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ASSESSING THE IMPACT OF LEATHER INDUSTRY TO WATER QUALITY IN THE AOJING WATERSHED IN ZHEJIANG PROVINCE, CHINA

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Abstract. The development of the leather industry in the Aojiang watershed of Zhejiang province increased the release of waste water. In the waste water, ammonium nitrogen (NH_4^+-N) and germanium (Ge) are the main pollutants. In recent years, literature has documented that the intake of high concentrations of NH_4^+-N and Ge harms human health and biological species. This paper focuses on assessing the trends of NH_4^+-N and Ge concentrations in the released waste water in Aojiang watershed and on understanding their relationships with the released waste water using regression and correlation statistics. The paper also utilizes the integrated pollution index to evaluate the water quality in the watershed. Preliminary results show that, from 1992 to 1998, the concentrations of NH_4^+-N and total Ge increased 13 and 14 times, respectively, and they decreased somewhat after 1998. The concentrations of NH_4^+-N and total Ge are positively correlated to the amount of released waste water. These concentrations of NH_4^+-N and Ge, respectively, exceed 12 and 3 times, of the water standards. The water quality in the watershed degraded from Type III in 1992 to over Type V in 2003 when they were compared with the national water quality standards. It appeared that the pollution had positive correlation with leather industry production. The degraded water has no doubt affected human health and the ecosystem health. These results can provide scientific information for the local government to reasonably adjust the industry structure and reduce the pollution to protect the environment.

Keywords: watershed water quality, NH_4^+-N, Ge, leather industry waste water, impact assessment, ecological risks

1. Introduction

The increased number of leather industries in the Aojiang watershed of Zhejiang province generated a large amount of waste water carrying toxic contaminants that harm human and ecosystem health. Although the emerging leather industry stimulated the local economy (Wenzhou Statistics Bureau, 2001), the cost of environmental pollution has to be dealt with in order to sustain the balance of industrial development, human health and ecology (Wenzhou EMC, 2002; Handsell *et al.*, 2003; Xie, 2004). Ammonium nitrogen (NH_4^+-N) and total germanium (Ge) are the main pollutants in the released waste water of the leather industry (Wenzhou EMC, 2002).

The impact of high concentrations of nitrogen in drinking water to cause methemoglobinemia or blue baby syndrome, a condition where nitrate interferes with the transport of oxygen in babies, and rivers nitrifications where high nitrogen concentrations present has also been well documented (Cooke and Kalita, 2002; Cébron *et al.*, 2003; Skipton and Hay, 2004; Portage County Ground water, 2004). One study conducted in 1997 reported that nitrate levels in excess of 4 mg-N/L were found to be highly correlated with increased risk of non-Hodgkins lymphoma in rural areas (Cooke and Kalita, 2002).

Germanium is a heavy metal that is known to irritate the eyes, the skin and the respiratory tract (Lide, 1998; Lenntech, 2004). The substance in blood may cause effects in lesions of blood cells. Exposure may result in death for fish species (Lenntech, 2004). Wang and Ding (2002) reported that the high concentration of the Ge could harm human body by causing the protein fixation and stimulating digestion system.

High concentrations of NH_4^+-N and Ge have impacted the local ecosystem and human health (Xie, 2004). According to the news media network, Xie (2004) reported that, a town called Shuitou, in the Aojiang watershed, the crops had little harvest; air quality was full of bad smell; and water was contaminated; strange diseases frequently prevailed; and few youngsters could pass physical exams for army admission. Although these phenomena can be the integrated effects of many factors in the region, the reduction of the NH_4^+-N and the Ge pollutants in water bodies appeared to be a worthwhile and urgent environmental goal. Therefore, the objectives of the study are to examine the concentrations of NH_4^+-N and Ge in the released waste water, to assess the water quality in the Aojiang watershed, and to understand the impact of these pollutants to human health and ecosystem health in the watershed and the surrounding ocean.

2. Materials and Methods

2.1. Study site

Aojiang watershed is located in the southern Wenzhou metropolitan district in Zhejiang Province. It is one of the four major watersheds releasing water to the open sea in Wenzhou. This watershed originated from the town of Shuitou in Pingyang County (Figure 1) covering 82 km in length and running through four main towns including Shuitou, Xiaojiang, to Aojiang and Longgang and then releasing water to the ocean. Among these towns, the leather industry is mostly concentrated in Shuitou, the head water of the watershed.

Using the national standards for determining the surface water quality functions (National EPA, 1997), Wenzhou Environmental Protection Bureau assigned surface water quality as Type III function for Aojiang watershed in 1992.

2.2. Data sources

The monitoring data of water quality was from the Wenzhou Environmental Monitoring Center and Pingyang Environmental Monitoring Stations (Zhang,

Figure 1. Aojiang watershed and the sampling locations.

2004). The economic data of agriculture, industry and populations was based on the Wenzhou Statistics Yearbook (2001). The waste water release was calculated using the method of coefficients which was based on the integration of empirical release coefficients per unit product from multiple years and locations to estimate the amount of pollutants release. The formula is defined as $W = K * A * B * C + D$, where W refers to waste water release and the unit is ten thousand ton; K refers to the number of enterprises and the unit is number of enterprises; A refers to product waste water demand and the unit is ten thousand ton per ton of product and number of processing machines; B refers to number of processing machines and the unit is number of processing machines per enterprise; C refers to tons of product produced per unit of processing machine and the unit is ton of product per number of processing machines; and D means the other water use and the unit is ten thousand ton.

The sampling sites were in Litou, Jiangyu, Fangyanxia, and Jiangkou (Figure 1), and the sampling periods were from 1992 to 2003. Samples were taken two times during ‘Ping Shui Qi’ (the flat water period received the amount of water around 25% of annual rainfall), March, April, May, October, and November; then two times during “Feng Shui Qi” (the plentiful water period received the amount of water close to 60% of annual rainfall), June, July, August and September; and two times during ‘Ku Shui Qi’ (the scarce water period received the amount of water is about 15% of annual rainfall), December, January, and February. The annual concentrations were the averages of these samples.

2.3. Analytical Methods and Standards for the Evaluations

The “Ge-Rou” method is commonly used in the leather industry, and the chemistry reaction uses Na_2CrO_4 to change Ge to Ge^{3+} under certain suitable conditions. The main side products are the NH_4^+-N and the Ge. The analytical methods of GB/T7479-1987 (National EPA, 2001) and GB/T7467-1987 (National EPA, 2001) were used to analyze the concentrations of NH_4^+-N and Ge. The unit for measuring

TABLE I
Standard values of elements for Type III surface water environmental quality, GB3838-2002, (National EPA, 2001)

Monitoring constituents	Standards
pH	6–9
NH_4^+-N (mg/l)	1.0
Ge^{6+} (mg/l)	0.05
COD (mg/l)	20
BOD (mg/l)	4

TABLE II
Standard values of first class pollutant release, (GB8978-1996), (National EPA, 2001)

Monitoring constituents	Standards
pH	6–9
NH_4^+-N (mg/l)	15
Ge^{6+} (mg/l)	1.5
COD (mg/l)	100
BOD (mg/l)	20
Sedimentations (mg/l)	70

the contaminant concentrations is 'mg/l' and the unit for measuring the loadings is 'ton'. The monitoring samples were analyzed by Wenzhou Environmental Monitoring Center and Pingyang Environmental Monitoring Station.

Water quality was evaluated using the standards of GB3838-2002 (Table I, National EPA, 2001) for the Type III function district and waste water was evaluated using the standards of GB8979-1996 (Table II, National EPA, 2001) for the first class release.

2.4. Pollutants and method for assessment

Ammonium nitrogen and total germanium are the focus in the study. In addition, the amount of chemical oxygen demand (COD), biochemical oxygen demand (BOD), and sedimentation in the waste water was also examined.

An integrated pollution index and standard exceeding index were used in this study to evaluate the water quality. The integrated pollution index is defined as the addition of the ratio of annual average of the pollutants to their standards. The formula is as follows:

$$P_j = \sum_{i=1}^{n} P_{ij} \quad k_i = \frac{p_{ij}}{pj} \times 100\% \quad p_{ij} = \frac{c_{ij}}{c_{io}}$$

where P_j – water quality integrated pollutant index in j^{th} profile, P_{ij} – i^{th} pollutant integrated pollution index in j^{th} profile, C_{ij} – annual average of the i^{th} pollutant in the j^{th} profile, C_{io} – annual average of the i^{th} pollutant, k_i – dividing coefficient of the i^{th} pollutant in the j^{th} profile, n – number of pollutants in the evaluation

The exceeding index is defined as the ratio of the i^{th} pollutant to its standard. The formula is as follows.

$$T_i = \frac{B_{il}}{B_{io}}$$

where T_i – Exceeding index of the i^{th} pollutant, B_{il} – The i^{th} monitoring result on the i^{th} pollutant, B_{io} – Evaluation Standard for the i^{th} pollutant

Statistical regression and correlation methods were used in this study to assess the relationships of water quality and released waste water and other factors. The linear regression model is described as

$$Y = a + bX$$

where b – regression coefficient

$$b = \frac{\sum (x - \bar{x})(y - \bar{y})}{\sum (x - \bar{x})^2}$$

a – regression intercept

$$a = \frac{1}{n}\sum y - b\frac{\sum x}{n} = \bar{y} - b\bar{x}$$

Y – Concentrations of the pollutants
X – Amount of released waste water
Correlation coefficient:

$$r = \frac{S_{xy}}{\sqrt{S_{XX}S_{yy}}},$$

$$S_{xx} = \sum_{i=1}^{n} (x_i - \bar{x})^2,$$

$$S_{XY} = \sum_{i=1}^{n} (x_i - \bar{x})(y_i - \bar{y})$$

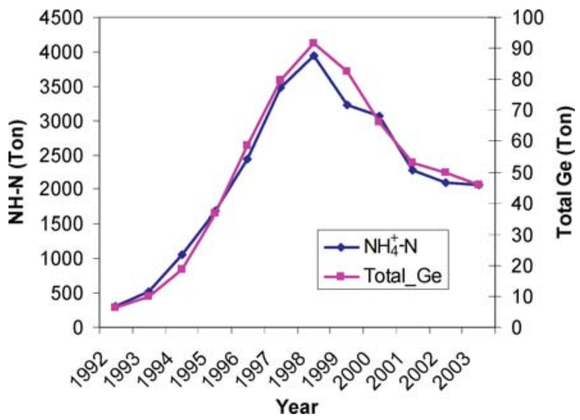

Figure 2. The load trends of ammonium nitrogen (NH_4^+-N) and germanium (Ge) from 1992 to 2003.

3. Results and Discussion

3.1. Results

3.1.1. *Changing Pattern of the NH-N, Ge and Released Waste Water*

Figure 2 shows the changing pattern of the ammonium nitrogen and total germanium. During the period of 1992 to 1998, the amount of NH_4^+-N increased with a slope of 651.6 and $r = 0.97$. The maximum amount of NH_4^+-N in 1998 reached 3952 tons. After 1998, the amount of NH_4^+-N in the released water started to decrease with a slope of -326.6 and $r = 0.87$. In 2003, the amount of NH_4^+-N decreased to 2069 tons, 47.6% decrease from 1998. Figure 2 also shows the change of the total germanium. From 1992 to 1998, the total germanium increased with a slope of 15.54 and $r = 0.96$. The maximum amount of total germanium was 91.9 tons in 1998. From 1998 to 2003, the amount of total germanium decreased with a slope of -8.98 and $r = 0.9$. In 2003, the amount decreased to 45.7 tons, a 50.2% decrease from 1998. These relationships are statistically significant at the significance level of 0.01. Although many factors could be attributed to the reduction of these two main pollutants, the actions from the local and provincial governments on environmental protection may have played a role in this reduction after 1998.

Figure 3 shows the relationship of ammonium nitrogen and germanium with released waste water. It is clear that the relationship between the ammonium and released waste water is linear, so does for germanium and waste water. Using annual pollutant loading and released waste water data, the amount of NH_4^+-N increased as waste water release increased. Y_1 (ton) $= 53.35 + 1.74 * X_1$ (ten thousand tons), where Y_1 is the amount of NH_4^+-N, and X_1 is the amount of released waste water from leather industry. The determination coefficient of this regression is 0.996 which means that more than 99% of the ammonium nitrogen was from the released waste water in leather industry in this region. The regression equation also indicated

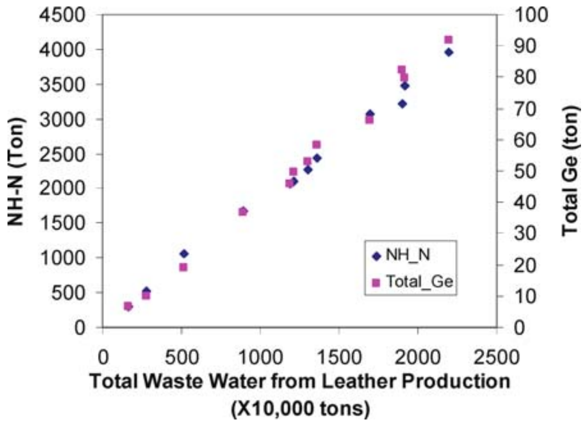

Figure 3. The relationship between the total waste water and ammonium nitrogen and germanium.

that the increase of releasing ten thousand tons of water, the amount of NH_4^+-N associated would be increased to 1.74 ton.

Similarly, the total germanium increased as the released waste water increased. Y_2 (ton) $= -1.95 + 0.0425 * X_2$ ten thousand tons), where Y_2 is the amount of total germanium and X_2 is the amount of released waste water from leather industry. This relationship indicated that total germanium is linearly associated with the released waste water. The determination coefficient for this regression is 0.994. When released waste water increased a ten thousand ton, the total germanium will increase the amount of 0.0425 ton.

3.1.2. *Waste Water and Integrated Pollution Index*

Figure 4 shows the relationship between integrated pollution index and the released waste water from leather industry. As the released waste water increases, the

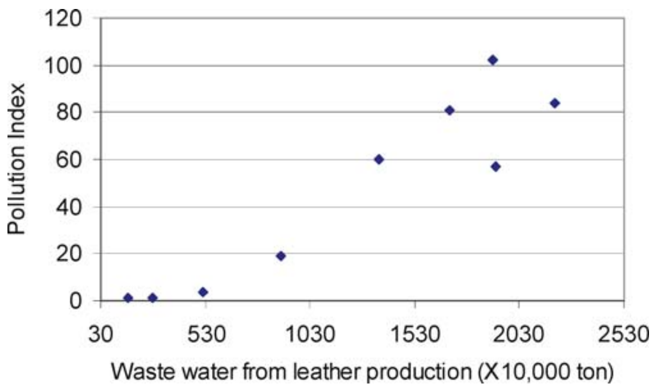

Figure 4. The relationship between the integrated pollution index and total waste water released in leather products production.

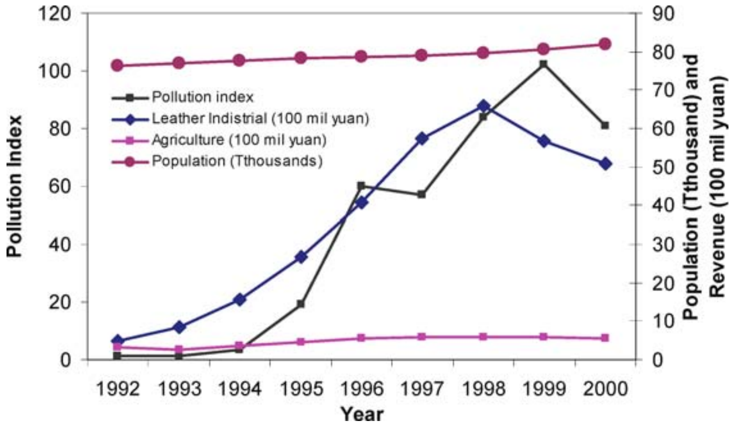

Figure 5. The relationship between the integrated pollution index and the economy of agriculture and industry and populations in Pingyang County, Zhejiang Province.

integrated pollution index also increased. There exists a linear relationship between the waste water and integrated pollution index. Thus, $P_j = -13 + 0.0482 * W$, where P_j is the integrated pollution index, and W is the released waste water from leather industry (ten thousand ton). This relationship is statistically significant, as the determination coefficient is 0.86, which indicates that 86% of the pollution was from the waste water. The linear relationship also illustrated that increasing every ten thousand tons of waste water release will lead to the increment of the integrated pollution index to 0.0482.

To further understand the integrated pollution index, Figure 5 shows the relationship of the pollution and the economics of agriculture and industry in the county where the watershed is located. During the period of 1992 to 2000, the population and the agricultural economics increased slightly, and the industrial value dramatically increased. However, the integrated pollution index increased from 1992 to 1996, a 4175% increase, decreased in 1997 about 5%, and then increased again in 1998 and 1999. After 1999, the integrated pollution index decreased 21%.

Table III shows the average exceeding index, its standard deviations and the coefficient of variations for the contaminants studied in the paper. These measures

TABLE III

Average exceeding index, its standard deviation, and coefficient of the variations for each pollutant from 1992 to 2000

	COD	Sedimentation	BOD	NH_4^+-N	Ge
Average	22	43	57	12	3
Standard deviation	2.13	1.51	3.18	0.59	0.16
CV%	9.7	3.5	5.6	4.9	6.2

were used to further evaluate the water quality in the watershed. The COD exceeded standard 22 times, sedimentations over 43 times, BOD 57 times, NH_4^+-N 12 times, and Ge 3 times (Table III). Their associated standard deviations were rather small. This may mean that the changes of these exceeding standards pollutants were small. Their associated coefficients of variations were as follows: 9.7% for COD, 3.5% for sediments, 5.6% for BOD, 4.9% for NH_4^+-N, and 6.2% for Ge respectively (Table III). From the data, one can see that the most severe pollutant was to the BOD. However, the most harmful pollutants to human are NH_4^+-N and Ge. As literature indicated, these two pollutants not only can harm human health, but also affect the aquatic species and ecosystem health (Bianchi *et al*., 1999; Radman *et al*., 2003; Wang and Ding, 2002). All the pollutants studied in this paper affect the water quality and the environment.

Following the single factor (National EPA, 2001) for determining water quality, the Aojiang watershed water quality dropped to Type IV or V in 2003 from the Type III in 1992. In 1997, one of the four profiles sampled for the monitoring still remained at Type III. This indicated that the water quality dramatically degraded in the last ten years in Aojiang watershed. When comparing with the Oujiang watershed that is near to the Aojiang watershed and with similar leather industry as the main local enterprises, we found that the integrated pollution index in Oujiang watershed also decreased since 1996 (Figure 6). The integrated pollution index in Oujiang watershed was negatively correlated with the released waste water. Although the leather industry is known to pollute the environment, large differences and opposite relationships of the integrated pollution index values can be found in Aojiang and Oujiang watersheds (Figure 6). The degraded water quality in Aojiang watershed not only has drawn the public concern, but also has attracted the attention from the local and provincial governments to consider the mitigation and litigation activities.

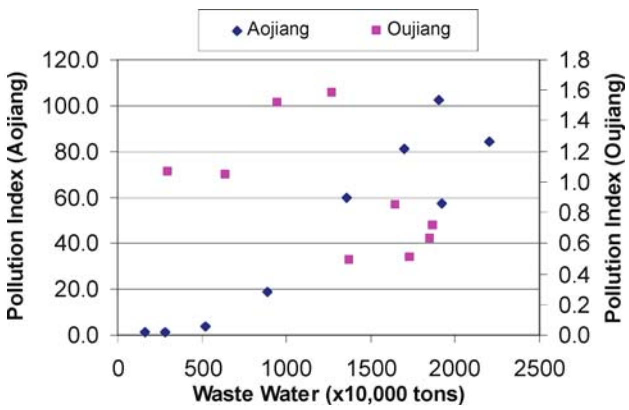

Figure 6. The relationship between integrated pollution index and waste water in Aojiang and Oujiang watersheds.

4. Discussion

In 1996, National Environmental Protection Agency (EPA) carried out the policy of "one control and two standards", and attacking "fifteen small" enterprises to dispose of poor processing enterprises (National EPA, 1997). This slogan of "one control and two standards" refers to controlling pollution and establishing the standards for surface water quality and waste water releases. These policy reinforcements have confirmed that controlling the production from the extreme small size, poor processing and managed enterprises could effectively reduce the waste water release from leather industry. These policies can also effectively reduce the release of pollutants. This may be one of the reasons why the loads of ammonium nitrogen and total germanium were reduced after 1998.

The results of decreasing trend integrated pollution index with the increased industry economy after 1999 illustrated that the leather industry associated pollutants could be reduced by using more advanced technologies and proper industry managements as well as implementing government regulations. Industries strictly following the guidelines of materials processing can also help reduce pollution. In addition, the proper investment in water treatment plants can effectively reduce the pollutants in released waste water. It is very clear that controlling the waste water release to the major river can improve the water quality in Aojiang watershed. This is especially important to control the release of NH_4^+-N and Ge from the areas where heavy leather industry is concentrated.

As literature well documented, high concentrations of nitrogen in drinking water can cause blue baby syndrome (Skipton and Hay, 1998). High nitrogen level can also cause river nitrification (Bianchi *et al*., 1999; Cébron *et al*., 2003) which led to degraded water quality and reduced the ecological biodiversities (Radman *et al*., 2003). High concentration of nitrogen can also reduce the blood oxygen transfer and destroy the tissues of respiratory system, which then leads to death by not being able to change oxygen and waste gas (Cooke and Kalita, 2002; Hubei Keliang Company, 2003).

As for the Ge toxicity, Wang and Ding (2002) considered that Ge^{3+} can fix protein and can harm human health. After intake of high concentrations of germanium, 0.015 to 0.033 mg/m^3, one can cause nose bleeds, voice loss, nose membranes shrinkage, even lung cancer (Wang and Ding, 2002), as well as harm fish species (Lide, 1998; Gao and Su, 2001; Lenntech, 2004). In addition, Li (2002) reported that the Ge^{3+} or Ge^{6+} could all form relatively stable compounds with ammonium, organic acids and proteins, which can adsorb to sediments. These sediments with the compounds then can contaminate crops and vegetables grown in these soils.

Although it is difficult to quantify the impact of ammonium nitrogen and germanium pollutants from the released waste water to human and ecosystem health, the potential long term effects of these pollutants to the human health and the ecological species in the watershed are foreseeable. According to the News Media Network (Xie, 2004), the "capital of leather industry" – Shuitou, the head water of the Aojiang watershed, Zhejiang province was named as one of the ten major environmental violation cases. Xie (2004) also pointed out that, in this region, the crops had little harvest; air quality was full of bad smell; the streams and rivers were full of blackish film floating on the water; and human health was severely impacted. The report also mentioned that the strange diseases frequently prevailed among the people in the region and fewer youngsters could pass physical exams for National Army admission. Due to the contamination of the head water, other towns in the Aojiang watershed down stream also suffered. This heavy impact of the pollutants forced many families to move out of the region (Xie, 2004).

Historically, the people in the Aojiang watershed, especially down streams, rely on fishing for living. Because of the advancement of science and technology, the tidal aquaculture activities and farm size near the bay area increased in the recent years. Up to this year, the farm size of aquaculture reached to 20 thousand Mu in Longgang town, and to one thousand Mu in Aojiang town (Wenzhou Ocean and Sea Bureau, 2003). The types of the fishery included crabs, shrimps, clams, and some fish types. These species require the water standard in Type III or better following GB3838-2002 (National EPA, 2001). If the water quality is less than the standard of Type III, the ecosystem health will be degraded and thus, the species numbers will automatically be reduced. Therefore, this degraded ecosystem and water quality will impact fishery economic development which then will influence people’s living standard in the region.

Following the current estimation, the annual waste water release in Aojiang watershed should be controlled to less than 2.9 million tons, which would be less than ten times of the current waste water release rate. If this goal can be achieved, then the water in the watershed can be maintained at the first level of waste water release standard, and the Type III surface water quality standard. In the same way, if the annual release of NH_4^+-N can be controlled to less than 43.5 tons, and total germanium annual release can be controlled to less than 4.35 tons, the waste water from the leather industry will meet the waste water release standards, GB8979-1996 (National EPA, 2001), the first level standard.

For improving the water quality in the watershed, it is critical and important to understand the needs of education on environmental protection, and understand the relationships between the environmental quality and economic development. Balancing these relationships can better foresee the long term and short term benefits. The deteriorating environment can affect the sustainable development of the economics in the region. Therefore, government agencies should work together with the local organizations to address the environmental quality issues in the watershed. Also the industries need to implement advanced technology and practical methods and managements to adjust for the environmental quality concerns.

Pollution prevention is often a better method than mitigation and litigation. It is also important for the local industry to improve resource use efficiency and to adjust the industry structure, and to promote “green” industry, and to control or reduce the pollution sources. Adjusting the leather industry in the total industry enterprise may be an effective way of reducing the nitrogen and germanium pollution in this watershed at the current conditions. Developing efficient water treatment plants can be another critical and effective means to protect the environment and the water in the watershed.

The analyses and the results from this study illustrated that the development of the leather industries in the Aojiang watershed was associated with sever pollution impact to human health and the degraded environment. Conventional models of depending on an economic impact estimate often lack the environmental linkages necessary for examining environmental stewardship and economic sustainability (Fenech *at al.*, 2003; Hansell *et al.*, 2003). In order to sustain the development of the economics, one should seriously consider the environmental protection when receiving the economic benefits and seriously bring economics and ecology together. A healthy environment can often lead to a harmony between the human and the ecosystems. Such an environment will lead people to be more productive and have a happier life to further generate capital by balancing economics and ecological sustainable development.

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