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Background levels and dietary intake of PCDD/Fs in domestic and imported meat in South Korea

MeeKyung Kim *, Sooyeon Kim, Seon Jong Yun, Dong-Gyu Kim, Gab-Soo Chung

National Veterinary Research and Quarantine Service, 480 Anyang 6-dong, Manangu, Anyang, Gyeonggido 430-824, South Korea

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Abstract

A survey was conducted in South Korea to determine residual levels and dietary intake of polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) from meat. Altogether 119 domestic and 164 imported samples of beef, pork, and chicken were examined. The mean levels of PCDD/Fs in upper bound were 0.21, 0.22, and 0.04 pg WHO-TEQ/g fat for beef, pork, and chicken, respectively. The low level of PCDD/Fs in chicken probably resulted from the low fat content in the samples used. The samples were separated into domestic and imported products in order to investigate the sources of contamination. PCDFs were the more dominant congeners in domestic beef and were similar to those found from emission of incineration. However, the congener profiles of domestic beef and incineration were difficult to compare because the environmental fate and animal metabolism were involved. The upper bound dietary intake of PCDD/Fs from beef, pork and chicken was calculated to be 0.04 pg/WHO-TEQ/kg bw/day. The combined consumption of beef, pork, and chicken was found to be 84.8 g per day for a person weighing 60 kg and represented 5.7% of their total daily food intake.

Keywords: PCDD/Fs; Dietary intake; Beef; Pork; Chicken

1. Introduction

Human exposure to environmental contaminants from food are various and complex. The dioxin crisis in Belgium in January 1999 affected Korea's pork importation and led to increased awareness of chemical residues in foods of animal origin. One of the major sources of dioxins (PCDD/Fs) in the environment is from combustion. Incineration of waste, including municipal solid waste, hospital waste and industrial hazardous waste, is probably the most significant source of PCDD/Fs (Goldfarb, 1989; Wagrowski and Hites, 1998). The potential precursors of dioxins in combustion processes are PCBs, PCP and chlorinated benzenes (Buser and Bosshardt, 1978; Fries et al., 2002). However, in most cases of dioxin contamination incidents in foods of animal origin, animal feeds have been identified as the source of the contamination (Covaci et al., 2002; Hoogenboom et al., 2004). Dioxins contained in the emissions from incineration plants have contaminated both crops and soil thus giving rise to a background level of dioxin in animal feeds (Malisch, 2000). Contaminated feed additives also increase the levels of PCDD/Fs in food of animal origin (Eljarrat et al., 2002; Esteban et al., 2002; Llerena et al., 2003). The hydrophobic properties of polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) result in their accumulation in the fatty content of food products (Liem, 1999; Ferrario and Byrne, 2000). Ingestion of PCDD/Fs by humans occurs mainly through the food chain and especially through foods of fish and animal origin (Schecter et al., 1994; Baars et al., 2004). The fate of PCDD/Fs in the environment and in the food chain affects the congener patterns of residual PCDD/Fs in food of animal origin (Schuler et al., 1997; Alcock et al., 2002). Although the congener patterns differ in various foods

* Corresponding author. Tel.: +82 31 467 1982; fax: +82 31 467 1872.
E-mail address: kimmk@nvrqs.go.kr (M. Kim).

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exposure levels to background contamination should be taken into account. Korea imports large amounts of food of animal origin from several different countries. Human exposure levels are periodically monitored in the interests of public health. The background level of PCDD/Fs and their congener profiles were investigated in domestic and imported beef, pork, and chicken which represented the majority of meats consumed in Korea from January 2001 to December 2002. Considering the global contamination of PCDD/Fs, the results from domestic and imported meat in this study indicate the general levels of exposure in Korea.

2. Materials and methods

2.1. Materials

PCDD/Fs and ^{13}C-labeled standards were obtained from Cambridge Isotope Laboratories (CIL, Andover, MA, USA). HPLC-grade hexane, nonane, methylene chloride, ethyl acetate, benzene, and toluene were obtained from J.T. Baker (Phillipsburg, NJ, USA). The domestic samples, which included 38 beef, 48 pork, and 33 chickens, were collected nationwide in South Korea. The imported samples comprised 98 beef products from Australia, Canada, New Zealand, and USA, 58 pork products from Austria, Australia, Belgium, Canada, Denmark, France, Hungary, Mexico, Poland, Sweden, and USA, and eight chicken products from Canada, China, Thailand, and USA, which were randomly collected through the national quarantine service program for imported meat.

2.2. Sample preparation

2.2.1. High fat sample analysis

The appropriate amount of fat was extracted from homogenized sample in an oven at 80 °C for beef and pork and at 50 °C for chicken before analysis (Covaci et al., 2002; Kim et al., 2004). The 5 g fat samples were solubilized in 200 ml of hexane and were spiked with a known amount (typically 100 µl of 4 ng/ml) of 15 ^{13}C-labeled standards using the isotope dilution method based on the US EPA 1613B protocol. Acidic silica gel (deactivated 30%) was added to the mixture and shaken for 2 h followed by elution through an anhydrous sodium sulfate column. The sample was concentrated to 1–2 ml and a small amount of hexane was added. A clean-up standard, $^{37}Cl_4$-labeled 2,3,7,8-TCDD (typically 100 µl of 0.8 ng/ml), was added to the sample and clean-up was performed using the Power-Prep™ (Fluid Management Systems, Waltham, MA, USA) system with silica, alumina, and activated carbon columns. The sample was loaded onto the column after it had been conditioned with hexane in the case of the silica column and toluene, ethyl acetate/benzene (1:1, v/v), 50% methylene chloride, and hexane for the carbon column. The sample was purified by elution from the silica and alumina columns with 90 ml of hexane and 60 ml of 2% methylene chloride/hexane followed by elution from the silica, alumina and carbon columns with 120 ml of 50% methylene chloride/hexane. Ethyl acetate/benzene (4 ml, 1:1, v/v) was then used to elute the sample from the carbon column followed by 10 ml of hexane. The final fraction containing PCDD/Fs was collected by reverse elution of the carbon column with 80 ml of toluene. The eluate was concentrated in a rotary evaporator and the concentrate was treated with nitrogen until almost dry. The extract was redissolved in 90 µl of nonane and 10 µl of ^{13}C-labeled 1,2,3,4-TCDD and ^{13}C-labeled 1,2,3,7,8,9-HxCDD were added as the internal standards. The final concentrated sample was analyzed by HR-GC/MS.

2.2.2. Low fat sample analysis

A 10 g homogenized sample mixed with 40 g anhydrous sodium sulfate was spiked with ^{13}C-labeled standards and allowed to stand for 12 h. The sample was extracted with methylene chloride/hexane (1:1, v/v) using a Soxhlet extractor for 18 h. The extract was then evaporated and redissolved in hexane. A clean-up standard was added and purification of sample was carried out as described above.

2.2.3. High resolution gas chromatography/mass spectrometry (HR-GC/MS) analysis

Seventeen PCDD/Fs were analyzed by GC (HP 6890, Hewlett–Packard, Palo Alto, CA, USA) using a DB5MS capillary column (60 m × 0.25 mm I.D., 0.25 µm film thickness, J&W Scientific, USA) and MS (Autospec Ultima, Micromass, Manchester, UK). Peak identifications were made by retention time and mass on two of the most abundant ions.

The results were reported as lower and upper bound TEQs. The lower and upper bound levels were calculated using non-detects are equal to zero and equal to congener's LOD, respectively. Recoveries of the internal standards were 71–129%. A set of experiments were carried out to control the quality of measurements. System blanks were performed with samples of ^{13}C-labeled standards spiked to corn oil.

3. Results and discussion

The biological metabolism and lifetime of an animal might affect the residual concentrations and pattern of congeners seen in meat tissues. Congener-specific PCDD/Fs concentrations by weight in beef, pork, and chicken are presented in Table 1. The mean concentrations represent the levels of PCDD/Fs in domestic and imported products. The total number of samples of each type of meat was as follows: 136 beef, 106 pork, and 41 chickens. The distribution patterns in beef and pork varied between the domestic and imported samples. The concentrations of PCDDs were higher than PCDFs except in domestic beef. The highest levels of PCDD/Fs concentrations by weight were found in domestic pork followed by imported

Table 1
Congener-specific concentrations of PCDD/Fs (pg/g fat) in major types of meat consumed in South Korea

Congener	Domestic			Imported		
	Beef ($n = 38$)	Pork ($n = 48$)	Chicken ($n = 33$)	Beef ($n = 98$)	Pork ($n = 58$)	Chicken ($n = 8$)
2,3,7,8-TCDD	nd	nd	nd	nd	nd	nd
1,2,3,7,8-PeCDD	nd	nd	nd	nd	nd	nd
1,2,3,4,7,8-HxCDD	0.02	0.05	nd	0.01	0.08	nd
1,2,3,6,7,8-HxCDD	nd	nd	nd	0.36	0.06	nd
1,2,3,7,8,9-HxCDD	0.05	nd	nd	nd	nd	nd
1,2,3,4,6,7,8-HpCDD	0.45	1.03	0.04	2.55	0.68	nd
OCDD	1.36	3.49	0.71	2.29	3.39	0.38
Sum of PCDDs	1.88	4.57	0.75	5.21	4.21	0.38
2,3,7,8-TCDF	nd	nd	nd	nd	nd	nd
1,2,3,7,8-PeCDF	nd	0.01	0.02	nd	nd	nd
2,3,4,7,8-PeCDF	0.55	0.12	nd	nd	0.03	nd
1,2,3,4,7,8-HxCDF	0.49	0.67	nd	0.03	0.11	nd
1,2,3,6,7,8-HxCDF	0.48	0.38	nd	0.01	0.19	nd
2,3,4,6,7,8-HxCDF	0.24	0.16	nd	nd	0.28	nd
1,2,3,7,8,9-HxCDF	nd	0.04	nd	nd	0.33	nd
1,2,3,4,6,7,8-HpCDF	0.51	2.73	0.03	0.63	0.45	0.18
1,2,3,4,7,8,9-HpCDF	nd	0.09	nd	0.12	0.45	nd
OCDF	0.09	0.24	nd	0.71	1.63	nd
Sum of PCDFs	2.36	4.44	0.05	1.50	3.47	0.18
Sum of PCDD/Fs	4.24	9.01	0.80	6.71	7.68	0.56

n = number of samples; nd = not detected.

pork. The levels of PCDD/Fs concentrations by weight were raised in domestic pork due to the presence of OCDD and 1,2,3,4,6,7,8-HpCDF and levels in imported pork were raised due to the presence of OCDD and OCDF. The concentration of 1,2,3,4,6,7,8-HpCDD was higher than that of OCDD only in the imported beef compared to all the other samples. The concentration of OCDF in imported beef and pork was higher than that in domestically produced beef and pork. The concentration of PCDFs in imported pork increased with the number of chlorines substituted. No clear conclusions can be drawn from these data as the imported pork originated from 11 different countries. The PCDD/Fs concentrations by weight was 9.01 pg/g fat in domestic pork and 7.68 pg/g fat in imported pork. The concentration of PCDFs decreased with the increase in the number of chlorine substitutions except for 1,2,3,4,6,7,8-HpCDF. Tetra-, penta-, and hexa-chlorinated PCDDs were found less frequently than the comparable homologs of PCDFs. Overall, the distribution pattern of PCDD/Fs in domestic samples was similar in beef and pork. The location in which the animals were raised had no major influence on the congener profiles of the meat samples. The results suggested that the sources of PCDD/Fs were very similar throughout Korea even though the feedstuffs were different. Congeners 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD which have the highest toxic equivalent factors (TEFs) were not found in domestic beef, pork, or chicken. Most chicken samples were free of PCDD/Fs compared to beef and pork. Chicken samples had been trimmed of fat which may be the reason for the low levels of PCDD/Fs found. The frequency of detection of OCDD was about 30% in 33 domestic chicken samples. OCDD was also the most concentrated congener in chickens. The frequency of detection for OCDD and 1,2,3,4,6,7,8-HpCDF was about 13%, respectively, in eight imported chicken samples. In the case of imported beef, pork, and chickens background information regarding the source and possible causes for the congener profiles found was not available.

Table 2 shows the TEQ concentrations of PCDDs or PCDFs in domestic and imported beef, pork, and chicken. The TEFs recommended by the World Health Organization (WHO) in 1998 were used to determine the toxicological concentration as TEQ. The lower levels of PCDD/Fs were 0.42, 0.23, and 0.003 pg TEQ/g fat in domestic beef, pork and chicken, respectively and 0.08, 0.14 and 0.002 pg TEQ/g fat in imported beef, pork and chicken, respectively. The upper levels of PCDD/Fs were 0.45, 0.26, and 0.04 pg TEQ/g fat in domestic beef, pork and chicken, respectively and 0.11, 0.17 and 0.04 pg TEQ/g fat in imported beef, pork and chicken, respectively. Relatively higher PCDFs were found in domestic products. The imported beef, originating mostly from USA, showed the highest level of PCDDs. The imported pork, mostly from European countries, showed a greater concentration of PCDFs than PCDDs. Because of the high level of 2,3,4,7,8-PeCDF in domestic beef, the sum of TEQ PCDD/Fs in domestic beef was more than five times higher than that of imported beef although the PCDD/Fs concentrations by weight of imported beef was 1.6 times higher than that of domestic beef. As determined from the TEQ level, the concentration of PCDDs in imported beef was six times higher than that in domestic beef. It is hypothesized that the sources of PCDD/Fs contamination differed between domestic and imported meat. The sources of PCDDs include kaolinite clay, ball clay, chemical impurities, or pentachlorophenol (Gaus et al., 2001; Prange et al., 2002; Ferrario et al., 2004; Huwe et al., 2004a), whereas PCDFs are generally produced from incineration processes (Cleverly et al., 1997; Chang-Chien et al., 2001; Kim et al., 2005; Yu et al., 2006). PCDFs were dominant in domestic beef as they are in incineration emissions. The congener patterns obtained from domestic beef samples closely

Table 2
Lower and upper bound concentrations of PCDDs and PCDFs (pg TEQ/g fat) in meat

PCDD/Fs	Domestic			Imported			Domestic + imported		
	Beef (*n* = 38)	Pork (*n* = 48)	Chicken (*n* = 33)	Beef (*n* = 98)	Pork (*n* = 58)	Chicken (*n* = 8)	Beef (*n* = 136)	Pork (*n* = 106)	Chicken (*n* = 41)
Lower bound (non-detects = 0)									
PCDDs	0.01	0.02	0.001	0.06	0.02	nd	0.05	0.02	nd
PCDFs	0.40	0.21	0.002	0.01	0.12	0.002	0.12	0.16	0.002
PCDD/Fs	0.41	0.23	0.003	0.07	0.14	0.002	0.17	0.18	0.002
Upper bound (non-detects = LOD)									
PCDDs	0.04	0.04	0.03	0.09	0.05	0.03	0.08	0.05	0.03
PCDFs	0.41	0.22	0.01	0.02	0.12	0.01	0.13	0.17	0.01
PCDD/Fs	0.45	0.26	0.04	0.11	0.17	0.04	0.21	0.22	0.04

n = number of samples; nd = not detected.

resembled those obtained from stack gas or fly ashes from incinerators in Korea (Yu et al., 2006). However, metabolic and accumulation processes of these congeners in the animal may change the pattern of PCDD/Fs in the beef samples examined.

Figs. 1 and 2 show the congener profiles of TEQ levels. The congener profiles differed between domestic and imported beef. However, the congener profiles were similar in domestic and imported pork although the toxicological levels were different. Pork was mostly imported from countries in Europe. The majority of beef and chicken was imported from non-European countries. PeCDFs and HxCDFs were dominant congeners in domestic beef and pork but 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, and 2,3,7,8-TCDF of relatively high TEFs were not found. Concentrations of 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF, and OCDD were highest in beef, pork, and chicken, respectively. Guruge et al. (2005) reported that the concentrations of PCDD/Fs in the fat of domestic pigs and chickens in Japan were 0.63 pg TEQ/g lipid and 1.71 pg TEQ/g lipid,

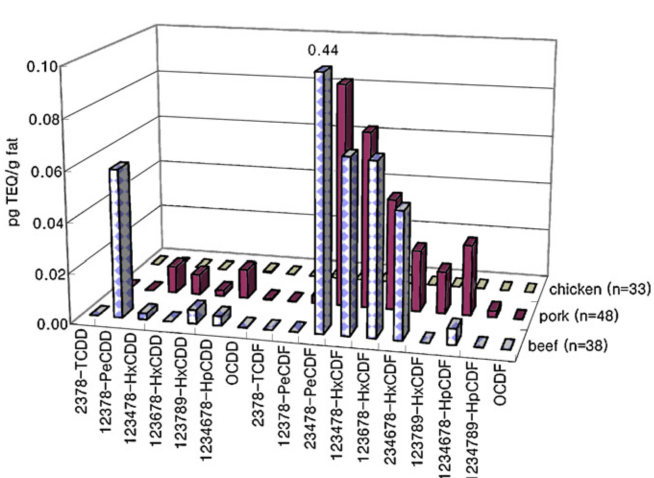

Fig. 1. Congener profiles for TEQ levels of PCDD/Fs in domestic beef, pork, and chicken. 2,3,4,7,8-PeCDF was found in the highest concentration (0.44 pg TEQ/g fat) in beef.

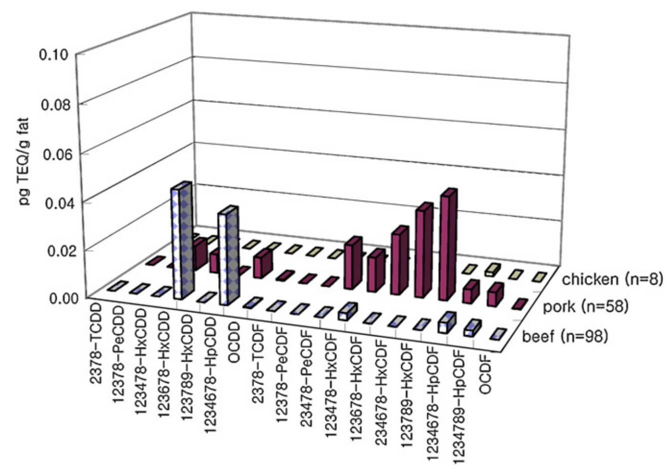

Fig. 2. Congener profiles for TEQ levels of PCDD/Fs in imported beef, pork, and chicken.

respectively. This trend was the reverse of that observed in domestic pork and chickens in South Korea. A survey in the United States using the International Toxic Equivalence Factor (I-TEFs) showed that the average concentrations of PCDD/Fs in 63 samples of beef and in 56 samples of market hogs were 0.35 and 0.42 pg TEQ/g fat, respectively (Ferrario et al., 1996; Lorber et al., 1997). It is difficult to compare the levels of PCDD/Fs reported in the literature because almost all samples were different in terms of the environmental conditions of the livestock and their feed regime. In addition, the final concentrations were calculated differently and were based on the use of either WHO-TEFs or I-TEFs.

Concentrations and estimated dietary intakes of PCDD/Fs are presented in Table 3. Lower and upper bound of mean concentrations derived from the sum of the concentrations found in samples of domestic and imported meat were used to calculate the dietary intakes. Information regarding the daily consumption and fat content of meat was obtained from the Food Balance Sheet published by the Korea Rural Economic Institute (2003). The daily consumption of beef, pork, and chicken was 85 g/day per

Table 3
Average concentrations and estimated dietary intakes of PCDD/Fs

Meat	Concentration (pg TEQ/g fat)		Daily consumption[a] (g)	Fat content[a] (g)	Dietary intake of PCDD/Fs (pg/kg bw/day)[c]	
	Lower bound	Upper bound			Lower bound	Upper bound
Beef	0.17	0.21	22.41 (1.5)[b]	2.06 (2.4)[b]	5.84×10^{-3}	7.21×10^{-3}
Pork	0.18	0.22	45.21 (3.0)	7.93 (9.3)	23.8×10^{-3}	29.1×10^{-3}
Chicken	0.002	0.04	17.21 (1.2)	1.72 (2.0)	0.06×10^{-3}	1.14×10^{-3}
Sum			84.83 (5.7)	11.71 (13.7)	29.7×10^{-3}	37.5×10^{-3}

[a] Data from Food Balance Sheet published by Korea Rural Economic Institute (2003).
[b] In parentheses: percentage of the total consumption of food or fat.
[c] bw: Body weight = 60 kg.

capita which represented 5.7% of the average total food consumption. The consumption of meat fat made up 13.7% of the daily total fat intake. The lower bound of the dietary intake of PCDD/Fs by Koreans through beef, pork, and chicken consumption was 0.03 pg TEQ/kg bw/day and 1.8 pg TEQ/person/day for an adult weighing 60 kg. The upper bound of the dietary intake was 0.04 pg TEQ/kg bw/day and 2.4 pg TEQ/person/day for an adult. The values represent a low intake when compared with the tolerable daily intake (TDI), 1–4 pg TEQ/kg bw/day established by WHO. The dietary intake of PCDD/Fs had changed slightly since 2000 (Lim et al., 2002). Intake through pork had increased with the increasing rate of consumption and residual levels of PCDD/Fs in pork. The relative decrease in the consumption rate and residual levels of PCDD/Fs in chicken was probably responsible for the decrease in intake of PCDD/Fs from this type of meat. The WHO-TDI included coplanar PCBs and the dietary intake of coplanar PCBs (PCB-77, 126, and 169) from beef, pork, and chicken in Korea was calculated to be 0.05 pg/WHO-TEQ/kg bw/day using the TEQ values from Kim et al. (2004). The dietary intake of coplanar PCBs was 1.7 times greater than that of PCDD/Fs. This indicated that the selection of compounds for the total dietary study is an important factor. However, coplanar PCBs were excluded in this study because coplanar PCBs were not analyzed in imported meat for the national quarantine service program. Fürst (2001) also emphasized the importance of coplanar PCBs for assessing human exposure. The concentrations found in dairy products were 0.77 pg/g fat for PCDD/Fs and 27.7 pg/g fat for coplanar PCBs (77, 126, and 169). Huwe et al. (2004b) found, however, 0.75 pg/g lipid of PCDD/Fs and 0.14 pg/g lipid of coplanar PCBs (77, 126, and 169) in beef and 0.16 pg/g lipid of PCDD/Fs and 0.03 pg/g lipid of coplanar PCBs in market hogs in the USA. In addition, the levels of PCDDs were much higher than that of PCDFs. The major reason for this might be the different sources of contamination. When studying human exposure to this type of contamination it is difficult to compare the results from different studies regardless of whether dioxins (PCDD/Fs) alone are monitored or whether dioxin-like PCBs are included in the study. This is primarily due to differences in the food group being studied, TEF, consumption data, and the method of calculation for non-detectable quantities. Several studies have reported dietary intake from different aspects. In the USA over 60% of total dietary intake is made up of beef, pork, and poultry. The levels ranged from a low value of 0.081 pg/I-TEQ/kg bw/day to a high of 2.174 pg/WHO-TEQ/kg bw/day (Schecter et al., 1994). The rate of consumption of beef, pork, and poultry in the USA is 147 g/day. The estimated daily intake of PCDD/Fs through meat and meat products by the general population living near a hazardous waste incinerator in Catalonia, Spain was reported to be 5.6 pg WHO-TEQ/day (Bocio and Domingo, 2005). Meat and meat products made up 8.8% of the total amount of food consumed in the population. Another study from Spain reported the dietary intake of dioxins to be ca. 35.0 ± 8.06 pg WHO-TEQ/day from pork and chicken meat (Fernández et al., 2004). Based on this study and previous reports, the dietary habits of each country should be taken into account in any study investigating human exposure levels and regulations regarding residual limit of PCDD/Fs from food.

4. Conclusions

The toxicological levels of PCDD/Fs were very low in beef, pork, and chicken in Korea. Different congener profiles were found according to the type of meat and the origin of product. PCDFs were more dominant than PCDDs in domestic beef and pork. The background levels of PCDD/Fs in beef, pork, and chicken were well below the European Union maximum residual limit. The estimated dietary intake of PCDD/Fs was within the TDI established by the WHO. The findings show that current background exposure is within safe levels, however monitoring should be continued to maintain public safety with regard to the consumption of food of animal origin.

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