Pak. J. Sci. Ind. Res. 2010 **53** (4) 192-197

Characteristic Trend of Persistent Organochlorine Contamination in Imported Red Kidney Beans

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(received July 16, 2009; revised April 28, 2010; accepted May 3, 2010)

Abstract. Residual level of persistent organochlorines (OC) such as ΣHCH, dieldrin and endrin were measured in red kidney bean samples from consignments imported from Ethiopia during 2004-2006. OCs, mainly ΣDDT and ΣHCH along with breakdown products (>65%), were detected in 80% of the samples analyzed and the highest concentration was 37 ng/g. In 2004, 20% and in 2006, 40% samples were found contaminated with detectable levels of OCs. Beta-HCH, however, was not detected in any sample in 2004 and HCB, in 2005. In 2004, average residual concentration of OCs in individual sample was 0.03-0.180 mg/kg and in 2005, 0.004-0.09 mg/kg.

Keywords: red kidney bean, organochlorine pesticides, Ethiopia, GC/ECD analysis

Introduction

Pakistan imports red kidney bean (RKB) from developing countries such as Ethiopia for human consumption as a source of protein, carbohydrates, dietary fiber, calcium, magnesium, zinc, iron, copper, phosphorus, potassium and vitamins.

Pakistan Export Promotion Bureau has implemented WTO standards for the import and export of agricultural products. Accordingly, pesticide residues are not permitted beyond the permissible limits of 0.1 ppm for ΣOCs including DDT. Pakistan Export Promotion Bureau has issued a list of pesticides (Table 1) which are banned in Pakistan (Mazari, 2005).

In the present work, 50 samples of red kidney been (RKB) were screened for a total of 37 pesticides and the results were compared with the standard values. Calibration curves of working standards were used to evaluate the linearity of the gas chromatograph response each day of analysis and pesticide residues were quantified based on these external standards.

Materials and Methods

The analytical method was validated and based on AOAC (2005).

Chemicals. High purity pesticide grade solvents (hexane, dichloromethane) and certified ACS reagents were used as received, but reagent grade acetone and hexane were distilled and evaluated by GC. Florisil was activated at 675 °C for one hour and deactivated with few drops of water with gentle shaking before use.

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Table 1. List of pesticides banned in Pakistan

Active ingredients	Formulations	Pesticides not registered
B.HC	Dichlorvos (above 500 g/L)	Aldrin (POP/PIC)
Binapacryl	Phophamidon (above 500 g/L)	Mirex (POP)
Bromophos ethyl	Methamidophos (above 600 g/L)	Chlordane (POP/PIC)
Captafol	Phophamidon (above 500 g/L)	Dinoseb (PIC)
Chlordimeform		Ethylene-di-bromide (PIC)
Chlorobenzilate		Parathion (PIC)
Chlorthiophos		Fluroacetate (PIC)
Cyhexatin		
Dalapon		
DDT		
Dibromochloropropane + Dibromochloropropene		
Dicrotophos		
Dieldrin		
Disulfoton		
Endrin		
Ethylene dichloride + Carbontenachloride		
Leptophos		
Mercury compound		
Mevinphos		
Toxaphene		
Zineb		
Heptachlor		
Methyl Parathion		
Dibromochloropropane + Dibromochloropropene		

Pesticide standards. A mixture of 37 pesticide standards was purchased from Dr. Ehresnstorfer's Laboratory, Germany and certified reference material (IAEA-406) from International Atomic Energy Agency. Stock solutions of pesticides (Table 2) were prepared using pesticide grade solvents.

Spiking solutions for measuring method efficacy (percent recovery) were prepared from stock solutions. Calibration standard solutions for at least three concentrations were also prepared from stock solutions in hexane. All stock, spiking and calibration standards were stored at 4 °C.

Sampling. Fifty (50) samples (approximately 200 g each) of red kidney bean were collected from open market, Karachi, between June 2004 and July 2005 for pesticide residue analysis. A sub-sample of 50 g each was ground in a grinding mill, passed through a 30 mesh sieve, and the fine flour was stored in desiccator at room temperature (25-30 °C).

Table 2. Recovery of pesticides from red kidney bean samples (n= 10)

Organochlorine	Recovery (%)	RSD (%)	LOD (mg/kg)	LOR (mg/kg)
alpha-HCH	95.4	4.6	0.0310	0.0940
HCB	97.8	5.1	0.0240	0.0729
beta-HCH	92.4	5.0	0.0304	0.0920
gamma-HCH	98.5	4.1	0.0292	0.0886
delta-HCH	97.9	5.2	0.0024	0.0072
epsilon-HCH	95.4	4.5	0.0540	0.1637
2,4,4-Trichlorobiphenyl	97.8	5.2	0.0607	0.1840
Heptachlor	92.4	6.0	0.0152	0.0461
2,2,5,5-Tetrachlorobiphenyl	98.5	4.3	0.1968	0.5963
Aldrin	97.9	5.4	0.1584	0.4800
Isodrin	95.4	4.7	0.0072	0.0219
Heptachlor-exo-epoxide (cis-isomer B) + Oxy-chlordane	97.8	5.2	0.0236	0.0716
Heptachlor-*endo*-epoxide (trans- isomer A)	92.4	6.0	0.0266	0.0806
cis-Chlordane(alpha)	95.4	4.7	0.0310	0.0940
Oxy-chlordane	97.8	5.2	0.003	0.0729
trans-Chlordane(gamma)	92.4	6.0	0.002	0.0920
2,4-DDE	98.5	4.3	0.003	0.0886
2,2,4,5,5-Pentachlorobiphenyl	97.9	5.4	0.002	0.0072
cis-Chlordane(alpha) + alpha-Endosulfan	95.4	4.7	0.003	0.1637
4,4-DDE	97.8	5.2	0.002	0.005
Diedrin	92.4	6.0	0.003	0.01
2,4-DDD	95.4	4.7	0.002	0.006
2,2,4,4,5,5-Hexachlorobiphenyl	97.8	5.2	0.007	0.02
4,4-DDT + 2,2,3,4,4,5-Hexachlorobiphenyl	92.4	6.0	0.003	0.01
Methoxychlor	98.5	4.3	0.007	0.02
2,2,3,4,4,5,5-Heptachlorobiphenyl	97.9	5.4	0.003	0.01
Methoxychlor	95.4	4.7	0.007	0.02
chlorpyrifos	97.8	5.2	0.003	0.01
Mirex	92.4	6.0	0.003	0.008
Heptachlor-endo-expoxide	98.5	4.3	0.002	0.005
Heptachlor-exo-epoxide (cis-isomer B) + Oxy-chlordane	97.9	5.4	0.003	0.01
Endrin+beta-Endosulfan	95.4	4.7	0.002	0.005
Dichlorvos	97.8	5.2	0.003	0.01
2,2,3,4,4,5-Hexachlorobiphenyl+4,4-DDT	92.4	6.0	0.002	0.006
2,2,4,4,5,5-Hexachlorobiphenyl	98.5	4.3	0.007	0.02

Extraction: A sample of ground RKB (20-50 g) in duplicate, including spiked sample and blank, was blended with a mixture of 350 mL H_2O (double distilled) + CH_3CN (7+13) at high speed in a stainless steel blender for 5 min, filtered with suction through sintered Buckner funnel (0.45 µ) into 500 mL suction flask and the filtrate of residues was transferred to petroleum ether. To the measured volume (with a measuring cylinder) of extract/filtrate 100 mL petroleum ether was added and transferred to one liter separator. It was shaken vigorously for 1-2 min and 10 mL saturated NaCl solution and 600 mL H_2O was added in separating funnel along with approx. 15 g anhydrous sodium sulphate (Na_2SO_4). It was shaken vigorously for avoiding prolonged soaking of Na_2SO_4 then the solvent was concentrated to 10-15 mL in K-D concentrator and the solution was transferred directly to Florisil column for clean up prior to determination (AOAC, 2005).

Clean up. ***Column chromatography.*** A glass column (22 mm id) was packed to 10 cm length with activated Florisil and topped with anhydrous Na_2SO_4 to additional 1 cm. The column was washed with 40-50 mL petroleum ether (40-60 °C) but was not allowed to dry. The petroleum ether extract of sample was applied to the packed column of Florisil at about 5 mL/min with 200 mL of 6% and 15% petroleum ether-ether mixture. Each elute (50 mL) was concentrated to suitable definite volume (1 mL) on a rotary evaporator (Kudernadanish evaporator).

Sample extract was quantitatively transferred to the GC auto sampler vial.

Gas liquid chromatography. Determination of chlorinated pesticides was made on a Perkin Elmer gas chromatograph, Clarus-500 equipped with Ni-63 electron capture detector (ECD). Analysis was performed on a cross bond 5% biphenyl, 95% dimethyl polysilicone capillary column (30 m length, 0.35 mm ID and 0.50 µm df.) obtained from M/s Analytical Measuring System, sole agent of Perkin Elmer Pakistan. The oven was operated under temperature conditions: initial temperature at 100 °C (held for 5 min) increased at the rate of 4 °C/min to a final temperature of 240 °C (held for 10 min). Injector temperature was maintained at 250 °C and detector, at 300 °C.

Recovery studies. Recovery study was carried out for each of the 37 pesticides. A 20 g control (pesticide free) RKB sample (purchased from market and checked to be pesticide residue free) was spiked with appropriate standard solution to the final concentration of 0.05-0.50 mg/kg, vigorously vortexed so as to distribute the added pesticide and extracted as detailed above. Data of average recovery and RSD (n = 10) for

organochlorine in the samples are shown in Table 2. In most cases, recoveries of the spiked pesticides ranged between 92 and 99% (5-20% SD) and represent triplicate analyses.

Quality assurance and quality control. Following the published procedures for quality assurance and quality control (EURACHEM, 1998; ICH Q2B 1996), the limit of detection (LOD) was established at a signal to noise ratio of 3; whereas the limit of quantification (LOQ) was 10 times that of LOD.

Results are calculated on the basis of the mean value of specimens. Recovery (%), RSD (%), LOD and LOR were calculated during the analysis; values are given in Table 2.

Results and Discussion

Physical observations. Beans were in very good appearance having bright red colour and typical kidney shape with very few broken beans.

Pesticide residues. Total 50 samples of RKB were analyzed for pesticides and metabolites including ΣHCH, ΣDDT (p,p-DDD, p,p-DDE, p,p-DDT), dieldrin and endrin and hexachlorobenzene (HCB). A number of pesticides have been determined by HPLC in a number of food commodities (Westbom *et al.*, 2008). In our studies, we used the GLC method.

Figure 1 summarizes the occurrence of pesticides detected in the samples; Table 3 and 4 summarize the residual concentration of chlorinated hydrocarbons in a number of red kidney bean samples collected from the consignments imported from Ethiopia during 2004 and 2005.

A comparison of data in Table 3 and Table 4 clearly indicates a reduction trend in OCs residue. For example, in 2004, in individual samples, average maximum residual concentration of chlorinated pesticides was 0.018 ppm that declined to 0.009 ppm in 2005. Average residual concentration for ΣHCH for 2004 was 0.004 ppm, but 0.003 ppm in 2005. HCB was not detected in 2004 and 2005 samples. Similarly, highest γ-HCH concentration of 0.012 ppm in 2004-declined to 0.010 ppm in 2005.

In 2004, average residual concentration for ΣDDT and its metabolites was 0.013 ppm, but decreased to 0.005 ppm in 2005. The o,p-DDD and pp-DDE were detected at 0.001-0.007 ppm in 2004, and 0.002-0.093 ppm in 2005. These values are comparable with the bean data analyzed in 1999 by Munshi *et al*. (2001). Concentrations of all major OCs investigated were less in 2005 samples as compared to those in 2004 (Fig. 2 and 3).

In 2004 samples, the highest residual concentration for p,p′-DDD was 0.007 ppm, but 0.093 ppm in 2005, which is below the maximum acceptable daily intake (ADI) set at 0.1 ppm (FAO/WHO, 1998).

Average residual concentration of endrin in 2004 and 2005 was 0.018 ppm (Fig. 2) and 0.002 ppm (Fig. 3), respectively- a decline. The same trend is seen in the highest concentration

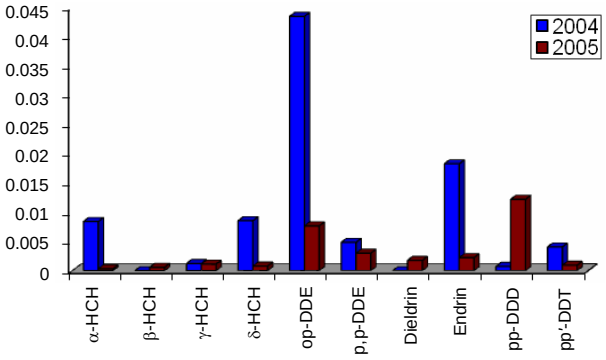

Fig. 1. Level of organochlorine contamination in red kidney bean during 2004-05.

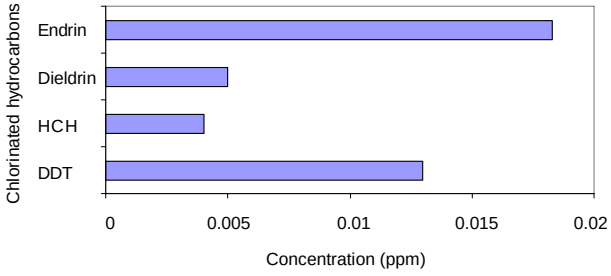

Fig. 2. Presence of DDT, HCH, dieldrin and endrin in red kidney bean during 2004.

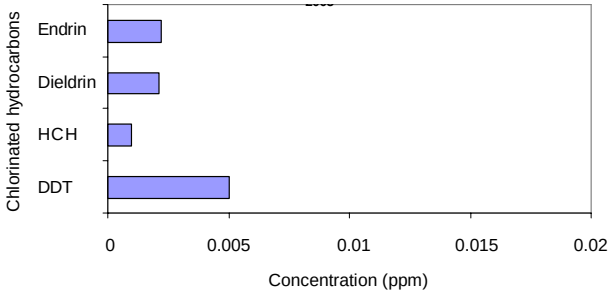

Fig. 3. Presence of DDT, HCH, dieldrin and endrin in red kidney bean during 2005.

Table 3. Residual concentration of chlorinated pesticide (ppm) (2005 samples)

α - HCH	HCB	β - HCH	γ - HCH	δ - HCH	op - DDE	p,p DDE	Dieldrin	o,p DDD	Endrin	pp - DDD	DDT
<LOD	<LOD	<LOD	<LOD	0.001	0.0145	<LOD	<LOD	0.004	<LOD	<LOD	<LOD
<LOD	<LOD	<LOD	<LOD	0.001	0	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
<LOD	<LOD	<LOD	<LOD	<LOD	0.013	<LOD	0.002	<LOD	<LOD	0.027	<LOD
<LOD	<LOD	<LOD	<LOD	<LOD	0.089	<LOD	0.002	<LOD	<LOD	0.017	<LOD
<LOD	<LOD	<LOD	<LOD	<LOQ	0.0105	<LOD	0.002	<LOD	<LOD	0.023	0.008
<LOD	<LOD	<LOD	<LOD	<LOD	0.0204	0.001	0.009	0.021	<LOD	0.030	<LOD
<LOD	<LOD	<LOD	<LOD	0.006	<LOD	0.006	0.003	<LOD	0.009	0.038	<LOD
<LOD	<LOD	<LOD	0.003	0.0021	<LOD	<LOD	0.016	<LOD	0.015	<LOD	0.012
<LOQ	<LOD	<LOD	<LOD	0.007	0.098	0.012	0.004	<LOD	0.015	0.093	<LOD
0.006	<LOD	<LOD	<LOD	0.0015	0.010	0.006	<LOD	<LOD	0.008	0.013	<LOD
0.004	<LOD	<LOD	<LOD	0.0013	<LOD	0.008	<LOD	<LOD	0.007	0.002	<LOD
0.005	<LOD	<LOD	0.01	<LOD	<LOD	0.004	<LOD	<LOD	<LOD	0.030	<LOD
0.003	<LOD	<LOD	<LOD	<LOD	0.021	<LOD	0.002	<LOD	0.008	<LOD	0.015
0.006	<LOD	<LOD	0.0018	0.006	0.013	0.008	<LOD	0.003	<LOD	<LOD	0.007
0.008	<LOD	<LOD	0.027	0.007	0.011	0.002	0.003	<LOD	<LOD	<LOD	0.011
0.012	0.006	<LOD	0.035	0.006	0.013	0.003	0.006	<LOD	<LOD	<LOD	0.013
0.022	0.003	<LOQ	0.002	<LOD	0.010	<LOD	0.001	0.008	0.007	0.014	0.044
<LOD	0.005	<LOD	0.005	<LOD	0.005	0.008	<LOD	<LOD	0.007	0.013	<LOD
<LOD	0.006	<LOD	<LOD	<LOD	0.014	<LOD	<LOD	0.001	0.004	<LOD	<LOD
<LOD	<LOD	<LOD	<LOD	<LOD	0.022	<LOD	0.002	0.0061	0.087	0.004	<LOD
<LOQ	<LOD	0.002	<LOD	<LOD	0.003	<LOD	<LOD	0.005	<LOD	0.003	<LOD
<LOD	<LOD	<LOD	<LOD	<LOD	0.082	0.005	<LOD	0.017	0.001	<LOD	<LOQ
<LOD	0.002	<LOD	0.002	0.001	0.015	0.009	<LOD	0.008	0.004	<LOD	0.006
0.004	0.005	0.007	<LOQ	0.010	0.013	0.001	<LOD	0.015	0.021	0.004	<LOD
0.008	0.006	0.006	0.010	0.006	0.006	0.014	0.010	0.042	0.016	0.002	<LOD

Table 4. Residual concentration of chlorinated pesticide (ppm) (2004 samples)

α - HCH	β - HCH	γ - HCH	δ - HCH	o,p'- DDD	op - DDE	Dieldrin + p,p- DDE	Endrin	pp - DDD	pp'- DDT	o,p'- DDT
<LOD	<LOD	0.003	<LOD	0.010	0.017	<LOD	<LOD	0.003	0.004	0.005
<LOD	<LOD	<LOD	<LOD	0.010	0.024	0.002	<LOD	<LOD	0.006	<LOQ
<LOD	<LOD	<LOD	0.004	<LOD	0.034	0.0083	<LOQ	<LOR	<LOQ	0.006
<LOD	<LOD	<LOD	<LOD	0.005	0.016	<LOD	0.007	<(LOQ)	0.002	0.012
<LOD	0.012	<LOD	<LOD	0.004	0.025	<LOQ	0.0003	<LOD	<LOD	0.006
<LOD	<LOD	<LOD	<LOD	0.002	0.025	<LOD	<LOQ	<LOD	<LOD	0.006
0.042	<LOD	<LOD	0.025	<LOD	0.064	0.004	0.050	<LOD	0.003	0.005
0.042	<LOD	<LOD	0.025	<LOD	0.064	0.004	0.051	<LOD	0.003	0.005
<LOD	<LOD	0.009	0.025	<LOD	0.072	0.004	0.044	0.007	<LOD	0.012
<LOD	<LOD	0.034	<LOD	<LOD	0.015	0.003	0.0285	<LOD	<LOD	0.005
0.041	<LOD	<LOQ	<LOD	<LOD	0.002	<LOD	<LOD	<LOD	<LOQ	<LOD
<LOD	<LOD	0.001	<LOQ	0.003	<LOD	<LOD	<LOD	<LOR	<LOD	<LOD
0.021	<LOD	<LOD	0.005	0.004	<LOD	0.004	<LOD	0.004	0.003	<LOD
<LOD	<LOD	<LOD	<LOD	0.001	<LOD	<LOQ	0.004	0.005	<LOD	<LOD
0.023	0.010	<LOD	<LOQ	0.005	0.003	<LOQ	0.006	0.003	<LOD	0.006
<LOD	<LOD	0.003	0.005	<LOD	0.002	0.005	0.003	<LOD	<LOD	0.005
0.042	<LOD	<LOD	<LOD	<LOD	<LOD	0.004	<LOD	<LOD	<LOD	<LOD
0.061	0.010	<LOD	<LOD	<LOD	0.006	<LOD	<LOD	0.002	<LOD	0.003
0.010	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.008	0.004	<LOD	<LOD
<LOD	<LOD	0.004	0.006	<LOQ	<LOD	<LOD	<LOD	0.001	0.012	<LOQ
<LOD	<LOD	<LOD	<LOQ	0.004	<LOD	<LOD	<LOD	<LOD	0.005	0.004
0.010	<LOD	<LOD	<LOD	<LOQ	0.004	<LOD	<LOD	<LOD	0.006	<LOD
0.020	<LOD	<LOQ	<LOD	<LOD	<LOQ	0.003	0.005	<LOD	<LOD	<LOD
<LOD	0.021	<LOD	<LOD	<LOQ	<LOD	0.003	0.006	<LOD	<LOD	0.005
0.030	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.004	<LOD	0.006	0.003

<LOD = lower than detection limit; <LOQ = lower than quantification limit; <LOR = lower than reportable value.

as well: in 2004 it was 0.028 ppm which reduced to 0.021 in 2005. Average residual concentration of dieldrin + p,p′-DDE in 2004 was determined as 0.005 ppm which in 2005 reduced to 0.003 ppm which is very low as compared to the tolerance limits. Similar studies were carried out previously on locally grown bean and vegetables. In 1999 analysis, no pesticide was detected in celery, cucumber, sugar beet and tomato collected from local markets in Karachi (Munshi *et al*., 2001). Residue data from that study are reported in Table 5 and 6.

Table 5. Pesticide residues in general food samples of Karachi Market in 1999

Commodity	Number of samples	Pesticides detected	Frequency (%)	Range (ppm)	Tolerance (ppm)
Celery	16	Endrin	26	0.10 - 0.80	15.0
		Endosulfane	30	0.05 - 0.90	5.0
Cucumber	35	Heptachlor	14	0.007-0.18	0.40
		Alpha-BHC	21	0.08 - 0.40	5.0
		Dieldrin	19	0.02 - 0.07	0.10
Sugar beet	19	DDT	2	0.03-0.05	0.2
Tomato	10	Endrin	6	0.02-0.04	0.50

Table 6. Food monitoring: Tolerance

Commodity	Pesticide	Highest concentration (ppm)	Tolerance (ppm)
Celery	Heptachlor	0.30	15.0
	Endosulfane	0.20	5.0
Cucumber	HCB	0.12	0.4
	Alpha- BHC	0.10	5.0
	Dieldrin	0.07	0.1
Sugar beet	DDT	0.03	0.2

Each group of chlorinated hydrocarbons in all of the samples was found to be within the tolerance limits as reported in literature (FAO/WHO, 1998). The Pest Management Regulatory Agency (PMRA) of Pakistan and Export Promotion Bureau have recently approved applications to amend the registration of environment-friendly pesticides in order to allow their use for the control of pests of cereals, rice, black beans, mung beans and white and red kidney beans as a post-emergent treatment. Griffitt and Szorik (1989) in PMRA determined that MRL of 0.1 ppm for each pesticide including its metabolites in red kidney bean and mung bean would not pose an unacceptable health risk to the public. These limits are in agreement with the MRLs set by the Joint FAO/WHO (1998).

The PMRA has also been requested to establish maximum residue limit (MRL) for residues of environment-friendly pesticides and its metabolites resulting from their use in dry beans (which include black bean, cranberry bean and white bean), dry lima bean, kidney bean and mung bean, in order to permit the sale of food containing these residues, after the review of all available data.

Endosulfan was found in about 30% of samples at level of 0.05 to 0.9 ppm. BHC, dieldrin, and endrine were detected in cucumber. DDT was found at a concentration of 0.03 ppm in one of the 36 (2%) sugar beet samples analyzed.

These problems are also represented by accumulation of organochlorine pesticide (OCs) residues in different environmental samples and hosting of at least 50,000 tons of obsolete pesticides, as well as tens of thousands of tons of contaminated soil, within the framework of the Africa Stockpiles Program (Westbom *et al*., 2008).

Conclusion

This study concluded that OC residues in imported RKB samples were below the recommended maximum residue level established by the Joint FAO/WHO (1998) and should not pose health issues if consumed as part of normal diet. Further it ensures the safety of imported RKB in Pakistan. It would be worthwhile to conduct another survey in 2010-2011 to ensure that OC concentration is declining and that there has not been any misuse of banned OCs locally and/or in countries which exports grains/pulses to Pakistan.

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