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# 我国东南地区饮用水源地多种农药的赋存特征及健康风险评估

何姝<sup>1,2</sup>, 董慧峪<sup>3</sup>, 任南琪<sup>1,2\*</sup>

(1. 哈尔滨工业大学环境学院, 哈尔滨 150090; 2. 哈尔滨工业大学(深圳)土木与环境工程学院, 城市水资源与水环境国家重点实验室, 深圳 518055; 3. 中国科学院生态环境研究中心饮用水科学与技术重点实验室, 北京 100085)

**摘要:** 为阐明我国东南地区典型饮用水源地农药类微污染物的污染特征及生态风险, 检测评估了某省 7 个水库的苯并咪唑类、酰胺类、三唑类和有机磷类等 19 类共 55 种常用农药的检出频率、检出浓度以及每种农药对于绿藻、水蚤和鱼类这 3 种不同营养级生物的风险商. 在分析的 55 种农药中, 多菌灵和乙草胺的检出频率为 100%, 12 种农药的检出频率在 80% 以上. 多菌灵的检出浓度最高 ( $77.7 \text{ ng}\cdot\text{L}^{-1}$ ), 其次是乙草胺 ( $51.6 \text{ ng}\cdot\text{L}^{-1}$ ). 风险评估结果显示, 大部分农药在目标区域都处于低风险状态. 对于 3 种生物来说, 乙草胺是绿藻的风险主导型农药, 而多菌灵是鱼类和水蚤的风险主导型农药.

**关键词:** 农药类微污染物; 水库; 检出浓度; 检出频率; 风险商

中图分类号: X52; X820.4 文献标识码: A 文章编号: 0250-3301(2023)01-0180-09 DOI: 10.13227/j.hjxk.202202008

## Occurrence and Health Risk Assessment of Multiple Pesticides in Drinking Water Sources of Southeast China

HE Shu<sup>1,2</sup>, DONG Hui-yu<sup>3</sup>, REN Nan-qi<sup>1,2\*</sup>

(1. School of Environment, Harbin Institute of Technology, Harbin 150090, China; 2. State Key Laboratory of Urban Water Resource and Environment, School of Civil and Environmental Engineering, Harbin Institute of Technology (Shenzhen), Shenzhen 518055, China; 3. Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China)

**Abstract:** To elucidate the pollution characteristics and ecological risks of pesticide micro-pollutants in typical drinking water sources in Southeast China, the detection frequency, detection concentration, and risk quotient of each pesticide for three different trophic levels of organisms (green algae, daphnia, and fish) were analyzed for a total of 55 commonly used pesticides in 19 categories, including benzimidazoles, amides, triazoles, and organophosphates, in seven reservoirs in Southeast China. Among the 55 pesticides analyzed, two pesticides (carbendazim and acetochlor) had a detection frequency of 100%, and 12 pesticides had a detection frequency of 80% or more. The highest detection concentration was found for carbendazim ( $77.7 \text{ ng}\cdot\text{L}^{-1}$ ), followed by that of acetochlor ( $51.6 \text{ ng}\cdot\text{L}^{-1}$ ). The results of the risk assessment showed that most of the pesticides were at low risk in the target areas. For the three organisms, acetochlor was the risk-dominant pesticide for green algae, whereas carbendazim was the risk-dominant pesticide for fish and daphnia.

**Key words:** pesticide micro-pollutants; reservoir; detection concentration; detection frequency; risk quotient

农药是在农业生产中防止植物和农作物遭受例如病原体、真菌、害虫和杂草危害的化学药品. 全球农药的生产和使用随着时间的推移逐渐增长<sup>[1]</sup>. 农药中含有一种或多种活性物质, 残留农药可以通过地表径流或农田排水进入地表水体, 或渗入土壤污染地下水<sup>[2-4]</sup>, 进而对整个生态系统结构、功能以及人类健康产生严重危害<sup>[5-9]</sup>.

农药在水生环境中还可能引起鱼类种群的生理行为变化<sup>[10]</sup>. 农药浓度的增加可能导致鱼类种群数量减少<sup>[11]</sup>. 此外, 源水和饮用水中农药的出现可能与白血病、脑癌、淋巴瘤、乳腺癌、前列腺癌、卵巢癌和睾丸癌有关<sup>[10]</sup>. 大部分农药可能会造成中度甚至重度的呼吸系统和神经系统损伤<sup>[12]</sup>. 目前, 尽管有学者研究出了相对安全的农药来替代高毒性农药, 但其水生环境污染问题还远远没有得到解决<sup>[13]</sup>. 水库水作为饮用水供给链的前端, 是保证居民用水安全的重要一环. 因此, 有必要调查农药在水

库水中的浓度水平和生态风险.

本研究以我国东南某省某地区 7 个水库为目标区域, 在每个水库设置 4~9 个采样点. 采用超高效液相色谱串联三重四极杆质谱仪对 55 种农药进行检测分析, 并系统评估该区域 55 种农药的生态风险, 在此基础上提出了针对该地区的饮用水环境风险管理建议.

### 1 材料与方法

#### 1.1 样品采集与保存

本研究于 2021 年 6 月采集东南某省 7 个水库样品, 分别编号为 A、B、C、D、E、F 和 G. 每个采样点采集 5 L 水样并装入预先清洗过的琥珀色玻璃

收稿日期: 2022-02-05; 修订日期: 2022-04-19

作者简介: 何姝(1980~), 女, 博士研究生, 高级工程师, 主要研究方向为水环境监测与评价, E-mail: heshu@yctestng.com

\* 通信作者, E-mail: riq@hit.edu.cn

瓶,4℃储存后运送入冷库保存,3d内进行过滤和固相萃取等前处理。

### 1.2 样品制备与分析

用于样品制备和分析的其他化学试剂均为HPLC级或分析级。水样通过固相萃取(SPE)进行富集,使用Oasis HLB固相萃取柱(Waters, Millford, MA, USA)。固相萃取前,水样通过1 μm玻璃纤维滤膜(GF/B, Whatman, Mainstone, England)过滤,然后用40% (体积分数) H<sub>2</sub>SO<sub>4</sub>酸化至pH 3.0,最后用0.5 g·L<sup>-1</sup> Na<sub>2</sub>EDTA螯合。固相萃取柱需要提前活化[5 mL 甲醇+5 mL 稀盐酸(1:23)+5 mL 超纯水],使用固相萃取柱富集每个水样(1.0 L)并用4 mL×4 甲醇洗脱,然后用高纯度氮气将样品吹脱至400 μL。进样前,用超纯水将样品复溶至1 mL。提取的水样用超高效液相色谱串联三重四极杆质谱仪(UPLC-MS/MS)(带电喷雾离子源)(Agilent 1290/6420 UPLC-MS/MS, Agilent, USA)进行分析;使用C18柱进行目标分析物的色谱分离,具体操作参数见表1。

表1 超高效液相色谱串联三重四极杆质谱仪(带电喷雾离子源)工作参数

Table 1 Operating parameters of the ultra-performance liquid chromatography-triple quadruple mass spectrometer (with electrospray ion source)

工作参数	设定值
离子源	ESI 源
电离模式	正离子模式
毛细管电压/kV	3.0
离子源温度/℃	150
锥形反吹气流/L·h <sup>-1</sup>	50
溶剂气体温度/℃	500
溶出气体流量/L·h <sup>-1</sup>	900
监控模式	MRM 模式

### 1.3 质量保证和质量控制

在确定的保留时间内,根据母离子和两个丰度最高的产物离子完成对目标农药的定性分析。在样品分析前,通过测量农药回收率、检测和量化极限以及回归系数来进行方法验证。磷酸三苯酯作为内标物在仪器分析前加入每个预处理的样品中,以监测仪器分析中的底物效应。配置不同浓度梯度的混合标准溶液(0.1~2 000 ng·L<sup>-1</sup>),用于绘制每个目标物的标准曲线,用内标法进行定量分析。在样品中加入0.1、0.5、1、10、50、100、500和1 000 ng·L<sup>-1</sup>的混合标准溶液,按照优化方法进行平行实验( $n=3$ ),计算目标物的回收率和相对标准偏差(RSD)。

55种目标农药的平均回收率在78.62%~123.62%之间,RSD在2.10%~21.30%之间。每种农药的LOD和LOQ分别被定义为信号/噪声(S/N)

为3和10时所对应的浓度。所建立的UPLC-MS/MS方法具有良好的相关系数( $r^2 > 0.99$ )。

### 1.4 风险评估

将每种农药的浓度作为实测环境浓度(MEC),利用无观察效应浓度值(NOEC)来计算预测无效应浓度(PNEC)。本研究参考Ogbeide等<sup>[14]</sup>和Papadakis等<sup>[15]</sup>的方法,优先采用NOEC值,当大部分目标农药的NOEC值不可得时,采用半致死浓度(LC<sub>50</sub>)来代替NOEC值。具体公式如下所示。其中,公式(2)中所有农药的NOEC值均由LC<sub>50</sub>代替。在对每种农药的分析中,考虑了绿藻、水蚤和鱼类的慢性毒性数据,并分别计算了不同的风险商(RQ)。为了考虑从单一物种毒性推断到生态系统毒性所产生的不确定性,通过将LC<sub>50</sub>值除以评估因子100计算PNEC值,以评估农药的慢性毒性<sup>[16]</sup>。LC<sub>50</sub>值通过美国环保署的模型(ECOSAR)计算得到<sup>[17]</sup>。当水中的农药浓度较低时,不存在急性毒性,但其慢性毒性和各种农药的相互作用不容忽视<sup>[18]</sup>。

$$RQ = MEC/PNEC \quad (1)$$

$$PNEC = NOEC/100 \quad (2)$$

式中,当 $0.01 \leq RQ < 0.10$ 时,该农药对目标生物的风险低;当 $0.10 \leq RQ < 1$ 时,为中风险;当 $RQ \geq 1$ 时,为高风险<sup>[19]</sup>。

## 2 结果与讨论

### 2.1 农药的检出频率和浓度水平

东南某省饮用水源地主要检出的农药及其浓度见表2。在55种农药中,均未出现含量超过欧盟立法为饮用水中的单个农药规定的100 ng·L<sup>-1</sup>限值。所有目标农药中,多菌灵的检出浓度最高,为77.7 ng·L<sup>-1</sup>。类似地,周怡彤等<sup>[20]</sup>在太湖流域西北部地表水的检测中也发现多菌灵的浓度最高[未检出(N.D.)~114.44 ng·L<sup>-1</sup>]。乙草胺、甲霜灵和异丙甲草胺的检出浓度次之,分别为51.6、37.6和37.9 ng·L<sup>-1</sup>。乙草胺在水生生态系统中具有较高的稳定性和持久性<sup>[21, 22]</sup>。之前有报道提出乙草胺可以诱发斑马鱼的生殖毒性、免疫毒性和内分泌毒性<sup>[23]</sup>,因此可能对目标区域的鱼类等水生生物具有潜在危害。甲霜灵在中性和弱酸性的水体环境中表现出较高的稳定性<sup>[24]</sup>,这也是其检测浓度相对较高的原因。异丙甲草胺不仅会干扰植物细胞分裂的功能,影响光合作用<sup>[25]</sup>,还会影响蝌蚪的隐藏行为<sup>[26]</sup>。据统计,东南某省2020年水稻种植面积为636.02×10<sup>3</sup> hm<sup>2</sup>,占全省总粮食作物耕种面积的64%<sup>[27]</sup>,而多菌灵、乙草胺、甲霜灵和异丙甲草胺作为广泛使用的杀菌剂和除草剂,在水稻种植及生长过程中使

表 2 中国东南地区 7 个饮用水源地农药污染物浓度范围与检出频率

Table 2 Concentration ranges and detection frequency of pesticide-pollutants in drinking water sources in southern China

分类	农药名称	浓度范围/ng·L <sup>-1</sup>							检出频率/%	
		水库 A	水库 B	水库 C	水库 D	水库 E	水库 F	水库 G		水库 H
苯并咪唑类	多菌灵	1.8~35.6	1.3~14.8	9.4~18.9	6.7~9.0	7.9~77.7	4.4~30.1	2.5~19.9	7.7~23.2	100
	涕必灵	0	0.1~0.3	0	0	0	0~0.2	2.5~19.9	0.1~4.9	34.21
	甲基硫菌灵	0	0	0	0	0	0~0.2	0	0	2.63
	咪唑胺	0	0	0	0	0	0	0~0.2	0	2.63
氨基甲酸酯类	异丙威	0~0.4	0~0.2	0~0.2	0~0.1	0~0.2	0~0.7	0~0.1	0	39.47
	仲丁威	0	0	0~1.5	0	0~0.8	0~0.4	0~0.1	0~0.1	26.32
	克百威	0	0~0.3	0	0	0	0~1.5	0.1~0.3	0.1~0.3	36.84
	抗蚜威	0	0	0	0	0	0	0	0~0.1	2.63
	丁硫克百威	0	0	0	0~0.5	0	0~0.2	0	0	10.53
	氯虫苯甲酰胺	1.2~2.5	0.4~0.8	0	0~0.2	0	0	0	0	28.94
酰胺类	丁草胺	8.9~12.2	8.9~17.8	2.7~12.3	0~5.5	0.5~5.5	1.2~4.5	2.2~10.4	1.1~10.5	97.37
	乙草胺	17.8~32.4	16.1~51.6	12.4~22.9	10.7~22.4	2.6~22.4	8.8~20.5	10.9~28.5	8.5~31.2	100
	丙草胺	19.3~25.2	9.0~15.6	2.4~8.5	0~3.7	1.0~4.6	0~0.5	0~4.7	0~2.1	73.68
	甲霜灵	0~0.6	0~2.5	0.7~2.1	0.8~1.2	0~2.2	0~0.7	0.1~0.4	0.4~37.6	86.84
	异丙甲草胺	0~0.3	1.2~20.6	6.6~28.9	2.5~2.8	1.1~23.1	0.1	1.2~4.0	2.6~37.9	94.74
	啶虫脒	0	0~1.4	0.5~3.9	0.9~1.6	0.2~2.2	0~17.4	0.4~0.9	0.1~1.3	81.58
三唑类	啶酰菌胺	0	0~2.1	1.0~3.2	0~1.5	0~4.1	0	0~1.7	0~0.4	57.89
	腈菌唑	0~1.6	0~0.8	1.1~6.4	0.2~0.4	0~7.0	0	0~0.2	0.1~0.9	60.53
	多效唑	0~0.1	0.2~1.3	0.5~3.9	0.6~0.7	0~29.5	0.1~0.7	0.6~1.3	0.4~1.2	89.47
	三唑醇	0~3.4	0~0.7	0.5~4.2	0~0.3	0~3.1	0	0~0.4	0.1~1.6	63.16
	戊唑醇	0~0.2	0.3~1.4	0~0.1	1.2~1.6	0.1~60.1	0.1~0.4	0.3~3.4	1.7~6.6	97.37
	已唑醇	0	0~0.8	0~0.1	0.1~0.2	0~0.8	0~0.1	1.5~2.7	0.5~2.7	57.89
	烯唑醇	0~0.6	0	0~0.3	0	0	0	0	0~1.0	15.79
	氟环唑	0	0~5.1	0	0	0~0.2	0	0	0~0.1	15.79
	丙环唑	0~4.8	0~0.1	0.2~0.5	0~0.1	0~3.8	0~0.2	1.2~2.4	0~1.9	71.05
	苯醚甲环唑	0~3.3	0.1~0.5	0.2~0.7	0.2~0.3	0.2~2.1	0~1.9	1.9~3.1	1.3~4.3	89.47
三嗪类	莠灭净	0	0	0.1~6.0	0~0.1	0~0.9	0~0.1	0.3~0.5	0.3~0.5	57.89
	吡蚜酮	0	0	0	0	0	0	0	0~0.2	2.63

续表 2

分类	农药名称	浓度范围/ng·L <sup>-1</sup>								检出频率/%		
		水库 A	水库 B	水库 C	水库 D	水库 E	水库 F	水库 G	水库 H			
有机磷类	乐果	0~0.8	0	0	0	0	0~0.9	0	0	13.16		
	水胺硫磷	0~12.4	0~0.4	0~12.4	0	0~2.4	0~0.2	4.3~6.7	0.8~5.3	55.26		
	二嗪磷	0	0	0.1~1.1	0	0	0~0.2	0	0	13.16		
	灭草松	0.7~1.0	0.6~1.1	5.5~22.3	0	0.2~1.5	0	0	0	50		
	草铵膦	0	0	0	0	0	0	0~0.5	0~0.2	7.89		
	辛硫磷	0	0	0	0	0	0	0	0	0		
	毒死蜱	0.5~0.8	0.8~2.5	2.7~12.5	0	0.5~3.7	0.5~1.6	0~1.2	0~0.5	71.05		
	草甘膦	3.5~10.6	3.3~10.2	0~1.7	0	0.3~2.9	0~0.5	0	0	55.26		
	苯胺基嘧啶类	噻霉胺	0~0.1	0.1~0.9	0.2~1.8	1.1~1.2	0~1.0	0	0.3~0.5	0.3~0.5	76.32	
		吡虫啉	0~26.5	0~4.7	1.1~11.6	1.5~2.9	0~9.1	1.1~4.6	1.6~5.0	5.7~10.3	89.47	
噻虫嗪		0	0~1.0	0~17.5	0	0~8.1	0~2.9	0~2.4	0.5~21.7	47.37		
噻虫胺		0	0	0~1.2	0	0	0	0	0	5.26		
呋虫胺		0.5~1.3	0	0~1.0	0~0.2	0	0~0.2	0	0	26.32		
烯啶虫胺		1.2~2.4	0	0~2.0	0~0.3	0	0	0~0.3	0~0.2	28.95		
噻二嗪类		噻嗪酮	0.1~3.7	0.1~0.7	0.1~1.0	0.4~0.6	0.3~6.6	0.2~4.0	0~3.7	0.8~1.7	97.37	
		吗啉类	0~0.7	0.3~10.9	0.6~15.4	2.7~4.8	0~5.6	0~2.5	0~0.7	1.5~10.4	76.32	
		甲氧基丙烯酸酯	啉菌酯	0~5.2	0.1~0.6	0.4~2.4	0.9~1.1	0.3~2.4	0~1.0	0~5.2	1.7~11.4	84.21
			啉菌酯	0	0	0	0~0.2	0	0	0	0	2.63
	甲氧虫酰肼		0	0	0	0~0.3	0	0~0.5	0~0.5	0~0.6	21.05	
	吡啶啉类		0	0	0	0	0	0	0	0	0	
	三嗪类		1.1~1.5	0.6~2.1	0~0.5	0	0~0.8	0	0	0	36.84	
	吡嗪酮类		1.3~8.2	3.7~8.2	0~0.8	0	0~0.5	0	0	0	36.84	
	苯乙酸类		0.4~1.1	0	0	0	0	0	0	0	10.53	
	二噻类		0	0	0	0~0.2	0	0	0	0	2.63	
生物源	阿维菌素		0.5~1.2	0.6~2.0	0~0.5	0	0~1.2	0	0	0	42.11	
	甲氨基阿维菌素苯甲酸盐		0	0	0	0	0~0.2	0~0.5	0	0	15.79	
	稻瘟灵	0~12.3	0~0.2	0~0.3	0	0~3.2	0~0.2	4.2~6.9	1.2~6.0	57.89		





$\text{mg} \cdot (\text{kg} \cdot \text{d})^{-1}$ , 以 bw 计]<sup>[39]</sup> 以及斑马鱼的发育毒性 ( $0.16 \mu\text{g} \cdot \text{L}^{-1}$ )<sup>[40]</sup>. 由于其对非目标生物的高毒性, 多菌灵在一些发达国家, 如美国已被禁止使用<sup>[41]</sup>, 但在其他许多国家仍在使用<sup>[42]</sup>.

对于绿藻来说, 高风险农药主要为乙草胺, 如图 3 所示. 在 7 个水库中, 乙草胺存在的生态风险位点最多, 其中  $\text{RQ} > 1$  的位点有 33 个,  $0.10 \leq \text{RQ} < 1$  的位点达 4 个, 共占总采样点的 100%; 其次是莠灭净,  $0.10 \leq \text{RQ} < 1$  的位点占比为 5.40%. 乙草胺是

世界范围内使用最广泛的除草剂之一, 已在地表水、土壤和地下水中检测到其残留<sup>[43]</sup>. 它不仅对浮游动物和低等脊椎动物有剧毒, 而且可以在人类肝癌细胞系中诱导氧化应激和抗氧化活性<sup>[44,45]</sup>. 有研究表明, 除草剂和杀虫剂引起的浮游植物丰富度和组成的改变与生态系统的变化相联系<sup>[45]</sup>, 因此, 不应忽视其对水生生态系统的潜在负面影响, 需要采取适当措施, 减少使用量, 并通过化学或生物降解等方法来降低其风险水平.

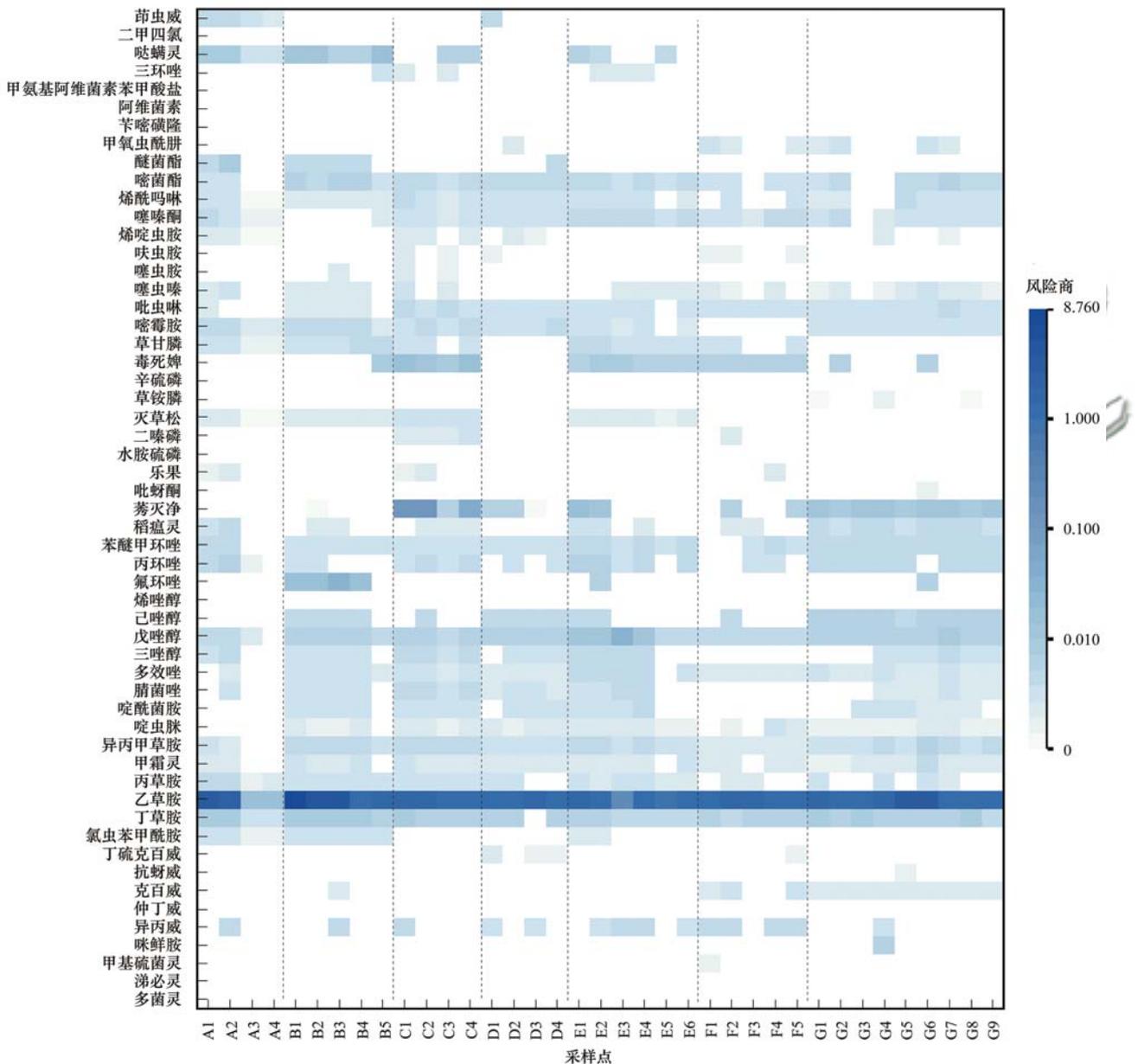


图 3 55 种农药对绿藻的风险评估

Fig. 3 Risk assessment of 55 pesticides on green algae

### 3 结论

本研究对东南某省某区域 7 个水库水样进行检测, 选择了 19 类农药进行综合评价. 同时, 选择 3 种不同营养级的水生生物进行了风险评估. 在 55 种农

药中, 26 种农药检出频率大于 50%, 12 种农药检出频率在 80% 以上, 表明该省区域农药使用的广泛性和其种类的多样性. 在风险评估中发现, 大部分农药对于 3 类生物都处于低风险状态, 其中多菌灵、毒死蜱、吡啶酮和戊唑醇为目标区域的潜在风险污染

物,可作为水库的指示性农药来检测和评估该区域的水处理效率。初步风险评估发现,对于绿藻而言,乙草胺是其生态风险主导的农药;对于水蚤和鱼来说,生态风险主导的农药均是多菌灵。尽管该区域整体生态风险水平较低,但不应忽视这些农药对水生生态系统的潜在负面影响,应重点关注这些农药在农业活动中的使用,不仅需要提高居民意识、减少使用量,还应通过加强目标区域的水环境治理来降低风险水平。

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