重金属和有机磷农药对真鲷和平鲷幼体的联合毒性研究*

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摘要 以真鲷和平鲷幼体为实验材料, 采用联合指数相加法, 研究了重金属和农药对真鲷和平鲷幼体的急性毒性和联合毒性效应. 实验结果表明: 铜、锰、甲胺磷和甲基异柳磷对真鲷和平鲷幼体的毒性顺序是: 甲基异柳磷> 锰> 铜> 甲胺磷. 甲胺磷和Mn对真鲷和平鲷幼体毒性相同; Cu对2种鱼的毒性几乎相同; 而甲基异柳磷对真鲷的96h LC_{50}为0.02mg/L, 对平鲷幼体的96h LC_{50}为0.0014mg/L, 前者是后者的14倍. 对真鲷, 甲基异柳磷的毒性比甲胺磷毒性大175倍, 而对平鲷幼体, 甲基异柳磷的毒性比甲胺磷毒性竟大2500倍. 联合毒性实验结果表明: 铜-锰、铜-甲基异柳磷对真鲷和平鲷幼体的联合毒性为拮抗作用, 甲基异柳磷-甲胺磷的联合毒性为协同作用.

关键词 重金属, 有机磷农药, 联合毒性作用, 真鲷, 平鲷.

关于重金属和农药污染对海水养殖造成影响的研究工作, 国内开展了很多研究[1-4], 但多数是研究单一因子对海洋生物的毒性效应, 而在海水中实际存在着多种污染物, 它们的作用无疑是综合的. 用重金属和农药的单一毒性实验结果, 往往不能客观地反应污染物共存时对海洋生物的危害程度, 必须研究毒物的联合毒性效应.

已有研究表明环境因子及重金属对鲷科鱼类早期幼体存活率有很大影响[5]. 但有关重金属和农药对鲷科鱼类幼体的联合毒性研究尚未见报道. 本文研究了重金属和农药对真鲷(*Pagrosomus major*)和平鲷(*Rhabdosargus sarba*)幼体的急性毒性和联合毒性效应, 为混合污染物的联合毒性作用的研究提供一些资料, 并为渔业环境监测, 污染物排放及人工育苗水质管理提供参考.

1 材料和方法

1.1 实验材料

联合毒性实验用鱼(真鲷和平鲷)由厦门水产学院养殖系养殖场提供, 真鲷体长范围: 2.5–3.2cm, 平均体重0.42g, 平鲷体长范围: 2.8–3.5cm, 平均体重0.60g, 将鱼苗从养殖场运至实验室, 暂养3d后进行实验.

1.2 试剂

$CuSO_4 \cdot 5H_2O$(A.R.), K_2MnO_4(A.R.), 用蒸馏水分别配成含Cu^{2+}为2mg/ml, 含Mn^{2+} 1.59mg/ml的母液; 将市售40%(乳剂)甲胺磷和10%(乳剂)甲基异柳磷分别稀释成10000mg/L和10mg/L(根据农药瓶上所标示的有效成分计算)的母液.

1.3 实验步骤和方法

(1) 测定单一毒物对真鲷幼体和平鲷幼体的毒性 预备实验, 以3倍之差估计3个浓度, 每一浓度用5条鱼苗, 观察24h, 找出铜、锰、甲胺磷和甲基异柳磷的100%致死浓度和最大耐受浓度.

根据预备实验结果按等比级数设5个浓度组, 一个对照组, 同时设有一个平行组, 每一浓度放鱼10尾, 随时捞出死亡个体, 记录各组鱼苗24h、48h、96h死亡数, 实验容器用体积为6L的聚乙烯塑料桶, 内装实验液4L供实验用, 实验期间全天充气, 实验用海水采用经沉淀、砂滤的海水, 盐度26—28, 水温度25—27℃, pH为8.04—8.01, 为防止饵料影响, 实验期间不喂食.

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以浓度的常用对数为横坐标, 死亡率的概率单位为纵坐标, 求出各自的半数致死浓度(LC_{50}).

(2) 联合毒性实验 在单一毒性的基础上, 按毒性单位1∶1进行5个浓度组的联合毒性实验, 其中真鲷幼体的毒性实验配比是: 铜和锰是6.4∶1, 甲胺磷和甲基异柳磷是50∶1, 铜和甲基异柳磷是25∶1, 平鲷幼体毒性实验的配比是: 铜和锰的浓度比为6.4∶1, 甲胺磷和甲基异柳磷的浓度比为200∶1, 铜与甲基异柳磷浓度比为133∶1, 各试验组均设平行组, 分别记录各组24h、48h、96h 的死亡个体数.

(3) 联合毒性的评价方法 实验得到单一毒性和联合毒性 LC_{50}后, 用水生毒理联合效应相加指数法求 S[6]

$$S = \frac{Am}{An} + \frac{Bm}{Bn} + \frac{Cm}{Cn}$$

式中, S 为对海洋生物毒性相加之和, A、B、C 分别为实验毒物, m、n 分别为单一毒性 LC_{50}值及混和毒物 LC_{50}值. 若 $S \leq 1$时, 相加指数 $AI = 1/S - 1.0$; $S > 1$时, $AI = S(-1) + 1.0$, 最后用相加指数 (Aditive Index) 法评价毒物的联合效应, $AI > 0$时为协同作用; $AI < 0$时为拮抗作用; $AI = 0$时, 为相加作用.

2 实验结果与讨论

2.1 Cu、Mn、甲胺磷和甲基异柳磷对真鲷和平鲷幼体的单一毒性作用.

实验测得Cu、Mn、甲胺磷和甲基异柳磷对真鲷和平鲷幼体的96h 半数致死浓度(表1). 从表1可见4种污染物对真鲷和平鲷幼体的毒性顺序是: 甲基异柳磷> Mn> Cu> 甲胺磷, 4种污染物中锰对真鲷和平鲷幼体的毒性作用相同, 甲胺磷亦是如此, 铜对真鲷和平鲷幼体的96h 的半数致死浓度分别为1.65、1.78mg/L, 毒性也几乎相同, 而甲基异柳磷对真鲷和平鲷幼体96h 半数致死浓度分别为0.02和0.0014mg/L, 二者相差14倍, 对真鲷, 不同污染物其毒性也有很大差别, 如甲基异柳磷的毒性是甲胺磷的175倍, 对平鲷, 甲基异柳磷的毒性是甲胺磷的2500倍.

表1 污染物对真鲷和平鲷幼体的半数致死浓度

实验鱼类	96h LC_{50}/ mg·L^{-1}			
	Cu	Mn	甲胺磷	甲基异柳磷
真鲷	1.65(1.4-1.9)	0.26(0.19-0.33)	3.5(3.36-3.63)	0.02(0.01-0.03)
平鲷	1.78(1.55-2.01)	0.26(0.19-0.33)	3.5(3.36-3.63)	0.014(0.0008-0.002)

铜、锰是生命活动所必需的微量元素, 构成酶的活性基团, 或是酶的组成成分, 但当其浓度超过生物的生态阈值时, 会引起生物中毒. 铜和锰都是水环境优先污染物. 本实验结果表明, 高价锰的毒性比铜离子强, 据文献[7]报道它们的毒作用机理是: 高价锰可显著减少琥珀酸脱氢酶(SDH) 和细胞色素氧化酶(CCD) 的活性, Cu^{2+} 可使肝溶酶体膜磷脂发生氧化反应, 导致溶酶体膜的破裂, 水解酶大量释放, 从而引起肝组织坏死.

本实验结果表明, 甲基异柳磷比甲胺磷对真鲷幼体的毒性大175倍, 对平鲷幼体毒性竟大2500倍. 甲胺磷和甲基异柳磷都是胆碱酯酶(ChE) 的强烈抑制剂, 但甲胺磷对 ChE 的抑制作用比较慢, Eto 等人认为甲胺磷在活体内抑制 ChE 的过程首先是 $CH_3S—P$ 键中硫原子氧化成亚砜, 然后通过这一具有活性中间产物抑制机体的 ChE[8]. 因此, 它属于间接 ChE 抑制剂, 并且蓄积作用也不强, 所以对水生生物来说相对毒性较低, 96h 半数致死浓度达3.5mg/L. 我国目前对农药毒性分级标准, 农药对鱼类的96h LC_{50}的值大于10mg/L 时, 为低毒农药, 介于1—10mg/L 时为中毒农药, 小于1mg/L 时为高毒农药, 根据这一标准, 本实验结果说明甲胺磷属中毒农药 , 甲基异柳磷属于高毒农药, 但根据陈碧鹃[9]等报道: 甲基异柳磷对紫贻贝的96h LC_{50}值为2.4mg/L, 本实验结果甲基异柳磷对真鲷和平鲷幼体的96h LC_{50}分别为0.02、

0.0014mg/L, 根据农药评价标准将得出同一种农药属不同毒性等级的结论, 这主要是因为生物耐污能力不同, 通过比较可以看出紫贻贝耐受能力最强, 真鲷和平鲷幼体的敏感性都高于紫贻贝, 其中贝类耐受甲基异柳磷的能力比真鲷高120倍, 比平鲷幼体高达1700倍, 这些结果可为安全使用农药或进行农药的环境安全评价提供一个毒性定量关系的参考依据, 同时也说明评价农药毒性时应选择敏感生物作为指标. 甲基异柳磷对真鲷和平鲷幼体毒性大小的差异还可能是对不同鱼类的乙酰胆碱酯酶的反应不同, 或是不同鱼类乙酰胆碱酯酶结构的差异所造成. 另外农药本身的差异如甲基异柳磷难溶于水, 而甲胺磷溶于水, 甲胺磷属间接毒性, 也是主要原因之一.

2.2 联合毒性作用

在单一毒性实验的基础上, 按毒性单位1∶1进行联合毒性实验, 测得铜-锰、铜-甲基异柳磷和甲胺磷-甲基异柳磷对真鲷和平鲷幼体的联合作用的96h 半数致死浓度(表2—4).

表2 **Cu** 和 **Mn** 对真鲷和平鲷幼体的联合毒性

实验鱼类	96h LC_{50}/mg·L^{-1}		*AI*	作用
	Cu	Mn		
真鲷	2.2	0.3	-1.48	拮抗
平鲷	2.1	0.3	-1.33	拮抗

表3 **Cu** 和甲基异柳磷对真鲷和平鲷幼体的联合毒性

实验鱼类	96h LC_{50}/mg·L^{-1}		*AI*	作用
	Cu	甲基异柳磷		
真鲷	1.346	0.054	-2.51	拮抗
平鲷	1.198	0.002	-1.09	拮抗

表4 甲胺磷和甲基异柳磷对真鲷和平鲷幼体的联合毒性

实验鱼类	96h LC_{50}/mg·L^{-1}		*AI*	作用
	甲胺磷	甲基异柳磷		
真鲷	1.8	0.009	0.04	协同
平鲷	1.75	0.006	0.09	协同

从表2中可看出, 铜、锰对真鲷和平鲷幼体的联合毒性实验所得的相加指数 *AI* 分别为-1.48、-1.33都小于0, 其结果为拮抗作用, 即当二者同时存在时毒性减弱, 铜和锰之间的拮抗关系的发生可能是铜竞争与锰结合部位, 从而铜抑制了锰的吸收, 这与王安利等[10]研究的铜、锰对中国对虾仔虾的联合毒性也是拮抗关系这一结论相同.

从表3中可见铜和甲基异柳磷对真鲷和平鲷幼体的联合毒性所得的相加指数分别为-2.51、-1.09, 均小于0, 也属拮抗作用, 表4列出了甲胺磷和甲基异柳磷联合毒性所得的相加指数均大于0, 联合作用为协同作用.

甲胺磷和甲基异柳磷96h 联合毒性实验结果为协同作用, 二者联合作用具有增毒、提高药效的作用. 从有机磷农药对真鲷和平鲷幼体单一毒性和联合毒性 LC_{50}值比较来看, 不同的有机磷农药有的对不同生物毒性相同, 有的毒性差别很大. 主要原因既同农药结构不同有关, 也同鱼类本身生理功能(主要是酶结构) 差异有关, 而农药联合作用的毒性是各农药之间相互作用的结果, 毒性大小主要同农药单剂的结构、毒性 LC_{50}值和有效浓度及农药的选择性有关. 因此正确地预测农药联合毒性效应, 还必需深入分析各农药的结构与毒性大小的定量结构关系(QSAR), 以及毒作用机制.

铜和甲基异柳磷的作用是拮抗关系, Stromgren 等报道: 石油和铜的混和污染对贻贝壳的生长和摄食影响的关系是拮抗关系, 而不是简单的叠加关系相类似, 但目前对这种拮抗机制还没有做出合理的解释. 笔者认为本实验很可能是有机磷农药对神经系统的麻痹作用减少了对铜的吸收, 从而减少了铜的毒性.

当然联合作用方式可随观察指标的不同而有差别, 实验结果也不宜任意外推现场条件, 否则会得出错误的结论.

3 有机磷农药联合作用的危险评价

当多种农药共存时, 尽管其毒性不同, 但对生物的作用方式和性质是相似的, 其联合毒性可以被一定量的其它农药所代替. 而农药的有

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因而用脱氮硫杆菌处理垃圾填埋场硝化渗滤污水时需要消耗碱度.

从图10中还以看出,产气速率存在一个最高值,表明此时 NO_3^--N 去除速率达到最大.

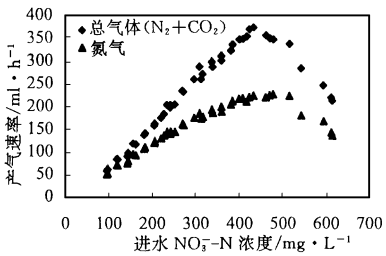

图10 进水硝酸盐浓度与产气速率的关系

3 结论

(1) 脱氮硫杆菌处理垃圾填埋场硝化渗滤污水是可行的. 当HRT 为5.71h时, NO_3^--N 处理浓度高达400mg/L,处理效率接近100%.

(2) 达到完全反硝化所需的最小 HRT 取决于硫磺粒径及进水硝酸盐浓度.

(3) 在 HRT 恒定的条件下,达到完全反硝化所能处理的最高硝酸盐浓度与硫磺粒径有关.

(4) 最大硝酸盐体积负荷取决于硫磺粒径,但最大表面负荷是自养反硝化过程的控制因素,与硫磺粒径无关.

(5) 自养反硝化过程除产生大量氮气外,还有一定量的 CO_2.

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效浓度以相同的毒性单位表示时,那么,联合作用的有效浓度为单一农药的有效浓度之和,即

$$c = \sum_{i=1}^{n} c_i$$

如果将联合作用的有效浓度 c 和联合毒性 LC_{50} 值的比定义为危险度(W),

$$W = \frac{c}{LC_{50}}$$

当 $W = 1$时,相当于海域中的农药的总有效浓度 LC_{50} 值. 西内康浩(1986)[11] 曾把危险度分为3级,当 $W \leq 0.1$时为低毒; $0.1 < W < 5.0$为中毒毒性; $W \geq 5.0$时为剧毒. 此危险评价在海域农药污染的联合毒性的评价时,具有实际应用价值.

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troaromatic compounds in the ecosystem, the study on the mixtoxicity of 2, 4-DNT and other 6 kinds of nitroaromatics to the algae (*Scenedesmus obliquus*) was carried out. 48h-EC_{50} of monotoxicity of 7 kinds of compunds and 48h-EC_{50} of mixtoxicity of 2, 4-DNT + 6 kinds of compounds were measured. Results indicated that there are synergisms between, 2, 4-DNT + 4-NAn, 2, 4-DNT + 4-NAnis, and 2, 4-DNT + 1, 4-DNB as well as antagonisms between 2, 4-DNT + 4-NT, 2, 4-DNT + 4-NPh and 2, 4-DNT + 4-NCB. Under the microscope (400 times) 3 kinds of toxic symptoms, the inhibition to the filial spores forming and releasing, the irregular big cells appearing, nucleus and cell organs as well as the protoplast distegrating were observed.

Key words: mixtoxicity, 2, 4-DNT, nitroaromatic compounds, algae, *Scenedesmus obliquus*.

Study on the Volatile Property of Organics in Coke-Plant Wastewater under the Aerated Stripping Condition. He Miao, Zhang Xiaojian etal. (Dept. of Environ. Eng., Tsinghua University, Beijing 100084): *Chin. J. Environ. Sci.*, **18**(5), 1997, pp. 34—36

A study was conducted to determine the volatile property of organics in the coke-plant wastewater under the condition of aerated stripping. The results show that there are different volatile characteristics among the organics. 11 kinds of compounds such as ethylbenzene and naphthalene are more volatile. The removal rate can get to 20% - 40% after an aeration of 12 hours. Their volatile rate constant K_v have a fairly good linear relation with the Henry constant (H), of which relation equation is $K_v = 2.906 \times 10^3 H + 0.0146$. Volatile effect plays a very important role in the removal process of these organic compounds. 12 compounds like phenol have a medium volatile property. The other compounds like resorcinol have a low volatility.

Key words: volatile property, coke-plant wastewater, aerated stripping, volatile organic compound, volatile rate constant.

Activity, Kinetics and Spatial Variation of Dissolved Alkaline Phosphatase in Lake Donghu. Zhou Yiyong, Li Jianqiu, Chen Xudong et al. (Institute of Hydrobiology, The Chinese Academy of Sciences, Wuhan 430072): *Chin. J. Environ. Sci.*, **18**(5), 1997, pp. 37—40

The extra cellular enzyme activities in aquatic environments are of monitoring significance. Dissolved alkaline phosphatase activities (APA_{Diss}) at the littoral outfalls, which receive the domestic waste water were significantly higher than those in pelagic zone in Donghu Lake. APA_{Diss} detected at the outfalls and in pelagic zone showed different relationships with soluble reactive phosphorus.

Key words: dissolved phosphatase, kinetics, characteristics, spatial distribution.

A Study on Bioaccumulation and Biomagnification of BHC and DDT in Baiyangdian Lake Foodweb. Dou Wei, Zhao Zhongxian (Institute of Zoology, Chinese Academy of Sciences, Beijing 100080): *Chin. J. Environ. Sci.*, **18**(5), 1997, pp. 41—43

BHC、DDT and their metabolites were analysed in Duancun area from Baiyangdian Lake ecosystem in June 1995. The content levels of pesticides in water and bottom sediments were relatively lower: in water, the average values of BHC and DDT were 0.3μg/L and 0.1μg/L, respectively; in sediments, the residue level of BHC was same with DDT with a mean concentration of 0.7μg/kg(wet). The organochlorine insecticides content in aquatic organisms was much higher: the average concentration of BHC in hydrophytes, planktons, benthic macroinvertebrates, young-of-the-year *Carassius auratus and* 2-year-old *Ophiocephalus cris-pus* was 19.0, 30.0, 60.9, 17.2 and 110.7 μg/kg, respectively; for DDT, the content sequence was 6.3, 21.0, 37.9, 19.4 and 124.4g/kg, respectively. So the lipophilic nature and low biodegradation rates of BHC and DDT led to the accumulation of these compounds and subsequent magnification of concentration in organisms progressing up the food chain. DDT had a much profound bioaccumulation potential than BHC for the bioconcentration factor of DDT in organisms (63.3- 1244) was well above that of BHC (63- 369). The ratios of BHC isomers monitored in organisms ($\delta > \alpha > \gamma > \beta$) were different from that in water and sediments ($\alpha > \delta > \gamma > \beta$). The predominant DDT derivative was p, p′-DDE. Base upon the observed that the original DDT (P, P′-DDT) was indentified in some specimens, it has been concluded that there was a recent input of DDT to Baiyangdian Lake.

Key words: Baiyangdian Lake ecosystem, BHC, DDT, foodweb, the bioconcentration factor.

Joints Toxicities of Heavy Metals and Pesticides to *Pagrosomus major and Rhabdosargus sarba* Larvae. Dai Jiayin, Zheng Weiyun, Wang Shuhong (Environ. Sci. Res. Centre, Xiamen University, Xiamen 361005): *Chin. J. Environ. Sci.*, **18**(5), 1997, pp. 44—46

The additive index of coeffects was used to study the toxicities of Cu-Mn, Cu-isofenphos-methyl and methamidphos-isofenphos-methyl

on *Pagrosomus major and Rhabdosargus sarba*. The acute toxicities of the four pollutants were in the sequence of isofenphos-methyl> Mn> Cu> methamidphos. Mn and methamidphos were equivalently toxic to *P. major and R. sarba*, Cu was almost equivalently toxic to them, while the 96h median lethal concentrations of isofenphos-methyl were 0.02mg/L for*P. major*, and 0.0014mg/L for*R. sarba*, respectively. The toxicity of isofenphos-methyl for *P. major* was 13 times higher than that for *R. sarba*. The toxicity of isofenphos-methyl were 174 times higher than that of methamidphos for *P. major*. The toxicity of isofenphos-methyl was 2500 times that of methamidphos for *R. sarba*. The additive toxicity of Cu-Mn and Cu-isofenphos-methyl for two species fishes was shown to be antagonistic, while that of methamidphos-isofenphos-methyl was synergism.

Key words: *P. major*, *R. sarba*, Cu, Mn, methamidphos, isofenphos-methyl, joint toxicity.

Microbial Treatment Technology for the Electroplating Wastewater. Wu Qianjing et al. (Chengdu Institute of Biology, Chinese Academy of Sciences, Chengdu 610041): *Chin. J. Environ. Sci.*, **18**(5), 1997, pp. 47—50

This paper has reported the new technology of electroplating wastewater and sludge treated by microbies on the basis of fundamental research, experiment and pilot scale. The demonstration project built on Chengdu Jinjiang Electronic Machine Factory has got a good result. Since then, four processes whose treating capacity ranged from 1 to 175 tons/d in Chengdu Hong Guang Industry Limited Company, Chinese People Liberation Army 5701 Factory etc. were established. The processes have run steadily, safely and reliably for two years. The level of chromium, zinc, copper, nickle, cadmium, lead, COD, BOD, SS, pH, color degree and NH_3-N in effluent is below the national GB8978-88 discharge standard. The water of effluent can be reused, the recovery of heavy metal of sludge can reach above 85%.

Key words: microbe, electroplating wastewater, heavy metal, treatment technology, sludge, reused, recovery.

Denitrification of Landfill Leachate by *Thiobacillus denitrificans*. Koenig Albert (Dept. of Civil and Structural Eng., The Uni. of Hong Kong), Liu Ling hua (Water Quality Research Center, China Institute of Water Resources and Hydropower Research, Beijing 100044): *Chin. J. Environ. Sci.*, **18**(5), 1997, pp. 51—54

The feasibility of nitrate removal from nitrified landfill leachate in reactors with different sulfur particle size was studied. The results indicate that ① the reactor with sulfur particle size of 2.8- 5.6 mm can effectively remove nitrate from nitrified leachate up to a concentration of 400 mg/L NO_3^--N at a hydraulic retention time of 5.71h; ② the minimum hydraulic retention time necessary for complete denitrification depends on sulfur particle size and influent nitrate concentration; ③ the maximum volumetric loading rate of NO_3^--N depends on sulfur particle size and is approximately 594, 278 and 175.4g/(m^3•d) for sulfur particle size of 2.8- 5.6mm, 5.6- 11.2mm and 11.2- 16mm, respectively; ④ the maximum area loading rate of NO_3^--N, approximately 0.68g/(m^2•d), appears to be the process limiting factor and is practically independent of sulfur particle size.

Key words: autotrophic denitrification, biofilm, *Thiobacillus denitrificans*, sulfur, landfill leachate, nitrate removal.

A Study on Treatment of Chemithermomechanical Pulping Wastewater with the New Technology by Combining ICZs and Activated Sludge Method. Chen Min (Guangdong Uni. of Tech., Environ. & Resource Eng. Dept., Guangzhou, 510090), Songnien Lou and H C laude Lavallée (Université du Québec à Trois-Rivières, Québec, Canada, G9A 5H7): *Chin. J. Environ. Sci.*, **18**(5), 1997, pp. 55—58

It is investigated that the treatment of Chemithermomechanical Pulping (CTMP) wastewater with new technology by combining Zone of Biological Population selection (ICZs) and activated sludge method. The control of sludge bulking in activated sludge system of CTMP treatment with this technology is discussed. The three month experiments showed that the slowness of filamentous microorganism growth is achieved and the sludgebulking is controlled with ICZs pretreatment by controlling DO below 0.06 mg/L and pH 7- 7.6. The biological treatment system with combining ICZs and activated sludge method can obtain efficient treatment. Removal of BOD_5, COD and TSS is above 97%, 80% and 90% respectively. ICZs pretreatment can remove 49.0% - 62.5% BOD_5 and 18.5% - 21.9% COD with only 20 minutes of HRT.

Key words: activated sludge, chemithermomechanical pulping wastewater, sludge bulking control, filamentous microorganism, zone of biological population selection, BOD_5, COD, TSS.

Numerical Simulation and Prediction for