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滇池大气沉降氮磷形态特征及其入湖负荷贡献

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摘要:为研究季节变化和降雨量对滇池各种氮磷形态浓度的影响,采用紫外分光光度法测定大气沉降的各种氮磷形态浓度,探讨滇池湖面氮磷对水污染的贡献.结果表明,滇池大气沉降氮浓度普遍符合雨季低,旱季高的特点;大气沉降氮磷负荷与降雨量正相关,季节性变化主要呈雨季高,旱季低.大气沉降氮负荷以 DIN 为主,占总氮沉降负荷的 63.70%;磷负荷以 PP 为主,占总磷沉降负荷的 45.54%,过度施肥和肥料中氮磷的流失是大气湿沉降中主要的氮磷来源.结合入湖河流数据,滇池大气沉降中 TN 和 TP 的沉降量分别为河流入湖负荷的 6.14% 和 12.76%,因而滇池主要污染来源仍然是入湖河流带来的负荷.但滇池大气沉降氮磷通量与其他地区相比处于中等偏上地位,所以该贡献仍需重视.

关键词:滇池;大气沉降;氮磷;形态特征;入湖负荷

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Characteristics of Nitrogen and Phosphorus Formation in Atmospheric Deposition in Dianchi Lake and Their Contributions to Lake Loading

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Abstract: To examine the effects of seasonal changes and precipitation on the concentrations of various nitrogen and phosphorus forms in Dianchi Lake, the concentrations of various nitrogen and phosphorus forms of atmospheric deposition were determined by UV spectrophotometry. Additionally, the contributions of nitrogen and phosphorus to water pollution in Dianchi Lake were discussed. The results showed that the atmospheric depositional nitrogen concentration in Dianchi Lake is generally consistent with the characteristics of the low rainy season and high dry season. The nitrogen and phosphorus load of atmospheric deposition was positively correlated with rainfall. Seasonal changes were mainly characterized by low dry season and high rainy season. The atmospheric depositional nitrogen load was dominated by dissolved inorganic nitrogen, which accounted for 63.70% of the total nitrogen deposition load. The phosphorus load was mainly PP, which accounted for 45.54% of the total phosphorus precipitation load. Excessive fertilization and loss of nitrogen and phosphorus from fertilizers are the major sources of nitrogen and phosphorus in atmospheric wet deposition. Combined with data from rivers entering the lake, the settlements of TN and TP in the atmospheric deposition of Dianchi Lake were 6.14% and 12.76% of the river load, respectively. Therefore, the primary source of pollution in Dianchi Lake was still the load brought by the river into the lake. However, the nitrogen and phosphorus fluxes in the atmospheric deposition of Dianchi Lake were at intermediate levels compared with other regions, so this contribution requires further investigation.

Key words: Dianchi Lake; atmospheric deposition; nitrogen and phosphorus; morphological characteristics; loading into the lake

外源性氮磷负荷是控制淡水生态系统初级生产力的关键因素之一^[1],外源氮磷负荷的持续增加是导致水生态系统退化,诱发水体富营养化和有害藻类水华的主要原因.其中大气干湿沉降是近年来逐渐被认识,且被认为是增长较快的氮磷外源负荷之一^[2]

大气干湿沉降中氮的来源分为自然源与人为源 两部分. 自然条件下固氮微生物或闪电作用产生的 含氮化合物称为自然源; 化石燃料燃烧、交通运输、化肥施用等人类生产生活等产生的含氮化合物

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称为人为源^[3]. 大气中铵态氮主要来自土壤、海洋、肥料和家畜粪便中铵态氮的挥发^[4], 大气硝态氮由雷击、生物固定或工业、汽车使用的化石燃料和生物燃料燃烧产生^[5]. 大气中有机氮来源较为复杂,可源于一次或二次反应^[6]: 二次有机氮化物包括烷基硝酸盐以及氨基化合物,NO_x 与挥发性有机物(VOCs)通过气固凝结作用可得到二次的有机硝酸盐气溶胶. 与氮相比, 大气中磷的存在比较稳定,主要来源于岩石风化、沙尘、生物质燃烧、花粉、工业生产等产生的气溶胶和颗粒^[3].

目前,大气氮素沉降已成为太湖流域农田供氮和水体氮污染的重要来源^[7];近30年来,我国长江流域平均每年大气氮沉降量可达到该流域氮素总输入量的20%~30%^[8];上海地区受大气氮污染的降雨中氮浓度已超过水体富营养化阈值0.20mg·L^{-1[9]}.因此,大气氮磷沉降不仅能维持初级生产力所需营养,还是外源污染负荷.

滇池是中国西南地区最大的高原湖泊,也是我国重度富营养化湖泊之一. 滇池流域大量污染物排人湖内,导致其水质已下降为劣 V 类水体. 即使已采取治理措施,但仍有水华暴发等自然现象发生. 雨季时,降雨导致大量污染物汇入水体,随温度升高,水质快速恶化. 本文通过滇池点位的表观观测,获得滇池地区不同形态氮磷的大气沉降通量,并分析时空变化以及各形态组成,分析其来源以及对滇池水污染的贡献. 同时引用大量数据比较滇池地区与其他流域大气沉降,讨论滇池地区大气沉降氦磷负荷特点与污染程度.

1 材料与方法

1.1 观测站位置与样品采集

2014 年在滇池北部和东部设置 2 个大气降雨样品收集点(图 1). 每次降雨后立即将收集箱内雨水带回实验室,样品混合均匀测定总氮(TN)、总磷(TP)浓度,样品通过 0. 45 μm 微孔滤膜后测定溶解性总氮(DTN)、总磷(DTP)、氨氮(NH₄⁺-N)、硝态氮(NO₃⁻-N)、溶解性无机磷(SRP)等,根据每月降雨及样品采集次数,计算得到每月多次降雨氮磷浓度平均值及标准偏差. 在此基础上,根据每月收集箱内雨水体积,并参考监测站 2014 年滇池流域降雨量及历年降雨量数据,计算滇池湖面大气湿沉降氮磷入湖负荷.

1.2 分析及计算方法

 ρ (TN)和 ρ (DTN)采用碱性过硫酸钾氧化-紫外分光光度法测定; ρ (NH₄+N)采用纳氏试剂分光光度法测定; ρ (NO₃-N)采用紫外分光光度法测定;

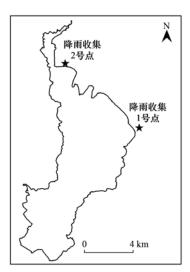


图 1 大气降水收集站点位置示意

Fig. 1 Location map of atmospheric precipitation collection site

ρ(NO, -N)较低忽略.

$$\rho(DON) = \rho(DTN) - \rho(DIN)$$

$$\rho(PN) = \rho(TN) - \rho(DTN)$$

$$\rho(DIN) = \rho(NH_4^+-N) + \rho(NO_3^--N)$$

式中,DON 为溶解性有机氮,DIN 为溶解性无机氮,PN 为颗粒态氮,单位均为 $mg \cdot L^{-1}$.

1.3 数据统计分析

数据统计分析及制图采用 Origin 8.5 和 Arcgis 10.2 软件.

2 结果与讨论

2.1 滇池湿沉降氮磷浓度及其形态特征

滇池流域大气湿沉降总氮、总磷及不同形态氮 磷浓度变化见图 2. 2014 年 1~12 月滇池流域大气 湿沉降总氮及不同形态氮浓度总体呈现"S"型曲线 变化趋势, 以 5 月最低, 9 月最高. 其中大气湿沉 降 TN 含量浓度变化在 0.66~2.0 mg·L⁻¹之间, 年 平均值为 1.36 mg·L⁻¹; PN 浓度变化在 0.04~0.35 mg·L⁻¹之间, 年平均值为 0.21 mg·L⁻¹; DIN 浓度 变化在 0.58~1.3 mg·L-1之间, 年平均值为 0.93 mg·L⁻¹; DON 浓度变化在 0.037~0.45 mg·L⁻¹之 间, 年平均值为 0.22 mg·L⁻¹. 大气湿沉降 DIN 浓 度明显高于青岛沿海地区[10], 9月 TN 浓度(2.051 mg·L-1)已高于湖泊富营养氮浓度阈值. 1~12 月 滇池流域大气湿沉降磷浓度与氮浓度变化趋势不 同. TP 和 PP 浓度变化分别在 0.037 ~ 0.081 mg·L⁻¹和 0.01 ~ 0.058 mg·L⁻¹之间, 年平均值为 0.13 mg·L⁻¹和 0.059 mg·L⁻¹, 二者均以 9 月处在 较低的水平, 其它月变化不明显. DIP 浓度变化在 0.019~0.025 mg·L⁻¹之间,年平均值 0.039 $mg \cdot L^{-1}$,以2~5月较低,6月显著增加,之后呈现

下降趋势. DOP 浓度变化在 $0.005 \sim 0.094 \text{ mg} \cdot \text{L}^{-1}$ 之间,年平均值为 $0.029 \text{ mg} \cdot \text{L}^{-1}$,以 10 月最高,是其它月的 3 倍左右, $5 \sim 9$ 月达到最低水平.

大气湿沉降氮磷浓度与温度、光照、降雨量和人类活动等因素关系密切,余辉等[11]的研究表明,太湖区域大气湿沉降氮浓度与降雨量变化趋势相反且符合夏季低、冬季高的规律.本研究大气湿沉降氮浓度年内变化也与降雨量有关,滇池位于云贵高原中部气候环境特殊,年内旱、雨两季分明[12].雨季(5~10月)来自海洋的西南暖湿气流长时间向该

区提供大量水汽,因而降水频率高,气溶胶等离子在空气中存留时间较短,从而造成雨水中氮磷浓度较低;旱季(11月至次年4月)受西风南支槽和二次蒸发的影响导致降水频率低,气溶胶等粒子在空气中存留的时间相对较长,所以在降水中容易出现较高的浓度值.而磷浓度的变化较为稳定,主要原因在磷酸钙等正磷酸盐和磷酸氢盐溶度积较小,难溶于水多以含磷颗粒物的形式存在在气溶胶中,不易随气流扩散到较远的地区.因而,滇池区域湿沉降的氮浓度更加符合旱季高,雨季低的变化趋势.

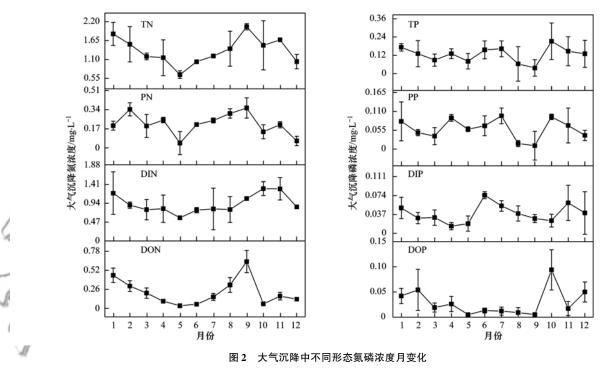


Fig. 2 Monthly changes in different forms of nitrogen and phosphorus concentration in atmospheric deposition

大气湿沉降氮磷浓度和负荷变化趋势与降雨量变化密切相关. 由图 3 可见,近 10 年来昆明地区的主汛期集中在 6~8 月,平均年降雨量为 719 $mm^{[13]}$. 2014 年滇池地区年降雨量为1 078 mm 远高于近 10 年降雨量. 与往年趋势一致,自 4 月起降雨量逐月大幅增加,8 月后逐步递减直至 10 月保持稳定且较低的降水量. 不同的是,7 月降水量出现倒峰,但其降雨量仍是非主汛期降雨量的若干倍. 经计算,2014 年 5~9 月的降雨量综合占全年降水量的 89.1%,与之对应月的 $\rho(TN)$ 、 $\rho(DIN)$ 、 $\rho(DIP)$ 、 $\rho(DOP)$ 受到雨水的淋溶作用均略低于其他月.

根据滇池全年降雨氮磷浓度及昆明站降水数据,2014年滇池湖面全年总氮沉降负荷为422.96t,这其中的溶解性总氮负荷为348.58t,颗粒态氮负荷为79.17t,溶解性无机氮负荷为269.41t,DON负荷为74.37t.大气沉降氮负荷以溶解性无

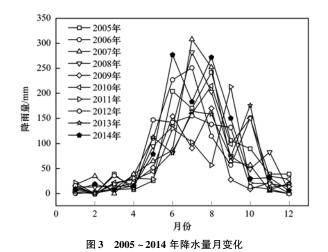
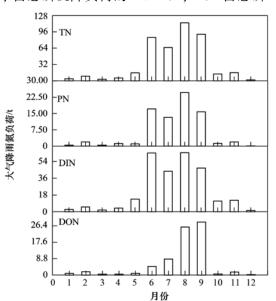


Fig. 3 Monthly changes in precipitation from 2005 to 2014

机氮为主,占总氮沉降负荷的63.70%,颗粒态氮占总氮沉降负荷的18.72%,DON占总氮沉降负荷的17.85%.大气沉降氮负荷主要集中在6、7、8和9月,占全年氮沉降负荷的84%以上(图4).

2014 年滇池湖面全年总磷沉降负荷为 35.80 t, 其中,溶解性总磷负荷为 19.95 t, 颗粒态磷负荷为 16.44t,溶解性无机磷负荷为 15.21 t, DOP 负荷为 4.45 t. 大气沉降磷负荷以颗粒态磷负荷为主,占总磷沉降负荷的 45.54%,其次为溶解性无机磷负荷,占总磷沉降负荷的 42.14%, DOP 占总磷



沉降负荷的 11.86%. 大气沉降磷负荷主要集中在 6~8 月,占全年氮沉降负荷的 75%. 雨季湿沉降中各形态氮磷的负荷占全年的氮磷指标负荷 70%以上. 夏季藻类大量繁殖,N、P等营养盐通过降水进入湖体很可能会促进藻类大量生长,加剧了滇池的富营养化.

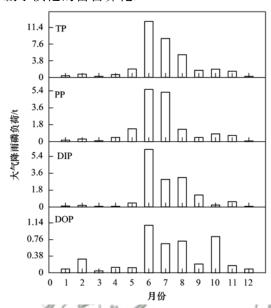


图 4 大气沉降中不同形态氮磷负荷月变化

Fig. 4 Monthly variation in different forms of nitrogen and phosphorus loading in atmospheric deposition

2.2 滇池湿沉降氮磷比及其对藻类的影响

研究大气湿沉降中不同形态的比例变化, 对分 析营养因子来源以及明确治理方向具有重要意义. 如图 5 所示, DIN 占 TN 的比例范围是 57% ~ 87%, 远高于 PN、DON 所占的比例. DON/TN 与 DIN/TN 处于此消彼长的趋势, DON/TN 有两个峰值, 分别 位于1月与9月.9月峰值非常明显,且从6月起 逐渐升高. 一般大气 DON 分为氧化态有机氮、还原 态有机氮和生物有机氮这3种,其中氧化态有机氮 通过无机氮与挥发性有机物(VOCs)反应生成[14], 雨季时段气温逐渐升高,且光照强烈,这些条件都 有助于有机氮的生成. 也就是说, 6~9 月有机氮的 持续升高与有机氮的持续降低均与有机氮的二次生 成有关. PP/TP 的波动范围 24%~71%, 旱季与雨 季相比波动较大,整体表现为旱季 > 雨季. PP/TP、 SRP/TP 的数值呈此消彼长的趋势, 雨季(5~10 月)时 SRP/TP 的数值明显升高, 9 月达到最大值 70.3%. 该现象说明, 旱季时期降雨量较少, 由生 物质燃烧、工业生产排放或农业施肥流失的颗粒态 磷在滇池上空富集: 雨季降雨量逐步增加对空气有 清洁作用因而 PP/TP 逐渐减少, 随之上升的是可溶 解在雨水中的磷酸盐. PP 进入水体后部分将直接 解吸进入水体,或形成沉积物称为内源磷负荷主要

来源,因此 PP 对水体富营养化的影响也不容忽视. DOP/TP 在 4~8 月间保持较低水平,降雨量减少后波动上升,全年均保持较低水平.目前评价湖泊磷污染特征仅考虑 SRP,但是何宗健等^[15]利用酶水解技术研究并指出滇池地区不同来源的 DOP 是与SRP 同等规模的生物可利用磷源,共同维持了滇池富营养化.

水体中氮磷比会影响水生植物初级生产力和群落组成,且对藻类暴发性生长具有重要意义.大气湿沉降氮磷负荷与日俱增将改变水体中的营养结构.如图6所示,TN/TP波动范围在6.6~56之间,除8、9月骤升至波峰56后,其他月的比值都在10上下波动.

结合上文中的浓度变化,9月峰值的产生主要与逐步上升的TN和TP的下降有关.DON/DOP,NP/PP的变化趋势与TN/TP相近,均在9月出现峰值.而DIN/DIP在4月与10月均出现峰值(图6).水体中氮磷比小于7~10时,藻类生长表现为氮限制状态,生物固氮作用可通过消纳水体中较多的TP,调节TP/TN;氮磷比大于22.6~30时,磷为生物限制因子,该环境将抑制氮素有机合成过程从而调节TP/TN^[16].滇池上覆水的年平均氮磷比为21,9月大部分形态的氮磷比均超过50.9月湿

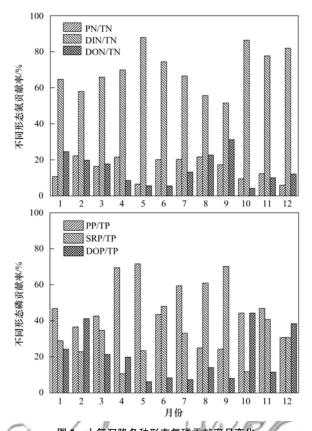


图 5 大气沉降各种形态氮磷贡献率月变化 Fig. 5 Monthly changes in various forms of nitrogen and phosphorus contribution rates in atmospheric deposition

沉降的较高氮磷比很可能将影响上覆水的营养结构,增加入湖区域水体蓝藻水华暴发的风险. 鉴于旱季各形态氮磷浓度较小,所以通过科学施肥,减少化石燃料等手段控制雨季氮素显得格外重要.

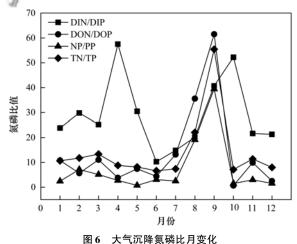


Fig. 6 Monthly changes in nitrogen and phosphorus in atmospheric deposition

刘莲等^[17]通过实验证明当N:P接近或低于Redfield比值时小球藻出现暴发性增殖.Paerl等^[18]认为富营养化水体的氮磷负荷都比较高,可能超过浮游植物的同化能力,氮磷比理论可能更适合于在类似安大略实验湖区营养盐相对缺乏的水体.谢

平^[19]在武汉东湖的围隔实验证明氮磷比无论小于大于 29 都能发生蓝藻(微囊藻)水华;蓝藻水华的暴发导致水体 pH 值上升,诱导沉积物大量释放磷,使得水体氮磷比降低,因此低氮磷比是蓝藻水华暴发的结果,而不是原因.许海等^[20]的研究结果表明,氮磷比对铜绿微囊藻和斜生栅藻(绿藻)生长的影响并不表现在一个确定值上,而与水体氮磷的绝对浓度有关,氮磷浓度比氮磷比对两种藻的生长影响更大.

2.3 滇池氮磷湿沉降入湖负荷贡献及对水质的 影响

滇池属于高原湖泊,湿沉降中营养因子的负荷变化趋势与其他地区相比存在一定的特殊性.因此本文选择珠江口^[21]与太湖流域^[22]以及气候条件较为相似的洱海^[23,24]的湿沉降氮磷浓度作对比.如图7所示,太湖水体受纳的TN湿沉降率波动在225~330 kg·km⁻²之间,分别在12月与4月出现两个波峰,分别与冬季供暖和春季降雨量增加、农业作业有关.TP湿沉降平均值为7.25 kg·km⁻².珠江口流域的平均月总氮湿沉降通量为194 kg·km⁻²,波动范围在0~661.9 kg·km⁻²,变化趋势仍然与当地播种施肥的时间有关.平均月总磷湿沉降通量为1.61 kg·km⁻²,与太湖地区相比,珠江口地区湿沉

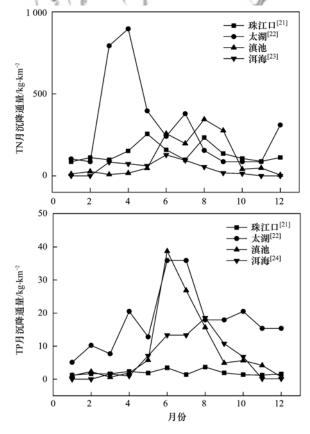


图 7 不同地区大气沉降氮磷通量月变化

Fig. 7 Monthly changes in nitrogen and phosphorous flux in atmospheric deposition in different areas

降中的营养因子较少. 洱海地区污染程度小, 大气 沉降中的营养因子与其它对比样数据相比相当低, 与滇池地区大气氮磷沉降季节性变化规律相似,5 ~10 月的氮磷总负荷均占全年负荷的 80% 以上. 滇池流域是重要的蔬菜和花卉生产基地, 化肥使用 量非常大(1015 kg·hm⁻²), 是全国水平(402 kg·hm⁻²)的 2.53 倍^[25]. 氮磷化肥使用时间应在雨 季开始之前(5月),考虑到氮磷挥发的滞后性,6 月时总氮总磷负荷明显升高与农业活动基本吻合. 可以推测, 滇池大气湿沉降氮磷的主要来源为农业 施肥.

通过不同湖泊年通量的对比可以分析滇池湿沉 降氮磷通量对水体的贡献. 本文收集了 4 处较为清 洁水域的湿沉降氮磷通量分别是弗拉特黑德湖、地 中海西、地中海东及波罗地海; 3 处人类活动频繁 的水域分别是斐伊川、滨海湾以及太湖(表1). 其 中弗拉特黑德湖位于美国西部,是世界最清洁湖泊 之一. 该地区的年大气沉降氮负荷仅有 335.8 kg·km⁻², 其中 82.6% 为无机氮, 氨氮居多. 地中 海地区与弗拉特黑德湖地区同为贫营养水域,东西 两地的大气无机氮年负荷分别为 512.4 kg·(km²·a)⁻¹和464.8 kg·(km²·a)⁻¹, 为弗拉特黑 德湖地区年大气氮负荷的1.7倍左右. 滇池, 冰海 湾(新加坡)、斐伊川(日本)等地的年大气氮沉降 负荷为弗拉特黑德湖地区氮负荷量的 4 倍以上

度污染的太湖地区是其负荷量 13.8 倍. 中国的滇 池、太湖地区的大气沉降磷年负荷与其他国家水域 相比处于较高水平,大小排序为滇池>太湖>斐伊 川>弗拉特黑德湖>波罗的海. 滇池湿沉降中的磷 负荷无论是总量还是波峰值均大于污染严重的太湖 地区,应格外重视. 综合以上对比, 滇池地区的氮 磷负荷变化特征均符合旱季低,雨季高的特点;年 湿沉降氮通量与其他人口密度大的地区相比处于中 等偏下水平,湿沉降磷年通量偏高.

滇池入湖污染源除大气沉降外,很大一部分营 养因子来自入湖河流. 结合同年滇池入湖河流水量 及水质数据, 滇池河流 TN 入湖负荷量为6 887.76 t·a⁻¹, TP 负荷量为 280. 51 t·a⁻¹. 滇池大气沉降中 TN 和 TP 的沉降量分别为河流入湖负荷的 6.14% 和 12.76%. 而太湖 2002~2003 年的研究结果为 17.3% 和12.8%[22],与之相比,滇池区域的干湿沉降中不 同形态的氮对湖泊富营养化的贡献并不严重. 但是 不仅湿沉降本身会带来一部分的氮磷负荷, 随机性 大雨产生的氮磷地表径流流失也会为湖水输入一笔 可观的氮磷负荷. 结合上文, 大气沉降中无机氮与颗 粒态磷对氮磷的贡献率最高,这两种成分经过时空 变化分析,确定主要来源应该是过度施肥导致,种植 生态拦截草也可以有效地控制土壤氮磷向水体迁 移[31]. 寻求合适的施肥量及施肥方式从而达到农业 与环境的共赢是目前待解决的问题.

表 1 不同地区大气湿沉降氮磷入湖负荷比较/kg·(km²·a)-1

Table 1	Comparison of atmospheric wet deposition of	f nitrogen and phosphorus in I	akes in different regions/kg·(km²·a)	•
	氮负荷		磷负荷	

位置	氮负荷		磷负荷			4± -6-			
14. 直.	TN	DIN	DON	PN	TP	DIP	DOP	PP	- 文献
弗拉特黑德湖(美国)	335. 8	277.4			29. 2	11.7			[26]
波罗的海	617	556	61		7. 3	5. 7	1.7		[27]
滨海湾(新加坡)	1 547. 2	1 078. 8	468. 4			21.5	29. 5		[28]
地中海西		464.8				17			[29]
地中海东		512.4				11.7			[29]
斐伊川(日本)	1 702. 3	1 562. 8	69.8	69. 8	53. 9	21. 2	4. 9	27.8	[30]
太湖(中国)	4 648. 8	2 827. 5			105. 7				[11]
滇池(中国)	1 363. 4	868.4	239. 8	255. 2	115. 4	49	14. 3	53	本研究

3 结论

- (1) 滇池地区大气沉降不同形态氮和有机磷浓 度雨季低,旱季高. TN、DTN、PN、DIN 的浓度均 在9月达到峰值. 大气沉降磷浓度总体呈波动式先 降后升的趋势, 但浓度远低于不同形态 N, 均在 10 月达到峰值(DIP 除外).
- (2) DIN 占 TN 的比例范围是 57% ~ 87%, 贡 献率最大. 由于 DIN 可在高温强光照的条件下二次 生成 DON, DON/TN 与 DIN/TN 全年处于此消彼长

的趋势. 磷指标中 PP 贡献率的波动范围 24%~ 71%, 贡献率整体表现为: 旱季 > 雨季. 各形态氮 磷比均在8、9月达到峰值, 且均在50以上, 远高 于上覆水中的氮磷比.

(3)大气沉降氮负荷以溶解性无机氮为主,占 总氮沉降负荷的 63.70%;磷负荷以 PP 为主,占总 磷沉降负荷的 45.54%. 大气沉降氮磷负荷与降雨 量呈正相关关系, 氮磷负荷主要集中在雨季的6、 7、8和9月,分别占全年大气氮磷沉降负荷的84% 和75%以上.

(4)经过多地区对比,滇池大气沉降年氮负荷与其他人口密度大的地区相比处于中等偏下水平,但磷负荷水平偏高.滇池大气沉降中 TN 和 TP 的沉降量分别为河流入湖负荷的 6.14% 和 12.76%,对滇池水体的贡献不大,很大一部分营养因子来自入湖河流.

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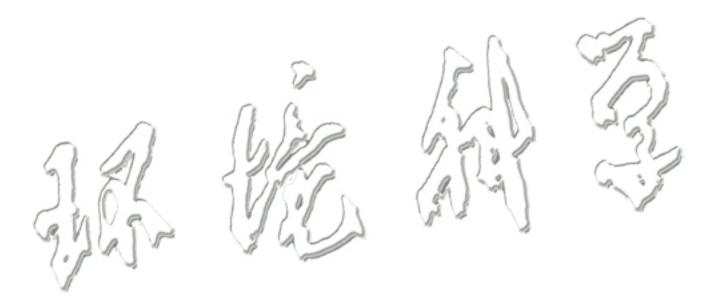
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