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ABSTRACT

Ten automatic samplers were used to collect dry deposits and rain water in the highly industrialized region of southwestern Taiwan since December 1985. During the dry season, whatever rain that falls frequently appears basic because it contains a large amount of basic minerals such as calcium carbonate released by several cement plants. During the wet season, however, the rain becomes increasingly acidic with the lowest pH value being 3.823.

Over sixty lakes all over Taiwan have been sampled. The results show that lake alkalinity is low in regions of igneous rocks but high for the sedimentary zone in western Taiwan, and variable for the eastern region with metamorphic rocks. The high-mountain lakes without local pollution, Shuang Lian Pyl and Taiy Feng Hui in Yilan County and Tian Chyr within the Yusan National Park, are acid-sensitive ones (with a pH of less than 6.3-6.7, a conductivity of less than 30-40 µS/cm, and an alkalinity of less than 300 eq/l). The Li Yu Tan at Puli, Li Yu Tan in Hualien County, Mei Hau Hui in Yilan County, Yuan Yang Hu in Hainan County, Tian Chyr on the Langyu Island and the Nan Jen Hu Group within the Kenting National Park could soon become acid-sensitive lakes. In general, low-elevation lakes are high in pH and alkalinity, which are related to the soil and rock type or man-made eutrophication from the inputs of fertilizers, pesticides and other agriculture chemicals. These lakes are not likely to be acidified in the near future.

Keywords: acid rain, lake acidification, water pollution, pH, alkalinity

I. Introduction

Lately acid rain has attracted much attention and concern worldwide. In Taiwan, the adverse effects of acid rain, the offspring of rapid industrialization, have also been felt and discussed. Lakes are showing both direct and indirect responses to the acidification of the water by acid rain. Directly, acid water affects the growth of organisms in lakes, changes the ecosystem, affects the growth of crops if acid water is used for irrigation, and speeds up the corrosion of generators and metal pipes if used to generate electricity [2, 3, 6-8, 16-18, 26, 27]. More circuitously, the release by acidification of heavy metals originally adsorbed on sediments results in high concentrations of heavy metals in the lake biota. Similarly, the heavy metal concentrations in crops will also increase if acidified water is used for irrigation. These processes pose serious health problems regardless of whether the acidified lakes constitute the source of drinking water, or whether man consumes food which has been irrigated with acid water and has accumulated high concentrations of heavy metals [5, 6, 9, 14, 15, 17-20, 25].

The process of lake acidification that leads to the adverse effects mentioned above obviously should be investigated thoroughly. Unfortunately, because of the lack of historical data, we do not know whether the lakes in Taiwan have changed, or will soon change their acidity. We report here the acidity of rain and lake water in Taiwan and the sensitivity of these lakes to acid rain using pH and alkalinity data. We also estimate possible future changes in lake acidity based on studies of total dissolved salt, rock and soil types [1, 10-12, 21-24].

II. Material and Methods

Ten automatic dry/wet samplers were used to collect dry deposits and rain water in southwestern Taiwan (Fig. 1). Dry deposits were collected periodically and examined by scanning electron microscope (JEOL JSM-35 CFSEM) and X-ray diffraction (Diano 8536 XRD). Over two hundred wet samples were collected and analyzed within 24 hours after the rain. The measurements include pH, alkalinity, conductivity, density, nitrate, sulfate, chloride, ammonia, calcium, magnesium, potassium and sodium. NBS pH 4.004 and 7.415 buffers were used to calibrate the Radiometer PHM 85 system, precise to ±0.003 units. Certified ERA (Environmental Resource Associate) potable water and NBS (National
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Bureau of Standards, USA) rain water samples were used as references. The precision and accuracy of the analyses are better than 94% (Table 1).

Over two hundred lake water samples were collected from 60 lakes (Fig. 1) either with a bucket (for small ponds) or with a Nansen bottle when boats were available. In remote areas without access to a boat, we used a remote-controlled vessel, which could be controlled to obtain samples from as deep as 15 m. Normally we collected samples at three depths at two locations for each lake. Temperatures and pH were measured immediately. Afterwards, samples were filtered with 0.45 μm Nalgene filter, stored in Amber heavy plastic bottles at 4°C, and shipped back to the laboratory for further analyses of the above mentioned parameters. The filter paper was pre-rinsed with sample before being used for filtration. The parameters measured are the same as those for rain water (Table 1). Reliability of data was further checked by correlating the sum of cations with the sum of anions. A linear correlation was found (r=0.98) with a slope of 1.03, which differs only slightly from the expected value of 1 (Fig 2). Detailed sampling and analytical procedures were described in [4] and [13].

III. Results and Discussion

Dry deposits consist mostly of quartz, gypsum and aragonite. Quartz probably comes from weathered rock and sand. Gypsum and aragonite probably come from several cement plants in this area. The latter is quite basic: one to fifty weight dilution of dry deposits with deionized/distilled water (pH = 5.66) results in solutions with pH values ranging from 7.34 to 10.23.

The pH of rain waters collected covers a wide range because of large differences in local conditions. pH values are frequently quite high in the dry season, perhaps because the samples contain a relatively large amount of airborne mineral particles. A pH value as high as 10.635 has been recorded, reflecting high gypsum and aragonite contents (Fig. 3). In general, the higher the alkalinity or conductivity, the higher the pH values appear. The pH range and the numerical mean of rain waters collected in each month are also shown in Fig. 3. The rain water becomes more acidic during the wet season (May-Sept). There is some indication that the rain water is less acidic at the onset of a rain. This phenomenon also suggests that basic particles contribute to part of our rain water samples. Later during a precipitation event, the water becomes more acidic because basic particles have previously been removed.

Rains brought in by three typhoons were less acidic. The mean pH values for normal rains in July through September were slightly more acidic than the mean pH values for all rains combined (Fig. 3).

Of the areas studied, Hsiao-gun (site 9) seems to be the most affected by the acid rain because it is adjacent to the heavy industrial area concentrated in southern Kaohsiung. Rain waters there are frequently below pH 5 (Fig. 4). The amount of nitrate is inversely correlated with pH, thus, nitrate apparently is a leading contributing factor for acid rain at Hsiao-gun. The pH also correlates inversely with excess sulfate (the amount of sulfate in rain water after correction for sea salts) between March and August, suggesting that excess sulfate also contributes to the formation of acid rain at this site. The effect of sulfate is potentially large because of its higher concentration than nitrate. The effect of excess sulfate, however, is not clear between December 1985 and Feb., 1986 because of high dust content in the samples.

Rain waters collected outside of the Kaohsiung area generally had a pH value greater than 5.0. The sulfate and nitrate concentrations are also lower in the less industrialized region. These results reflect the relation between acid rain and air pollution.

| Table 1. The Precision and Accuracy of the Selected Methods of Rainwater Analyses |
|---------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|
|                                  | NBS Rainwater                  | ERA Rainwater                   | Hsiao-gun Rainwater             |                                  |                                  |                                  |                                  |
|                                  | Certified                      | Dev. (%)                        | Certified                      | Dev. (%)                        | Certified                      | Dev. (%)                        | Certified                      |
| pH                              | b                               | 4.50                            | 4.30                           | 3.59                            | 3.58                           | 0.28                            |
| NO₃ (ppm)                       | a,c                            |                                 | 7.06                           | 7.50                            | 6.23                           | 18.0                            |
| Cl⁻ (ppm)                       | a,e                            | 0.24                            | 0.25                           | 4.2                             | 1.00                           | 0.98                            |
| SO₄²⁻ (ppm)                     | a,d                            | 2.69                            | 2.64                           | 1.9                             | 10.80                          | 10.66                           |
| Na⁺ (ppm)                       | a,f                            | 0.20                            | 0.21                           | 5.0                             | 0.419                          | 0.416                           |
| K⁺ (ppm)                        | a,f                            | 0.05                            | n.d                            | n.d                             | 0.106                          | n.d                             |
| NH₄⁺ (ppm)                      | a,g                            |                                 |                                | n.d                             | 11.0                           |
| Mg²⁺ (ppm)                      | a,f                            | 0.024                           | n.d                            | n.d                             | 0.051                          | n.d                             |
| Ca²⁺ (ppm)                      | a,f                            | 0.014                           | n.d                            | n.d                             | 0.049                          | n.d                             |
|                                  |                                 |                                  |                                |                                 |                                 |                                  |                                |
|                                  | a: Ion Chromatography         | d: Turbidimetric method         | g: Phenomen method              |                                  |                                  |                                  |                                  |
|                                  | b: Potentiometry              | e: Mercury thiocyanate          | n.d: not detectable             |                                  |                                  |                                  |                                  |
|                                  | c: Cadmium reduction          | f: Atomic absorption spectrophotometry |                                  |                                  |                                  |                                  |                                  |

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Fig. 1. Locations of automatic rain water samplers (+, numbered 1 through 10) in southwest Taiwan and lakes sampled all over Taiwan (*). Letters A through F denote the alkalinites of lakes. Zones I through III are described in the text.
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$$\text{Sum}(\pm) = \text{Sum}(-) \times 1.029 + 0.066 \\ r = 0.980$$

![Graph showing the relationship between sum of cations and sum of anions for lake waters.](image)

**Fig. 2.** The sum of cations [sum (+)] correlated with the sum of anions [sum(−)] for lake waters.

Besides the three reservoirs in Penghu County and the Nan Jen Hu group which contain mainly sodium and chloride, most other lakes are dominated by calcium and bicarbonate ions. Consequently, calcium correlates linearly with alkalinity ($r=0.85$, Fig. 5).

We divide lakes in Taiwan into six categories based on alkalinity according to the classification of Zimmerman and Harvey [28] (Fig. 1): A (0.05 meq/l); B (0.05-0.2 meq/l); C (0.2-0.5 meq/l); D (0.5-1.0 meq/l); E (1.0-2.0 meq/l) and F (>2.0 meq/l).

As a first approximation, we also divide Taiwan into three major zones according to the rock and soil compositions (Fig. 1). Zone I consists mainly of igneous rocks such as andesite, andesitic pyroclastics, basalt, and basaltic tuff and flows. These rocks have a low buffering capacity and the lakes in this region should have low alkalinites and potentially low pH values. Unfortunately, we have collected only one sample from zone I. The Tian Chyr on the Lanyu Island has a rather low alkalinity of 0.126 meq/l and a slightly acidic pH of 6.61 in November, 1985. Further acidification is possible because of the low buffering capacity of the lake water and the surrounding rocks and sediments. This lake, however, apparently dries up periodically. It was dry when we returned for resampling in February, 1987.

Zone II consists mainly of non-carbonaceous sedimentary and metamorphic rocks such as mudstone, sandstone, tuffaceous sandstone, shale, coaly shale, sandy shale, gravel, conglomerate, agglomerate, argillite, slate, phyllite, and schists. These rocks have buffering capacities

![Graph showing pH ranges of rain waters collected in southwest Taiwan.](image)

**Fig. 3.** The numerical mean and the pH ranges of rain waters collected in southwest Taiwan. Dashed line in July through September are values excluding rains brought in by typhoons.

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between igneous rocks (zone I) and carbonaceous rocks (zone III). The lakes in this region show a wide variety of alkalinity and pH values. The ones worthy of notice are Shuang Lian Pyi (alkalinity 0.074 meq/1, pH 6.02), Tauey Feng Hu (alkalinity 0.003-0.008 meq/1, pH 4.68-4.84), and Mei Hua Hu (alkalinity 0.425-0.437 meq/1, pH 7.65-8.16) in Yilan County, Li Yu Tan at Puli (alkalinity 0.199-0.205 meq/1, pH 8.88-8.91), Li Yu Tan in Hualien County (alkalinity 0.28-0.695 meq/1, pH 5.96-6.99), Yuan Yang Hu in Hsinchu County (alkalinity 0.44 meq/1, pH 7.37), Tian Chyr within the Yusan National Park (alkalinity 0.014 meq/1, pH 5.9) and the Nan Jen Hu group within the Kenting National Park (alkalinities < 0.115 meq/1, pH 5.05-6.86). These lakes are either acid-sensitive or might soon become acid-sensitive. Since little local pollution occurs near these lakes, acid rain and local geochemistry are possible causes of acidification.

Of particular interest is the Nan Jen Hu group which did not exist prior to 1980. The lake waters contain mainly NaCl which probably comes from rain water and sea spray. We do not know whether this group of lakes is in equilibrium (or at steady state) with their bottom sediments or the surrounding rocks and soils (acidic laterite). More intensive study of these lakes is currently underway.

Zone III consists mainly of gravel, sand, clay, limestone and the alluvium zone. Most lakes have a alkalinity higher than 1.0 meq/l and the waters are generally slightly basic (pH between 7.0 and 9.0). A few lakes are heavily polluted. We do not believe that any of these lakes are in danger of acidification in the foreseeable future.

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臺灣地區之酸雨及湖沼酸化程度

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摘要

由1985年12月開始，本研究利用三座自動集雨器，收集臺灣西南工業化地區之乾降及雨水。乾季所收集之雨水中常含有大量可能由水泥廠排放之硫酸鈣等鹼性物質，使得雨水呈現鹼性。而季時雨水則經常呈酸性，最高時 PH 爲3.823。

臺灣地區超過60個湖沼及水庫也曾加以研究。於火成岩地區之湖水總鹼度甚低，於西部沈積岩地區總鹼度甚高，於東部變質岩地區則湖水總鹼度變化甚大。無當地污染源之高山湖泊，如雙連埤，翠峰湖及玉山國家公園內之天池已經屬於酸化敏感湖泊（PH 小於6.3至6.7，導電度大於30至40μs/cm，鹼度小於300μeq/l）。埔里鯉魚潭，花蓮鯉魚潭，宜蘭梅花湖，新竹鯉魚湖，蘭嶼池塘以及墾丁國家公園內之南仁湖區可能很快會轉變成酸化敏感湖泊。低海拔湖泊一般來講 pH 及總鹼度較高，除與土壤，岩石之分佈有關外，亦有可能因為農業，肥料之流入，造成人為優養化。這些湖泊短期內不太可能酸化。