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Simulating the Environmental Risks Resulted from Using Different Biocides in the Cooling System of Coastal Nuclear Power Plant

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Abstract: The using of biocides to the incoming seawater of the nuclear power plant, as important algaecide tool in order to prevent bio-fouling in cooling water system. In this study Egyptian coastal (Sidi Kirir) area was selected as a cooling water source, and once through pass cooling water system plant was also suggested as nuclear power cooling water type, where its measured pH and alkalinity data were suitable for using modeling software (Quantitative Structure Activity Relationships (QSARs), which it will be used to predict the aquatic toxicity resulting from using different types of non-oxidizing biocides (chlorophenol, trichlorophenol and quaternaryphenyl) and oxidizing biocides (sodium hyphochlorite, chlorine dioxide and ozone) in the suggested once through cooling water system. field study survey was done, water samples was taken, and Physicochemical and biological parameters were measured as Statistical correlations for different Kow were being done in-order to select the lower environmental risk biocide which would be used in cooling water treatment system of the suggested nuclear power plant. Study results showed that the lowest LC50 in Fish and Daphnia that found in case of using Quaternary phenyl biocide, while the highest LC50 value was showed in Chlorophenol values of fish and Daphnia. On the Other hand the lowest EC50 value of green algae was found when using quaternary phenyl biocide. This support the using of quaternary phenyl as low ecological risk biocide fouling treatment method in the cooling water system at the selected site Sidi Kirir region when using non-oxidixing biocides. In case of using oxidizing biocides, the results showed that lowest LC50 in Fish and Daphnia were found when using ozone biocide while the highest LC50 values obtained in fish and Daphnia when using chlorine dioxide, while in case of EC50 results showed that the lowest EC50 value in green algae was found when using ozone biocide while the highest EC50 value of green algae found when using chlorine dioxide biocide this support the using ozone as low ecological risk ecological biocide in the treatment of the cooling water system at the selected site .

Keywords: Biocides, nuclear power, cooling water system

1 Introduction

Using seawater in a once through cooling system as a type of cooling water system can lead to biofilm formation in the condenser tubes. Biocides are chemical substances used to control bio-fouling in heat exchangers by killing the living organisms. However, chlorine is the most widely used chemical for disinfecting in water treatment, but there is a high risk of the potential production of hazardous halogenated byproduct [1]. Biocide is also called germicide and algaecide, micro biocides or fungicide. According to the incomplete statistics, nearly 80 kinds of biocides, generally divided into oxidizing and non-oxidizing biocide two categories, are used in the current domestic industrial circulating cooling water. The oxidizing biocide used commonly includes chlorine, hypochlorite such as sodium hypochlorite etc, chlorine dioxide, ozone, bromine and bromide. The non-oxidizing biocide used commonly includes chlorine phenol such as double chlorophenol, trichlorophenol and pentachlorophenol, quaternary ammonium salt such as benzalkonium chloride, quaternary phenyl, organic amine such as rosin amine, etc, organic sulfur compounds such as methylene sulfur, copper salts such as copper sulfate and quaternary phosphorus salt [2,3]. There are some factors to be considered when considering biocides dosing in sea water include:1)Biocide application rates and frequency, 2)Use of bio-dispersants or other cleaners, 3) Retention time and water velocity through the system,4) Cooling system pH, chlorides, TDS, or other pertinent analytical parameters, including any process

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contaminant load or cooling system nutrients, 5) Microbiological flora load, including slimes, biofilms,…etc. 6)The compatibility and possible effects on other chemical inhibitors, 7) Cost comparisons of a proposed biocide with other possible starting-point biocides, 8) Existing corrosion, deposits and other reported problems [4,5]. Modeling of biocides can be carried out using generic data for chemical release. Estimates of environmental release are improved by gathering information on the release of biocides from specific processes to develop emission scenarios. Risk characterization is also conducted regarding animals kept and used by humans. The humaneness of biocidal products targeted at vertebrates is also considered, e.g. for biocides directed against rats [6,7] . ChV, or Chronic Value, is defined as the geometric mean of the no-observed-effect concentration (NOEC) and the lowest-observed-effect concentration (LOEC). This can be mathematically represented as:

ChV = 10^([log (LOEC × NOEC)]/2)[8] *……………(1)*

Median Lethal Concentration LC50 is a statistically derived concentration of a substance that can be expected to cause death in 50% of test animals. It is usually expressed as milligrams (mg) of substance per liter (L) water. Log Logarithm; a base of 10 is used in ECOSAR calculations. The Lowest Observed Effect Concentration LOEC of a substance that produced statistically significant effects(9) . The octanol / water partition coefficient (Kow) is defined as the ratio of a chemical's concentration in the octanol phase to its concentration in the aqueous phase of a two-phase octanol/water system.

Kow = Concentration in octanol phase / Concentration in aqueous phase [8] *….(2)*

Values of Kow are thus, unit less. The parameter is measured using low solute concentrations, where Kow is a very weak function of solute concentration. Values of Kow are usually measured at room temperature (20 or 25'C). The effect of temperature on Kow is not great - usually on the order of 0.001 to 0.01 Iog Kow units per degree - and may be either positive or negative. Measured values of Kow for organic chemicals have been found as low as 10^{-3} and as high as 10^{7}, thus encompassing a range of ten orders of magnitude. In terms of log Kow, this range is from -3 to 7. It is frequently possible to estimate log Kow with an uncertainty (i.e., method error) of no more than 10.1-0.2 Iog Kow units [9] The octanol/water partition coefficient is not the same as the ratio of a chemical's solubility in octanol to its solubility in water, because the organic and aqueous phases of the binary octanol/water system are not pure. Log Kow is a very important parameter for predicting the distribution of a substance in various environmental compartments (water, soil, air, biota, etc). Substances with high Log Kow values tend to adsorb more readily to organic matter in soils or sediments because of their low affinity for water. Chemicals with very high Log Kow values (i.e. >4.5) are of greater concern because they may have the potential to bio-concentrate in living organism [9]. For above reason, n-octanol/water partition coefficient (Kow) is used as a screening test for bio-accumulation test. The assumption behind this is that the uptake of an organic substance is driven by its hydrophobicity. For organic substances with a Log Kow value below 4.5 it is assumed that the affinity for the lipids of an organism is insufficient to exceed the bio-accumulation criterion i.e. a BCF value of 2000.For some groups of chemicals, such as metals and surface active compounds, Log Kow is not a valid descriptor for assessing the bioaccumulation potential. Information on bioaccumulation of such substances should therefore take account of other descriptors or mechanisms than hydrophobicity. Kow or Log Kow is a key input parameter in environmental modeling tools to estimate environmental exposure levels [10]. ECOSAR software contains a library of class-based QSARs for predicting aquatic toxicity, overlaid with an expert decision tree for selecting the appropriate chemical class based on chemical structure. The ECOSAR program was developed by Environmental Protection Agency 's Office of chemical safety and pollution prevention. It is a screening level tool intended for use in application such as rapid screening of chemical for Eco-toxicity hazards and prioritization of chemical (biocides) for future work, thus it will be used to predict the aquatic toxicity of resulting from using different types of non-oxidizing biocides (chlorophenol, trichlorophenol and quaternary-phenyl) and oxidizing biocides (sodium hypho-chlorite, chlorine dioxide and ozone) in suggested once through cooling water system at the selected Sidi Kirir region, in Egyptian coastal area[10].

2 Material and Methods

Sidi kirir was selected as cooling water site (Once through pass cooling water system) by the study survey. Physicochemical and biological analysis was measured in the selected site during six months (seasonal variations) at suggested nuclear power plant cooling water site, different parameters were measured as (Temp, pH, alkalinity, hardness, total solids, NH3,…etc.) by using (Standard method for examination for water and wastewater analysis)(11) , as the measuring pH and alkalinity parameters of the selected site, were suitable for using (Quantitative Structure Activity Relationships (QSARs) modeling software, Statistical correlations for different Kow will be done for the resulted lower environmental risk biocide which would be used in cooling water treatment system of the suggested nuclear power plant. The QSARs in ECOSAR for both neutral organics and excess toxicity are based on a linear mathematical relationship between the predicted log Kow values and the corresponding log of the measured toxicity values (mmol/L). ECOSAR study criteria articulate that the toxicity should be measured at pH 7-8 (replicating environmental conditions). Inorganic and organic chemicals as well as polymers and other substances with average MW>1000 are outside the domain. . ECOSAR version (1.11) [12], was programmed to identify 111 chemical classes and allows access to 704 QSARs for numerous endpoints and organisms. In an earlier version (1.11) of ECOSAR, the types of chemicals often designed with surfactant properties are detergents, wetting agents, and emulsifiers. Within ECOSAR, the surfactants are grouped by total charge into four general divisions: anionic (net negative charge), cationic (net positive charge), nonionic (neutral), and amphoteric (positive and negative localized charges) surfactants. The QSARs for surfactants can be linear or parabolic and the toxicity is often related to the size of the hydrophobic component or the number of repeating hydrophilic components [12].

Table1: Sidi-kirir Mean average cooling water physicochemical and biological properties during the period 12/3/2020-16/8/2020.

Parameter(mean average)	Range	Mean average
Temperature in ^{0}C	25-28	27
pH	7.6-8.8	---
Alkalinity as $CaCO_3$ mg/l	140-178	170
Total solids mg/l	38144-38800	38760
Total hardness mg/l	4156-5290	5089
Chlorides	20789-21680	21567
Nitrates	4.1-6.9	5.6
NH^{3+} mg/l	2.1-7.6	2.8
Phosphates mg/l	3.8-7.6	5.8
Total plankton count Cell/l	236633-236856	236789

3. Reults and Discussion

3.1 Non oxidizing biocides.

Table 2: Using ESAR model to calculate the risk of different living organisms resulted from using chlorophenol biocide in cooling water.

Biocide	Organism	Duration	End point	Predicted in mg/l (ppm)	Base line toxicity SAR
Chlorophenol Clc2ccccc2O	Fish	96-hr	LC50	16.957	76.284
	Daphnia	48-hr	LC50	5.476	24.804
	Green Algae	96-hr	EC50	24.884	34.763
Molecular weight	Fish		ChV	1.752	7.585
	Daphnia		ChV	1.041	4.463
128.56	Green algae		ChV	11.529	9.403
Chlorophenol log Kwo			Maximum Log Kow	Baseline Toxicity SAR Limitations	
2.157	Fish 96-hr , Daphnia		LC50 =7.0	5.0	
	Green Algae		EC50 =0.7	6.4	
	ChV =8.0			8.0	

Table 3: Using of ESAR model to calculate risk of different living organisms resulted from using trichlorophenol biocide in cooling water.

Biocide	Organism	Duration	End point	Predicted in mg/l (ppm)	Base line toxicity SAR
Tri-chlorophenol Clc1cc[c10]Cl]Cl	Fish	96-hr	LC50	2.728	8.150
	Daphnia	48-hr	LC50	1.564	5.288
	Green Algae	96-hr	EC50	6.252	6.843
	Fish		ChV	0.340	0.932
	Daphnia		ChV	0.297	0.748
Molecular weight 197.45	Green algae		ChV	2.884	2.413
chlorophenol log Kwo			Maximum Log Kow	Baseline Toxicity SAR Limitations	
3.446	Fish 96-hr , Daphnia		LC50 =7.0	5.0	
	Green Algae		EC50 =0.7	6.4	
	ChV =8.0			8.0	

Table(1) showed the physicochemical and biological mean average values of the suggested once through pass cooling water(Sidi Kirir) as temperature, hardness, total alkalinity, nitrates, chlorides, and phytoplankton count will lead to the formation of bio-fouling and scale formation inside the selected cooling water site, also the measured pH values are ranged from 7.6-8.8 this values may affect the condenser tubes in the suggested selected site if there is no cooling water treatment as it was found that At pH 8.5, the corrosion rate of iron in untreated water reaches a minimum[7,13]. It was noticed that the significant effects of the pH of cooling water are occurred as follows: 1) Above pH 7.0 pitting occurs with polyphosphate, 2) Above pH 8.0 the chlorine become ineffective as biocide. On the other hand the pH ranged from 7.8-8.8 is suitable for using modeling software. ESAR version 1.11(Quantitative Structure Activity Relationships (QSARs) modeling software, The ECOSAR program was developed by Environmental Protection Agency's Office of chemical safety and pollution prevention[13].Table (2) showed that Log Kow of chlorophenol biocide (2.157) was lower than the endpoint of fish, Daphnid, green algae Log Kow so there will be some effects at saturation expected at each end point predicted. On the other hand Maximum log Kow of LC50 of fish and Daphnia96-hr 7.0 was higher than baseline toxicity SAR limitation and the EC50 for green algae 0.7 was also higher than baseline toxicity SAR limitation. Table(2) had showed also that LC50 end point predicted of Fish at 96-hr duration 16.957 mg/l was lower than base line toxicity SAR, also LC50 end point predicted of Daphnia at 48-hr duration 5.476 mg/l was lower than Base line toxicity SAR, and EC50 of end point predicted of Green algae at 96-hr duration 24.884 mg/l was lower than Base line toxicity SAR, while both predicted ChV end point of Fish, Daphnid,(1.75,1.041) was lower than base line toxicity while ChV of green algae was higher than Base line toxicity SAR limitations. Table (3) showed that Log Kow of chlorophenol biocide (3.446) is lower than the endpoint of fish, Daphnia, green algae Log Kow so there will be some effects at saturation expected at each end point predicted. On the other hand Maximum log Kow of LC50 of fish and Daphnia 96-hr 7.0 was higher than baseline toxicity SAR limitation and EC50 for green algae 0.7 was higher baseline toxicity SAR limitation. Table(3) had showed also that LC50 end point predicted of Fish at 96-hr duration 2.728 mg/l was lower than base line toxicity SAR, also LC50 end point predicted of Daphnia at 48-hr duration 1.564 mg/l was lower than Base line toxicity SAR, and EC50 of end point predicted of Green algae 96-hr duration 6.252 mg/l was lower than Base line toxicity SAR, while both predicted ChV end point of Fish, Daphnia,(0.340,0.297) was lower than base line toxicity while ChV of green algae(2.884) was higher than ChV Base line toxicity SAR limitations in case of algae(2.413).Table (4) showed that Log Kow of quaternary phenyl biocide (7.285) was higher than the endpoint log Kow of fish, Daphnia, and lower than green algae log Kow so there will be no effects at saturation expected at each end point predicted, this was observed also in decreasing the end point LC50 predicted of Fish at 96-hr duration base line toxicity SAR, also LC50 end point predicted of Daphnia at 48-hr duration Base line toxicity SAR, and EC50 of end point predicted of Green algae 96-hr duration Base line toxicity SAR. It was also observed that end point of EC50 green algae at 96 hr duration exceeds the water solubility by 10x , so there will be some effects in saturation in case of green algae duration. Fig (1) showed that lowest LC50 in Fish and Daphnia found in Quaternary phenyl biocide with values 0.005 and 0.004 mg/l respectively while the highest LC50 value of chlorophenol in fish and Daphnia were 16.95 and 5.47 mg/l respectively while in case of EC50 fig(1) showed that the lowest EC50 value in green algae was found when using quaternary phenyl biocide with 0.025 mg/l while the highest EC50 value of green algae found when using chlorophenol biocide with value 24.884 mg/l this support the using quaternary phenyl as low ecological risk biocide will be used in fouling treatment in the cooling water system at two selected site Sidi Kirir

Table 4: using ESAR model to calculate risk of different living-organisms parameter resulted from using Quarter phenyl biocide in cooling water.

Biocide	Organism	Duration	End point	Predicted in mg/l (ppm)
Sodium Hypochlorite	Fish	96-hr	LC50	16271.958
	Daphnia	48-hr	LC50	7088.866
	Green Algae	96-hr	EC50	1765.754
NaOCl	Fish		ChV	1163.636
	Daphnia		ChV	330
Molecular Wt 52.46	Green algae		ChV	256.326
Hypochlorite log Kwo			Maximum Log Kow	Baseline Toxicity SAR Limitations

3.2 Oxidizing biocides

Table 5: Using ESAR model to calculate Risk of different living organisms resulted from using sodium hypochlorite biocide in cooling water.

Biocide	Organism	Duration	End point	Predicted in mg/l (ppm)
Sodium Hypochlorite	Fish	96-hr	LC50	16271.958
	Daphnia	48-hr	LC50	7088.866
NaOCl	Green Algae	96-hr	EC50	1765.754
	Fish		ChV	1163.636
Molecular Wt 52.46	Daphnia		ChV	330
	Green algae		ChV	256.326
Hypoch			Maximum	Baseline

lorite log Kwo			Log Kow	Toxicity SAR Limitations

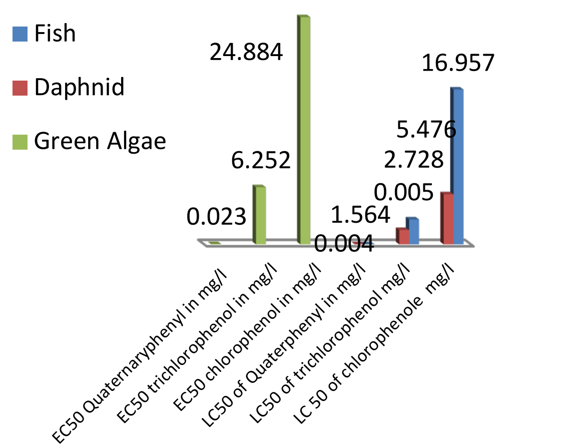

Fig 1: Determination of nonoxidizing biocides LC50 and EC50 fish, Daphnia and green Algae.

Table (5) showed that Log Kow of sodium hypochlorite biocide (-0.869) is lower than the endpoint Kow of fish, Daphnid, green algae Log Kow so there will be some effects at saturation expected at each end point predicted, this was observed also in increasing the end point LC50 predicted of Fish at 96-hr duration base line toxicity SAR, also LC50 end point predicted of Daphnia at 48-hr duration Base line toxicity SAR, and EC50 of end point predicted of Green algae at 96-hr duration Base line toxicity SAR. Table (4) showed that all the predicted level end point were not exceeding the water solubility 1E+006 by 10X, so that there will be some effects at saturation. Table (6) showed that Log Kow of chlorine dioxide biocide value (-3.221) is lower than the endpoint Kow of fish, Daphnia, and the green algae so that there will be some effects at saturation expected at each end point predicted, this was observed also in increasing the end point LC50 predicted of Fish at 96-hr duration base line toxicity SAR, also LC50 end point predicted of Daphnia at 48-hr duration Base line toxicity SAR, and EC50 of end point predicted of Green algae 96-hr duration Base line toxicity SAR. Table (6) showed that all the predicted levels end point were not exceeding the water solubility 1E+006 by 10X, so that there will be some effects at saturations . Table (7) showed that Log Kow of ozone biocide (-0.871) was lower than the endpoint Kow of fish, Daphnia, and green algae so that there will be some effects at saturation expected at each end point predicted, this was observed also in increasing the end point LC50 which was predicted of Fish at 96-hr duration base line toxicity SAR, also LC50 end point was predicted of Daphnia at 48-hr duration Base line toxicity SAR, and EC50 of end point was predicted of Green algae 96-hr duration Base line toxicity SAR. Table (7) showed also, that all the predicted levels end point were not exceeding the water solubility 1E+006 by 10X, so that there will be some effects at saturations and Fig (2) showed that lowest LC50 in Fish and Daphnia were found in ozone biocide with values 14937.231 and 6506.432 mg/l respectively while the highest LC50 value of chlorine dioxide in fish and Daphnia were 2.71e+06 and 9.49e+05 mg/l respectively while in case of EC50 fig(2) showed that the lowest EC50 value in green algae was found when using ozone biocide with 1619.687mg/l while the highest EC50 value of green algae found when using chlorine dioxide biocide with value 96353.992 mg/l this supporting the using of ozone as low ecological risk ionizing biocide as will be used in fouling treatment in the cooling water system at the selected site (Sidi Kirir) but in case of the high cost of using ozone specially in high flow rate of once through cooling water system as the result the using of sodium hypochlorite as second lowest LC50 of fish with 16271.958 mg/l and of Daphnia with 7088.85 mg/l on the other hand with second lowest green algae EC50 with value 1765.754 mg/l.

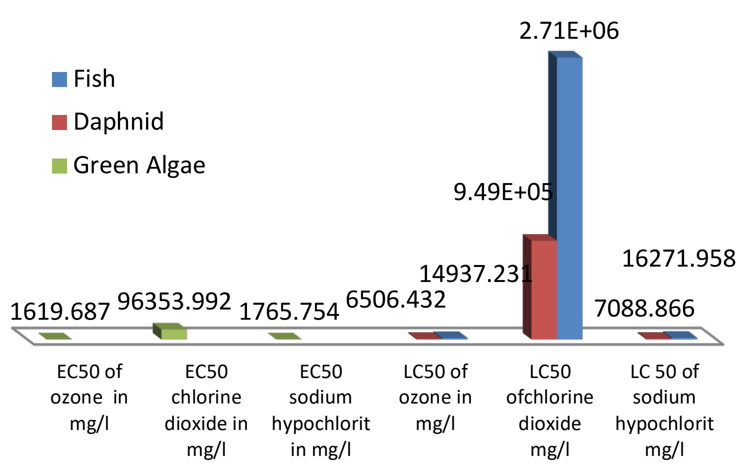

Fig2: Determination of oxidizing biocides LC50 and EC50 for fish Daphnia and green Algae.

Table 6: using ESAR model to calculate risk of different living organisms resulted from using chlorine dioxide biocide in cooling water.

Biocide	Organism	Duration	End point	Predicted in mg/l (ppm)
Chlorine Dioxide O=Cl=O Molecular Weight 67.45	Fish	96-hr	LC50	2.71e+006
	Daphnia	48-hr	LC50	9.49e+005
	Green Algae	96-hr	EC50	96353.992
	Fish		ChV	1.5e+005
	Daphnia		ChV	24201.813
	Green algae		ChV	8624.512
Chlorine dioxide log Kwo			Maximum logKow	Baseline Toxicity SAR Limitations
-3.221	Fish 96-hr , Daphnia		LC50=5.0	5.0
	Green Algae		EC50 =5.0	6.4
	ChV =8.0			8.0

Table 7: using ESAR model to calculate risk of different living organisms resulted from using ozone biocide in cooling water.

Biocide	Organism	Duration	End point	Predicted in mg/l (ppm)
Chlorine Dioxide O=Cl=O Molecular Wt 67.45	Fish	96-hr	LC50	2.71e+006
	Daphnia	48-hr	LC50	9.49e+005
	Green Algae	96-hr	EC50	96353.992
	Fish		ChV	1.5e+005
	Daphnia		ChV	24201.813
	Green algae		ChV	8624.512
Chlorine dioxide log Kwo			Maximum logKow	Baseline Toxicity SAR Limitations
-3.221	Fish 96-hr , Daphnia		LC50= 5.0	5.0
	Green Algae		EC50 =5.0	6.4
	ChV =8.0			8.0

4 Conclusions and Recommendation

The measured physicochemical and biological mean average values of the suggested once through pass cooling water(Sidi Kirir) as temperature, hardness, total alkalinity, nitrates, chlorides, and phytoplankton count will lead to the formation of bio-fouling and scale formation inside the condenser tubes of the selected cooling water site. Using biocides is added to the incoming seawater, in order to prevent bio-fouling in cooling water system, the complexity of a biocide reaction with different species that exist naturally in seawater is that it produces different by-products. It is necessary to identify the most effective by-product in controlling biofilm development.

Results showed that the lowest LC50 in Fish and Daphnia found in Quaternary ammonium salts biocide, while the highest LC50 value was showed in chlorophenol values in fish and Daphnia. On the Other hand the lowest EC50 value in green algae was found when using quaternary phenyl biocide. This support the using quaternary phenyl as low ecological risk biocide will be used in fouling treatment in the cooling water system at the selected site Sidi Kirir when using nonoxidixing biocides. In case of using oxidizing biocides, the results showed that lowest LC50 in Fish and Daphnia found when using ozone biocide while the highest LC50 value obtained in fish and Daphnia when using chlorine dioxide, while in case of EC50 results showed that the lowest EC50 value in green algae was found when using ozone biocide while the highest EC50 value of green algae found when using chlorine dioxide biocide this support the using ozone as low ecological risk ecological biocide in the treatment of the cooling water system at the selected site Sidi Kirir . Due to the high cost of using ozone(oxidizing biocide), as in high flow rate of once through cooling water system, So that the using quaternary ammonium salts (non-oxidizing biocides) will be best recommended economical and lowest ecological antifouling biocides, for the selected nuclear power plant cooling water system in Sidi Kirir.

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