For non-carcinogenic effects, the risk is expressed as a target hazard quotient (THQ). If the THQ exceeds unity (1), an adverse effect might occur for a lifetime in human population. The equation for estimating exposure of non-carcinogenic pollutants by marine organisms consumption is as follows:

$$THQ = \frac{EF \times ED \times IFR \times C}{RfDo \times BW \times AT}$$

where RfDo is the oral reference dose (2×10^{-5} mg/kg/day)[35)]. From this equation, a cancer risk was 1.67×10^{-2} and the value is below 1. The result indicates that the level of exposure is not likely to cause any adverse effect in the Korean population during their lifetime.

Judging from this investigation, the PCBs levels in various seafoods observed in the present study would not seem to cause cancer or reproductive disorders. However, PCBs has been pointed out to act as a dioxin-like compound and the effects of low-dose exposure remain clear. Therefore, more intensive studies on PCBs intakes through the seafood consumption will be necessary, in particular, for heavy fish consumers and their infants and fetuses. In addition, guidelines for the dietary intake of seafood should be established to protect the health of the Korean population.

4. Conclusion

The PCBs residues in the organisms varied from 0.02 to 102 ng/g dry weight. The levels in various marine organisms from Korea were lower than those found in other countries. The dietary intake of PCBs through the seafood consumption was estimated to be 0.81 ng/kg body weight/day. The relative contribution of each organism group to the total dietary intake was, in descending order, fish (50.3%), crustaceans (39.0%), cephalopods (4.9%), seaweeds (1.1%) and gastropods (0.2%). Lifetime cancer risk and target hazard quotient (THQ) due to the seafood consumption in the Korean population were below risk guidelines. Results indicate that the dietary intake of PCBs through the consumption of seafood from Korea seems to be safe for human health with negligible cancer and non-cancer risk. The current residues of PCBs in various marine organisms are not potential threats to public health in Korea.

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