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## RESIDUES OF CHLORINATED ORGANIC PESTICIDE IN SEDIMENTS AND FISH FROM THE EGYPTIAN RIVER NILE

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### ABSTRACT

Persistence of the residue of organo chlorine pesticides (OCPs) became a great task due to long lasting effect in environment. Subsequently the monitoring of these residue is a very important issue. The present study detect the persistence of OCP in Benisuef and Al Qalyubia governorates ecosystem in Egypt during 2018 and 2019.

Six different studied sites in two governorates Benisuef and AlQalyubia (three sites of each governorate) were investigated. The residues were determined in sediment and fish samples. The samples were analyzed by gas chromatography (GC) with electron capture detector (ECD).

Generally, total levels of OCPs residues (ppm) in sediments were found to be higher in Benisuef governorate comparing with AlQalyubia governorate in autumn and winter seasons, in spring season the residue detected only in Benisuef governorate, whereas in summer season were found to be only in AlQalyubia governorate. OCPs in fish muscles showed high levels in Benisuef governorate comparing with Al Qalyubia governorate in autumn season, in winter season it was higher in AlQalyubia governorate comparing with Benisuef governorate. While, in spring and summer seasons were found to be only in Benisuef governorate.

The current study reveal higher level of OCPs in both sediment layer and fish muscles. Which necessitates presence of intensive plan for monitoring water quality of River Nile regionally and seasonally.

**Keywords :** Persistence; Pollution; Residue; Organochlorine pesticides (OCPs).

### Introduction

Pollution is one of the serious problems that faces human societies in the whole world especially in the developing countries. Where the human activities can cause many problems in both water quality disruption in fish health, in addition to deleterious effects on human's environments and resources which makes the problem worse El-Mekkawi *et al.* (2009). Use of good bio-indicators for environmental pollution monitoring is important issue, fish is a good bio-indicator for detection of pollution in the aquatic ecosystem due to its direct exposure to pollutant and indirect via the food chain Sethuraman *et al.* (2013). Organochlorine pesticides are one of the chlorinated hydrocarbon derivatives. One of the most difficult problems related to these pesticides is their stability in both fresh and salt water furthermore it's resistant to photo degradation Jayaraj *et al.* (2016). In addition to the high persistence, the low polarity, low aqueous solubility and high lipid solubility (lipophilicity) result in bioaccumulation in the food chain carry a great risk to human health and the environment globally, with particular to the aquatic ecosystem, posing a serious threat to its aquatic organisms, including fish (Afful and Anim, 2010). With concern to human health organochlorine pesticides can cause many health defects as cancer and reproductive problems Sethuraman *et al.* (2013), as the organochlorines can be carcinogenic in animal experimental as revealed by The World Health Organization (WHO) Shakeel *et al.* (2010). The great problem is that the organochlorines residue

still persist in the environment till now although their use have been banned since the 1970s, or restricted Cruz *et al.* (2003). Nile River in Benisuef and Al-Qalyubia governorates expose of many pollutants including untreated sewage, agricultural and industrial as well as urban wastes which increase the concentration of pollutants and organochlorine pesticides, which affects the physio-chemical and biological parameters in two governorates. So, this study aimed to establish the relation between of (Benisuef and Al-Qalyubia governorates ecosystem) and environmental pollution in along year seasons (Autumn, Winter, Spring and Summer) of 2018 – 2019.

### Materials and Methods

#### Study areas

In this study, the persistence of OCPs was determined in Benisuef and AlQalyubia governorates in Egypt during 2018 and 2019. Three sites of each governorate were investigated; the three studied sites in Benisuef Governorate were (El wasta, Beba and Elfashn), the three studied sites in AlQalyubia Governorate were (Shubra, El Qanater and KafrShokr).

#### Sampling

##### Sediment samples

About 2 kg of sediment from each selected site under the study were taken at the same time at a depth 5 cm of sediment surface. The samples were wrapped in aluminum foil after cleaning from live plants and water then chilled in the

ice box. Samples were air dried in dark for 48 hours before analysis El-Mekawi *et al.* (2009).

### Fish samples

Healthy and vigorous fish (*Tilapia niloticas* pp) were collected by fishermen with average weight of 2-2.5 kg from the each site at the same times then they were kept in thermo insulator box and transported to the laboratory. Several washing of fish samples with deionized water was done to remove the sediments and other materials. The viscera of fish be removed and a muscle tissue sample (10 g) was taken from the dorsal muscle and were wrapped in aluminum foil then identify and kept in deep freezer until analysis Essumang *et al.* (2009).

### Soxhlet Extraction

Fifty-gram of air-dried sediment were weighted, and homogenized in ceramic mortar with 20 g anhydrous sodium sulfate. The homogenized samples were placed in conical flask then orbital shaker was used for shaking samples two hours with 130 ml mixture of 30% acetone in n-hexane as modification instead of n-hexane only to be suitable in sediment extraction then extract was dried through 50 g of anhydrous sodium sulfate Leyva-Cardoso *et al.* (2003). The obtained organic phase was treated with acid activated copper granules to remove sulfur. The extract was evaporated to about 1 ml using rotary evaporator.

Ten-gram of fish tissue samples were analyzed for PCB congeners and OCPs following well-established techniques UNEP (1991); IOC(1993). Fish sample (10 g of wet weight) was placed in ceramic mortar, anhydrous sodium sulfate (30 g) was added, and the mixture was well homogenized. The mixture was transferred to a pre-cleaned extraction thimble and the dehydrated tissue was extracted with 200 ml of 50% methylene chloride in n-hexane for 8 hours in a Soxhlet apparatus cycling 5-6 times per hour. The extracted solvents were concentrated with rotary evaporator to about 1 ml.

### GC analysis

The extracts were concentrated and injected into GC (Agilent 7890) equipped with a Ni63 ECD, a split/splitless injection inlet, capillary column capability, and a 7683A autosampler. Chemstation software was used for instrument control. GC analysis was conducted on a HP-5MS (Agilent, Folsom, CA) capillary column of 30 m, 0.25 mm id., 0.25  $\mu$ m film thickness. The oven temperature was programmed from an initial temperature 160 (2 min hold) to 240 °C at a rate of 5° C/min and was maintained at 240°C for 20 min. Injector and detector temperatures were maintained at 260 and 320 °C, respectively. Nitrogen was used as a carrier at flow rate of 3 ml/min.

### Target analytes:

Target analytes including: aldrin, o,p'-DDE, endosulfan, p,p' DDE, dieldrin,  $\alpha$ -HCH,  $\gamma$ -HCH, heptachlor, heptachlor epoxide and o, p'-DDT.

### Statistical analysis

Data obtained were statistical analysis using (ANOVA) test, Duncan Multiple Range Test, Fischer Exact Probability test, correlation and t-student test by SPSS 24 (2016).

## Results and Discussion

Fish are consider as good bioindicator for environmental monitoring as a result direct up take of contaminants from both water and food chain and more than that is the moderate ability of fish to metabolize organo chlorines subsequently fish is good reflection of contaminant load in the surrounding ecosystem Guo *et al.* (2008).

### Concentration of Chlorinated Organic Pesticide residues in Sediments

The concentration of residues of chlorinated organic pesticides was detected in sediment samples in four seasons; the table (1) & figure (1) represent the concentration of residues of chlorinated organic pesticides (ppm) in autumn season where the highest total level in Benisuef governorate was in Beba site, (0.148) ppm. While the highest total levels of OCPs residues in Al Qalyubia governorate was in Kafreshokr, (0.120) ppm. The frequency of occurrence for OCPs varied with congeners; aldrin was detected only at Elwasta in Benisuef governorate and at Shubra in AlQalyubia (0.086) and (0.083) ppm respectively, OP-DDE residues was detected only at Beba (0.031) ppm and Elfashn (0.033) ppm in Benisuef governorate, Endosulfan and PP-DDE residues were detected at Elfashn in Benisuef governorate (0.039) and (0.072) ppm respectively, dieldrin residues were found at Beba ( 0.117) ppm in Benisuef governorate and kafreshokr (0.120) ppm in Al-Qalyubia governorate and  $\alpha$ -HCH  $\gamma$ -HCH, heptachlor, heptachlor epoxide and OP-DDT were undetectable at all sites. This result reveal that the highest levels of OCPs residues was detected at Beba in Benisuef governorate.

The table (1) & figure (1) represent the concentration of residues of chlorinated organic pesticides (ppm) in winter season where the highest total level in Benisuef governorate was at Elwasta, (0.142) ppm. While the highest total levels of OCPs residues in AlQalyubia governorate was at El Qanater, (0.083) ppm. With respect to the frequency of occurrence for OCPs congeners; aldrin residues was detected at El Qanater and kafreshokr in AlQalyubia governorate at (0.083) and (0.079) ppm respectively, OP-DDE residues was detected at Elwasta (0.026) ppm and Beba (0.139) ppm in Benisuef governorate, and dieldrin residues (ppm) was detected only at Elwasta (0.116) ppm in Benisuef governorate. This result reveal that the highest levels of OCPs in winter season residues was detected at Elwasta in Benisuef governorate.

In spring season the table (2) & figure (1) represent the concentration of residues of chlorinated organic pesticides (ppm) where the OCPs residues were detected in Benisuef governorate only at Elwasta (0.124) ppm and Elfashn (0.089) ppm; OP-DDE residues was detected at Elwasta and El fashn (0.041) and (0.089) ppm respectively, and aldrin residues was detected at Elwasta (0.083) ppm.

With concern to the summer season the OCPs residues were found only in AlQalyubia governorate at Shubra (0.073) ppm; the only detected congeners is aldrin residue table (2) and figure (1).

The above result clears that the concentration of OCPs residues were higher in Benisuef governorate than with AlQalyubia governorate in autumn, winter and spring seasons where in summer season the OCPs residues were detected only at shubrain AlQalyubia governorate. With respect to the season of detection in Benisuef governorate the

highest concentration of OCPs residues were in autumn followed by winter and lowest concentration was in spring. But in AlQalyubia governorate the highest concentration of OCPs residues were in autumn followed by winter and lowest concentration was in summer.

**Concentration of Chlorinated Organic Pesticide residues in fish samples:**

The table (3) & figure (2) can show that the total concentration of OCPs residues in fish muscles in autumn season were detected in Benisuef governorate at Beba and El fashn, (0.275) and (0.212) ppm respectively, while in AlQalyubia governorate they were detected at El Qanater and kafreshokr (0.218) and (0.139) ppm respectively. The frequency of occurrence for OCPs varied with congeners;  $\alpha$ -HCH and  $\gamma$ -HCH residue were detected at El-Fashn, El-Qanater and kafreshokr at level of (0.066), (0.072), (0.068) ppm and (0.065), (0.066), (0.071) ppm respectively, also aldrin residue was detected at Beba in Benisuef governorate, El fashn and El Qanater at level of (0.077), (0.081) and (0.080) ppm respectively. While, levels of heotachlore epoxide and OP-DDE was detected only at Beba (0.056) and (0.142) ppm respectively.

The table (3) & figure (2) represent the concentration of chlorinated organic pesticides residues (ppm) in fish muscles in winter season where the highest total level was in AlQalyubia governorate at El Qanater (1.134) ppm, followed by Beba and El fashn in Benisuef governorate (0.165) and (0.150) ppm respectively and lowest level was at shubra in AlQalyubia governorate (0.091) ppm. With respect to the frequency of occurrence for OCPs varied with congeners; highest level of  $\alpha$ -HCH residue was detected at El Qanater, aldrin residue was detected only at El Qanater and  $\gamma$ -HCH residue was detected at Beba, El fashn and El Qanater (0.068), (0.069) and (0.082) ppm respectively.

Table (4) and figure (2) showed levels of OCPs residues in fish muscles in spring season at Elwasta and El fashn (0.141) and (0.238) ppm respectively; where the highest concentration level of  $\alpha$ -HCH and  $\gamma$ -HCH residues was detected at El fashn(0.166) and (0.072) ppm followed by Elwasta (0.072) ppm, (0.069) ppm in Benisuef governorate.

Table (4) and figure (2) showed levels of OCPs residues (ppm) in fish muscles in summer season at El wasta and

Beba (0.070) and (0.079) ppm respectively; levels of  $\alpha$ -HCH residue was detected only at Elwasta (0.070) ppm. While, aldrin residue was detected only at Beba (0.079) ppm in Benisuef governorate.

The above result clears that the concentration of OCPs residues in fish muscles were higher in Benisuef governorate than with AlQalyubia governorate in autumn, whereas in summer and spring seasons the OCPs detected only in Benisuef governorate. On the other hand in winter season the OCPs residues were higher in AlQalyubia governorate. With respect to the season of detection in Benisuef governorate the highest concentration of OCPs residues were in autumn followed by spring and winter while the lowest concentration was in summer. Butin AlQalyubia governorate the highest concentration of OCPs residues were in winter then autumn. While, the OCPs in spring and summer seasons were undetectable.

It is worth mentioning that the presence of OCPs residue without their metabolites is strong evidence of recent use of them Shalaby *et al.* (2018), especially that many previous studies has reported the concentrations of organochlorine pesticides in fish and sediment Yamashita *et al.* (2000). The sources of this residue may be related to wastewater from the densely populated area and agricultural wastes from cultivated areas El-Kady *et al.*(2017). Detection of such residue carry great risk to human health where the fish is one of most popular food El-Mekkwawi *et al.* (2009).

**Conclusion**

Organochlorine pesticides were detected in various sites of the study, with variation in the frequency of occurrence for OCPs among different congeners. The obtained result clears that both the sediment layer and fish are considered as important indicator in tested ecosystem because they contains significant level of OCPs residues in all of studied sites at in two governorates ecosystem (Benisuef and AlQalyubia). A recommendation and suggestions to intensive plan for monitoring water quality of River Nile regionally and seasonally. Strict application of the legislations concerning the protection of the River Nile and the environment, according to the laws 48/1982 and 4/1994.

**Table 1 :** Levels of organochlorine pesticides in sediments (ppm) at six sites in two governorates (Benisuef and AlQalyubia) in autumn and winter seasons

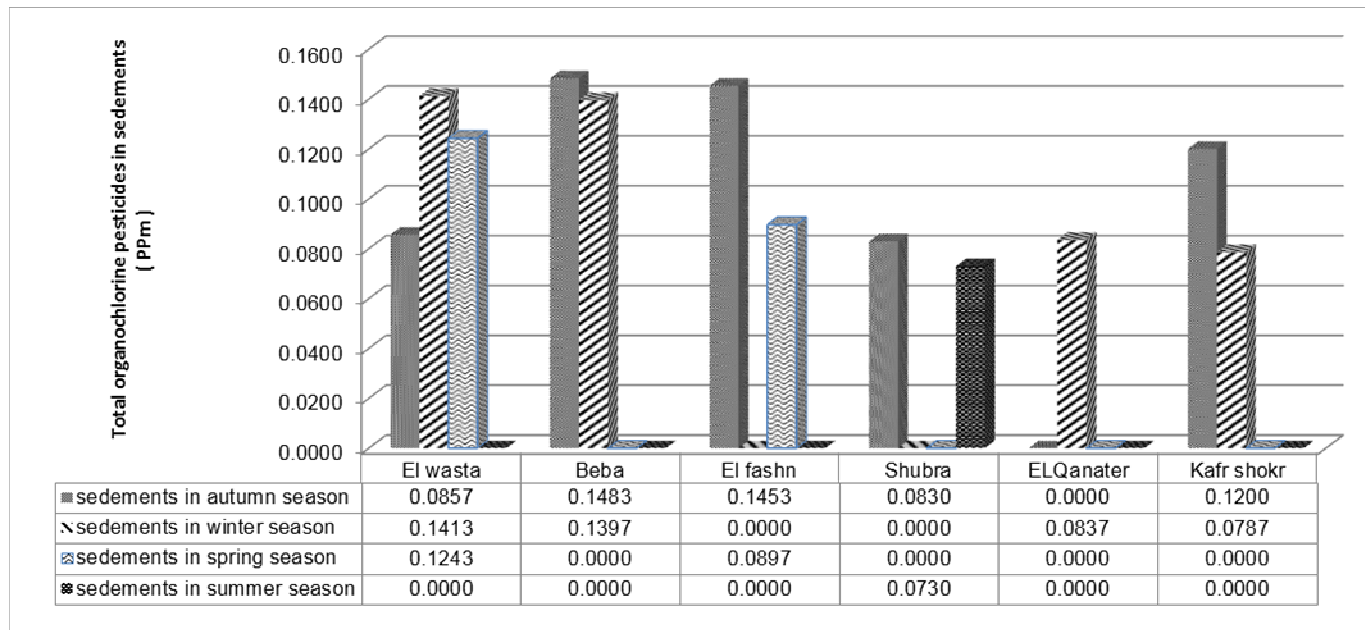
OCPs Region		Autumn season						Winter season			
		Aldrin	OP-DDE	Endosulfan	PP-DDE	Dieldrin	Total	Aldrin	OP-DDE	Dieldrin	Total
Benisuef Gov.	Elwasta	0.086±0.006	UD	UD	UD	UD	0.086±0.006a	UD	0.026±0.002a	0.116±0.003	0.142±0.005b
	Beba	UD	0.031±0.001	UD	UD	0.117±0.002	0.148±0.003c	UD	0.139±0.003b	UD	0.139±0.003b
	El fashn	UD	0.033±0.003	0.039±0.002	0.072±0.002	UD	0.144±0.005c	UD	UD	UD	UD
Al-Qalyubia Gov.	Shubra	0.083±0.003	UD	UD	UD	UD	0.083±0.003a	UD	UD	UD	UD
	El Qanater	UD	UD	UD	UD	UD	UD	0.083±0.002	UD	UD	0.083±0.002a
	Kafreshokr	UD	UD	UD	UD	0.120±0.003	0.120±0.003b	0.079±0.002	UD	UD	0.079±0.002a
F-calculated		0.144	0.800	-	-	0.377	48.799*	2.045	784.993*		88.728*

Data represented as mean ± SEUD = undetected

N = 3

a, b & c significant difference between similar litter using Duncan Multiple Range test for comparative of means at P < 0.05

\* Significant difference using ANOVA test at P < 0.05



**Fig. 1 :** Levels of total organochlorine pesticides(ppm) in sediments at six sites in two governorates (Benisuef and AlQalyubia) in autumn, winter, spring and summer seasons.

**Table 2 :** Levels of organochlorine pesticides (ppm) in sediments at six sites in two governorates (Benisuef and AlQalyubia) in spring and summer seasons.

Region \ OCPs		Spring season				Summer season	
		Aldrin	Heptachlor-epoxide	OP-DDE	Total	Aldrin	Total
Benisuef Gov.	Elwasta	0.083±0.003	UD	0.041±0.004a	0.124±0.007a	UD	UD
	Beba	UD	UD	UD	UD	UD	UD
	El fashn	UD	UD	0.089±0.003b	0.089±0.003b	UD	UD
Al-Qalyubia Gov.	Shubra	UD	UD	UD	UD	0.073±0.003	0.073±0.003
	El Qanater	UD	UD	UD	UD	UD	UD
	Kafrshokr	UD	UD	UD	UD	UD	UD
F-calculated		-	-	79.754*	20.067*	-	-

Data represented as mean ± SEUD = undetected

N = 3

a & b significant difference between similar litter using Duncan Multiple Range test for comparative of means at P < 0.05

\* Significant difference using ANOVA test at P < 0.05

**Table 3 :** Levels of organochlorine pesticides (ppm) in fish muscles at six sites in two governorates (Benisuef and AlQalyubia) in autumn and winter seasons.

OCPs Region		Autumn season						Winter season.			
		α-HCH	γ-HCH	Aldrin	Heptachlor-epoxide	OP-DDE	Total	α-HCH	γ-CH	Aldrin	Total
Benisuef Gov	Elwasta	UD	UD	UD	UD	UD	UD	UD	UD	UD	UD
	Beba	UD	UD	0.077±0.005	0.056±0.003	0.142±0.005	0.275±0.006c	0.097±0.004a	0.068±0.002a	UD	0.165±0.004b
	El fashn	0.066±0.003	0.065±0.002	0.081±0.004	UD	UD	0.212±0.007b	0.081±0.005a	0.069±0.004a	UD	0.150±0.001ab
Al-Qalyubia Gov	Shubra	UD	UD	UD	UD	UD	UD	0.091±0.004a	UD	UD	0.091±0.004a
	El Qanater	0.072±0.002	0.066±0.001	0.080±0.002	UD	UD	0.218±0.002b	0.096±0.039b	0.082±0.002b	0.087±0.003	1.134±0.041c
	Kafrshokr	0.068±0.006	0.071±0.002	UD	UD	UD	0.139±0.006a	UD	UD	UD	UD
F-calculated		0.533	3.508	0.242	-	-	95.010*	486.894*	5.542*	-	583.632*

Data represented as mean ± SEUD = undetected

N = 3

a, b & c significant difference between similar litter using Duncan Multiple Range test for comparative of means at P < 0.05

\* Significant difference using ANOVA test at P < 0.05

**Table 4** : Levels of organochlorine pesticides (ppm) in fish muscles at six sites in two governorates (Benisuef and AlQalyubia) in spring and summer seasons.

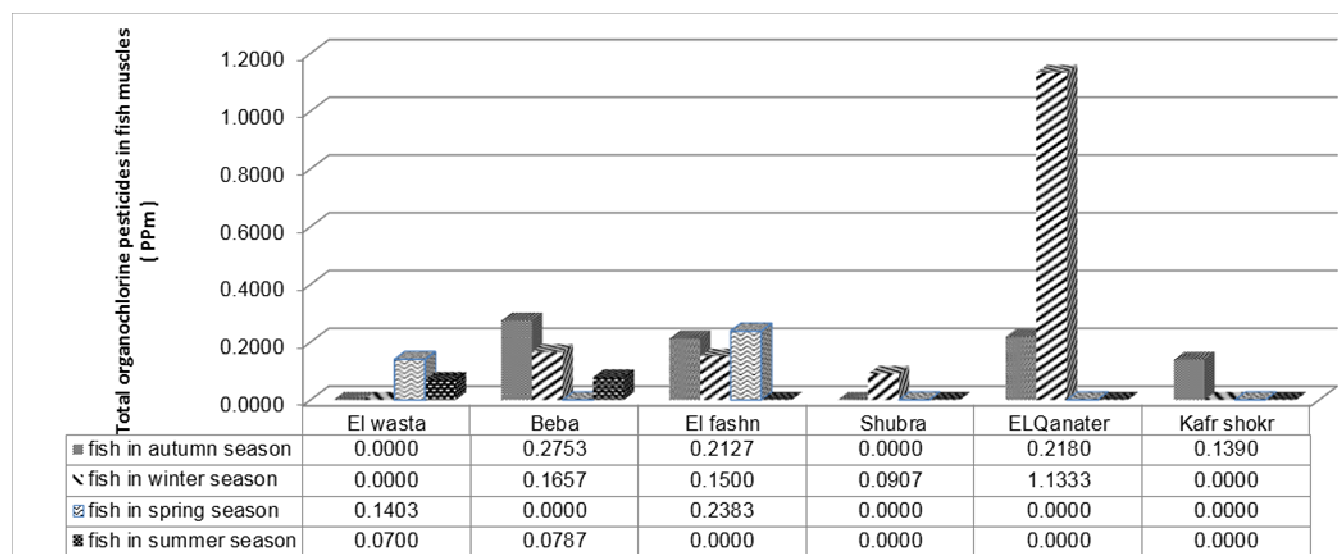
OCPs Region		spring season			Summer season		
		$\alpha$ -HCH	$\gamma$ -HCH	Total	$\alpha$ -HCH	Aldrin	Total
Benisuef Gov.	Elwasta	0.072± 0.002a	0.069± 0.002	0.141± 0.002a	0.070± 0.002	UD	0.070± 0.002
	Beba	UD	UD	UD	UD	0.079± 0.004	0.079± 0.004
	El fashn	0.166± 0.003b	0.072± 0.002	0.238± 0.005b	UD	UD	UD
Al-Qalyubia Gov.	Shubra	UD	UD	UD	UD	UD	UD
	El Qanater	UD	UD	UD	UD	UD	UD
	Kafrshokr	UD	UD	UD	UD	UD	UD
F-calculated		880.099*	1.513	336.327*	-	-	4.568

Data represented as mean  $\pm$  SEUD = undetected

N = 3

a&amp; b significant difference between similar litter using Duncan Multiple Range test for comparative of means at P &lt; 0.05

\* Significant difference using ANOVA test at P &lt; 0.05

**Fig. 2** : Levels of total organochlorine pesticides(ppm) in fish muscles at six sites in two governorates (Benisuef and AlQalyubia) in autumn, winter, spring and summer seasons.

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