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**Spatial and temporal
variability of heavy
metals in streams**

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Spatial and temporal variability of heavy metals in streams of the Flint Creek and Flint River Watersheds from non-point sources

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Abstract

Throughout the United States, non-point pollution is responsible for large quantities of heavy metals entering bodies of water. Pollution as a result of heavy metals can impact drinking water supplies, recreation, fisheries, and aquatic species. Presence of heavy metals such as lead (Pb), cadmium (Cd), and chromium (Cr), in surface water may pose great risks to human health as well as to aquatic animals. In order to understand water quality changes due to heavy metal elements and pH as a result of spatial and temporal variability and land use/land cover changes, there is a need to monitor water bodies on a constant basis. The purpose of this investigation was to assess the impacts of spatial and temporal variability on heavy metals and pH as a result of land use/land cover changes and provide a baseline for future water quality study from non-point sources in two watersheds. Spatial and temporal variability factors were not significant for all the heavy metal elements. Significant water quality changes occurred between 2003 and 2004 for the two of the five heavy metals (Pb, and Ni) and pH. However, this was not true for the other of heavy metals investigated (Cd, Cr, and Zn). There was no influence of watershed observed for any of the heavy metals and pH in this study. To accurately quantify environmental impacts of heavy metals as well as pH, land use changes, and natural processes leading to spatial and temporal variability of water quality variables, continuous monitoring of surface water is necessary to improve the water quality of these watersheds.

1 Introduction

There is considerable interest in protecting water quality in the streams and rivers at the watershed level. Integral to this concept is the need to continuously assess and monitor water bodies on a regular basis for heavy metals from non-point sources (NPS). Due to the complex and dynamic changes that occur in a water environment on a constant basis, it is often difficult to explain the spatial and temporal variations in water

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quality within the specific watershed. These variations may arise from several source including: topography, soil type (texture, pH), human activities, the amount and volume of runoffs, and climatic conditions (dry, wet season) within the watershed. The Flint Creek (FC) and Flint River (FR) watersheds, located in the Wheeler Lake basin in north Alabama, have been the focus for improvement of water quality for a number of special interest groups in recent years. The major sources of waste discharge in the watershed are from six communities and a number of public and private facilities in the area. About 36,000 people inhabit the area. The Flint River watershed project was initiated in January 1993 by the Environmental Protection Agency (EPA), Alabama Department of Environmental Management (ADEM), the United States Department of Agriculture (USDA-NRCS), and The Tennessee Valley Authority (TVA) as a cooperative effort among Federal, State and local organizations to improve and protect the water quality of the FC watershed. The multi-year cooperative effort was to document trends and improvements in water quality that result from implementation of best management practices. The FR is an important recreational and scenic resource; a 55 km section of the river is a popular canoe and tubing area and was designated a canoe trail by the Madison County Commission in 1993. The FR watershed is also home to many rare plants and animals. Increased anthropogenic inputs of heavy metals in soils have caused considerable concern relative to their effect on water contamination (Zhang et al., 2003). This study was undertaken to examine the impacts of spatial and temporal variability as well as land use/land cover changes on heavy metals in these two watersheds due to non-point source pollution. A significant drop in water quality at the FC and FR watersheds due to NPS pollutions can impact drinking water supplies, recreation, fisheries, and aquatic species. The presence of heavy metals such as lead (Pb), cadmium (Cd), and chromium (Cr), in surface water may pose great risks to human health as well as to aquatic animals. The objective of this investigation was to evaluate the impacts of spatial and temporal variability on heavy metals and pH as a result of land use/land cover changes and to provide a basis for future water quality study in two north Alabama watersheds from non-point sources.

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1.1 Materials and methods

1.2 Project background and approaches

The study areas chosen for this project are FC and FR watersheds, which are located in the Wheeler Lake basin in north Alabama. In north Alabama, FC Watershed has been identified as a target for special efforts to improve water quality. The FC watershed (Fig. 1) encompasses approximately 117 441 hectares in three counties: Morgan, Lawrence and Cullman. The primary land-use within the watershed is agriculture, with an over whelming majority being pasture and forest. Cullman and Morgan counties are two of the major agricultural counties in the state. The FC Watershed is a major tributary to Wheeler Lake on the Tennessee River.

The FR watershed (Fig. 1) includes approximately 147 151 hectares including half of Madison County (USGS, 2002). The Flint River watershed starts in Lincoln County, Tennessee, and is bounded by the mountains on the east border of Madison County, and drains into the Tennessee River. The watershed is primarily agricultural land in northern Alabama and south-central Tennessee (USGS, 2002). Urban and residential land uses represent less than 1%, but a growing part of land use in the watershed is residential growth from the City of Huntsville, Alabama, which spreads northward and eastward into the watershed.

1.3 Sampling, processing, and analysis

Three permanent sampling locations were established in each watershed and georeferenced using a Leica GS 50 GPS unit, for repeated measurement. Each sampling location was selected to represent maximum drainage area for each location. During each field visit, water samples were collected in 1-L LDPE (low density polyethylene) sample containers for analyses of heavy metals. All water samples were placed in a cooler with ice and transported to the laboratory for analysis. In the laboratory samples were acidified to pH2.0 with concentrated sulfuric acid at a rate of 2 ml/L of sample

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for storage in the refrigerator until analyses for heavy metals could be run. The water samples were collected from these sites twice per month in order to analyze the heavy metals. Measurements for pH were determined in situ during each sampling event using a Model 6600EDS-SV Clean Sweep water quality meter with new YSI 650 MDS Multi-parameter display system (YSI, Yellow Springs, OH). Standard methods and techniques found in the Standard Methods for the Examination of Water and Wastewater (Greenberg et al., 2000) was used for analysis of heavy metals. The water samples for heavy metals were analyzed with a plasma 400 Inductively Coupled Spectrophotometer (Perkin Elmer, Wellesley, MA). Data from all sampling locations were identified by year, watershed, and location. An analysis of variance (ANOVA) was performed for each indicator variable: cadmium, chromium, nickel, lead, zinc, and pH. Years (2003 and 2004), watersheds (FC and FR), and locations (one through six) were treated as independent factors. All analyses were performed using the general linear model (GLM) procedure of statistical analysis systems (SAS) version 8.2 (SAS Institute, 2001).

1.4 Land use/land cover within the watersheds

The Landsat 7 ETM⁺ image used in this study was acquired on 7 July 2002. The image was geometrically and radiometrically corrected and registered to the Universal Transverse Mercator (UTM) coordinate system using the NAD 83 datum and nearest neighboring re-sampling method. Careful selection of about 40 well-defined and well-distributed ground control points (GCPs) resulted in positional accuracy of root mean square error (RMSE) equal to or smaller than ± 0.25 pixels. The maximum likelihood classification algorithm using the ERDAS Imagine 8.7 image processing software was used to classify the land use/cover map of the two watersheds. An average accuracy of 86% and an overall accuracy of 83% were achieved with a Kappa coefficient of 0.79 using the supervised classification algorithm. The classification results are presented in Figures 2 (FC) and 3 (FR), respectively. Forest occupies the largest amount of land distribution covering over 57 411 and 49 465 hectares in the FC and FR watersheds,

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respectively (Table 1). Agriculture and pasture are the two next predominant land cover usages in both watersheds. Their percentages are about 23.12% and 17.68% for FC and for the FR watershed the percentages were 28.34%, and 22.47%, respectively. Water and commercial land cover constitute least area for both watersheds and the percentages of the land area covered are about 1.7%, and 3.04% for FC and 1.02%, and 4.88% for FR watersheds, respectively. It is important to note that all the different land use categories presented in Table 1, have the potential to impact water quality in these two watersheds.

2 Results and discussion

2.1 Rainfall within the watersheds

Monthly rainfall data during 2003 and 2004 for the two watersheds are shown in Figs. 4 through 7. Annual average precipitation for 2003 for the FC and FR watersheds were 1102 mm and 1162 mm, respectively. Annual average precipitation for 2004 for the FC and FR watersheds were 1135 mm and 1256 mm, respectively. The annual average rainfall data indicated that the 2004 monitoring period had slightly higher annual average rainfall precipitation (5%) for FC watershed compared to 2003 monitoring period. Similarly, the 2004 monitoring period is wetter and had slightly higher annual average rainfall precipitation (9%) compared to 2003 monitoring period for the FR watershed. Precipitation and seasonal changes resulting in hydrologic or in physicochemical water conditions may have influenced heavy metal transport and stability in the waters; and may play important role in the availability of some of the heavy metals elements. In order to accurately assess environmental impacts of spatial and temporal variability as a result of land use/land cover on heavy metals and other water quality variables, a continuous monitoring of surface water may help in attaining the desired water quality goals. Precipitation data were from the National Weather Service Forecast Office database, Huntsville, Alabama. Stream flow data are from US Geological Survey

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(USGS) Gage stations in north Alabama.

Mean spatial water quality parameters for heavy metals and pH at the Flint Creek and Flint River watersheds during the 2003 and 2004 monitoring periods at different sampling locations is presented in Table 2. The mean comparison among water samples collected for heavy metals from all sampling locations indicated that the highest mean concentration of Pb was observed at the FC watershed at location one with the value of 0.70 mg L^{-1} ; and this was significant ($P < 0.05$) compared to location two of the same watershed. However, this was not the case with the other locations ($P > 0.05$). The highest mean concentration of Cr was observed again at the FC watershed at location three with the mean value of 0.152 mg L^{-1} ; and this was not significant ($P > 0.05$) compared to the rest of the locations. The highest mean concentration of Cd was observed at the FR watershed at location four with the mean value of 0.008 mg L^{-1} ; and this was significant ($P < 0.05$) compared to locations one (0.005 mg L^{-1}) and six (0.004 mg L^{-1}) which were located in the FC and FR watersheds, respectively. Even though, the highest mean concentration of Ni was observed at the FC watershed at location three with the mean value of 0.055 mg L^{-1} ; and this was not significantly different than other locations. Again, the highest mean concentration of Zn was observed at the FC watershed at location one with the mean value of 0.138 mg L^{-1} ; and this was not significant ($P > 0.05$) compared to the rest of the locations. The highest mean pH level was observed at the FR watershed at location five (7.67 SU), this was significant ($P < 0.05$) compared to locations one, two, three, and six. However, the mean pH level at location five was not significantly different compared to location four.

Location did not show a significant effect on most of the heavy metals elements, except for Pb and Cd ($P < 0.05$) (Table 2). This study demonstrated that heavy metal concentrations did not vary significantly among locations during this study. Thus, spatial variability did not play a significant role for all of the heavy metals in our present study.

The mean concentrations for Pb and Ni were significantly higher during the 2004 monitoring period compared to the 2003 monitoring period (Fig. 8). However, no in-

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creases were seen for the other heavy metal elements. The mean pH level was also significantly higher during the 2004 monitoring period compared to the 2003 monitoring period (Fig. 8). Again, temporal variability did not play a significant role for all of the heavy metals. No significant differences were observed when the mean concentrations of heavy metal and pH levels were compared by watershed ($P>0.05$).

Although, the 2004 monitoring period had the higher mean concentrations for most of the heavy metals elements as well as the pH (Tables 3 and 4), it is evident that these increases were not significant for all cases. As shown in Table 2, there was no effect of locations on all heavy metals throughout the watersheds. Thus the null hypothesis that the means are equal at all locations and between years is rejected in both cases. Again no significant differences were observed when the means for heavy metal elements and pH were compared by watersheds.

Some of the differences in mean heavy metal as well as pH levels resulting from spatial and temporal variations may have been influenced by differences associated with land use, human activities, stream flow/volume, and other internal stream conditions at the time of sampling. Changes in hydrologic conditions, stream pH, or internal water conditions may have also influenced the availability or absence of heavy metals in these two watersheds. Transport and stability of heavy metals may also be dependent on other factors in water environment. These results of heavy metal concentrations at these watersheds highlight the need for continuous sampling under similar conditions and illustrate a potential use of models in helping to design and coordinate sampling.

3 Conclusions

Statistical analysis of the data from the two watersheds revealed significant increases in mean concentrations for Pb and Ni during the 2004 monitoring period compared to the 2003 monitoring period. However, no such increases were seen for the rest of the heavy metal elements (Cd, Cr, and Zn). Significant increase in mean pH level was also seen during 2004 in comparison to 2003 monitoring period. No significant dif-

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ferences were observed when the mean concentrations of heavy metal and pH levels were compared by watershed. This study also demonstrated that location did not show a significant effect on all metals. The role of pH in the availability or absence of heavy metals in the streams was not apparent in this study. Rainfall data for this study were similar for both watersheds for both years. The slightly higher rainfall that was seen during the 2004 monitoring period may have also affected some of the heavy metal elements and pH levels in these two watersheds as a result of pollutant wash offs into the receiving streams of these two watersheds. The results of this study yielded some intriguing observations for the effects of spatial and temporal variability particularly on heavy metal concentrations in these two watersheds. Seasonal differences in dynamic nature of water flow, hydrology of sampling sites, internal water conditions, and transportation rate of heavy metals for each sampling event may have played important role for this intriguing observation in this study. It is important to note that this study was performed under natural conditions where sampling is on a bi-weekly basis, whether runoff happens to be there or not. The findings of this investigation also highlight a continuous need of monitoring these two watersheds in order to see the interactive effects of spatial and temporal variations at these two watersheds. This study supports the importance of dissemination of educational programs at local and federal levels that will address best management practices that will reduce non-point source pollutions in these two watersheds and elsewhere.

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Table 1. Land use/land cover characteristics at the Flint Creek and Flint River watersheds for 2002.

Watershed	Land Use/Land Cover	Hectare (ha)	Percentage
Flint Creek	Water	1972	1.71
	Forest	57411	49.67
	Pasture	20444	17.68
	Residential	5528	4.78
	Agriculture	26724	23.12
	Commercial	3509	3.04
Total		115 588	100
Flint River	Water	1518	1.02
	Forest	49465	33.07
	Pasture	33619	22.47
	Residential	15286	10.22
	Agriculture	42393	28.34
	Commercial	7310	4.88
Total		149 591	100

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Table 2. Mean spatial heavy metals and pH comparison at the Flint Creek (FC) and Flint River watersheds (FR).

	Pb	Cr	Cd	Ni	Zn	pH
	mg/L					SU
FC						
Location 1	0.700 ^a	0.137 ^a	0.005 ^b	0.050 ^a	0.138 ^a	6.92 ^b
Location 2	0.218 ^b	0.10 ^a	0.006 ^{ab}	0.049 ^a	0.060 ^a	6.91 ^b
Location 3	0.395 ^{ab}	0.152 ^a	0.005 ^{ab}	0.055 ^a	0.052 ^a	7.03 ^b
FR						
Location 4	0.420 ^{ab}	0.047 ^a	0.008 ^a	0.045 ^a	0.069 ^a	7.18 ^{ab}
Location 5	0.341 ^{ab}	0.038 ^a	0.006 ^{ab}	0.037 ^a	0.049 ^a	7.67 ^a
Location 6	0.358 ^{ab}	0.035 ^a	0.004 ^b	0.035 ^a	0.057 ^a	6.97 ^b

Within column, means followed by a same letter are not significantly different at 5% level of probability.

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Table 3. Statistical data collected during the study period for the Flint Creek (FC) and Flint River (FR) watersheds, for lead (Pb), cadmium (Cd), and chromium (Cr).

Site ID	WS	n	Pb			Cd			Cr		
			mg L ⁻¹			mg L ⁻¹			mg L ⁻¹		
			Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
2003											
Site 1	FC	15	0.000	0.001	0.000	0.000	0.008	0.003	0.004	0.075	0.003
Site 2	FC	15	0.000	1.396	0.098	0.001	0.015	0.005	0.005	0.260	0.005
Site 3	FC	15	0.000	0.423	0.028	0.000	0.011	0.004	0.008	0.065	0.004
All sites	FC	45	0.000	1.396	0.042	0.000	0.015	0.004	0.004	0.260	0.004
Site 4	FR	15	0.000	1.888	0.279	0.000	0.051	0.008	0.005	0.193	0.008
Site 5	FR	15	0.000	2.646	0.200	0.001	0.009	0.006	0.003	0.073	0.006
Site 6	FR	15	0.000	4.518	0.325	0.000	0.006	0.003	0.007	0.130	0.003
All sites	FR	45	0.000	4.518	0.268	0.000	0.051	0.005	0.003	0.193	0.005
2004											
Site 1	FC	15	0.000	6.204	1.401	0.000	0.009	0.006	0.008	2.414	0.238
Site 2	FC	15	0.000	2.150	0.338	0.004	0.010	0.007	0.013	1.635	0.145
Site 3	FC	15	0.000	1.998	0.762	0.003	0.010	0.006	0.008	3.543	0.267
All sites	FC	45	0.000	6.204	0.833	0.000	0.010	0.006	0.008	3.543	0.216
Site 4	FR	15	0.000	1.888	0.562	0.003	0.029	0.007	0.001	0.009	0.036
Site 5	FR	15	0.000	1.916	0.482	0.003	0.009	0.005	0.006	0.125	0.041
Site 6	FR	15	0.000	1.837	0.391	0.000	0.008	0.005	0.007	0.104	0.035
All sites	FR	45	0.000	1.916	0.478	0.000	0.029	0.005	0.001	0.125	0.037



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Table 4. Statistical data collected during the study period for the Flint Creek (FC) and Flint River (FR) watersheds, for Nickel (Ni), Zinc (Zn), and pH.

Site ID	WS	n	Ni			Zn			pH		
			mg L ⁻¹			mg L ⁻¹			SU		
			Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
2003											
Site 1	FC	15	0.000	0.045	0.019	0.014	0.092	0.035	3.29	8.06	6.41
Site 2	FC	15	0.000	0.068	0.023	0.004	0.203	0.047	3.12	8.33	6.34
Site 3	FC	15	0.000	0.042	0.013	0.013	0.091	0.042	3.17	8.14	6.37
All sites	FC	45	0.000	0.068	0.055	0.004	0.203	0.041	3.12	8.33	6.37
Site 4	FR	15	0.006	0.091	0.042	0.012	0.193	0.065	3.60	8.29	6.42
Site 5	FR	15	0.000	0.069	0.024	0.005	0.129	0.048	6.18	8.29	7.25
Site 6	FR	15	0.000	0.059	0.025	0.005	0.232	0.047	3.73	8.16	6.06
All sites	FR	45	0.000	0.091	0.030	0.005	0.232	0.053	3.73	8.29	6.57
Site 1	FC	15	0.029	0.186	0.081	0.008	2.414	0.239	6.42	8.07	7.43
Site 2	FC	15	0.026	0.337	0.075	0.027	0.184	0.070	6.12	8.83	7.49
Site 3	FC	15	0.017	0.682	0.097	0.022	0.111	0.059	7.08	7.99	7.68
All sites	FC	45	0.017	0.682	0.084	0.008	2.414	0.122	6.12	8.83	7.53
Site 4	FR	15	0.000	0.069	0.047	0.014	0.259	0.083	7.37	8.37	7.95
Site 5	FR	15	0.003	0.089	0.050	0.012	0.083	0.048	7.24	8.90	8.09
Site 6	FR	15	0.021	0.065	0.045	0.020	0.114	0.061	7.25	8.08	7.89
All sites	FR	45	0.000	0.089	0.047	0.012	0.259	0.064	7.24	8.90	7.97

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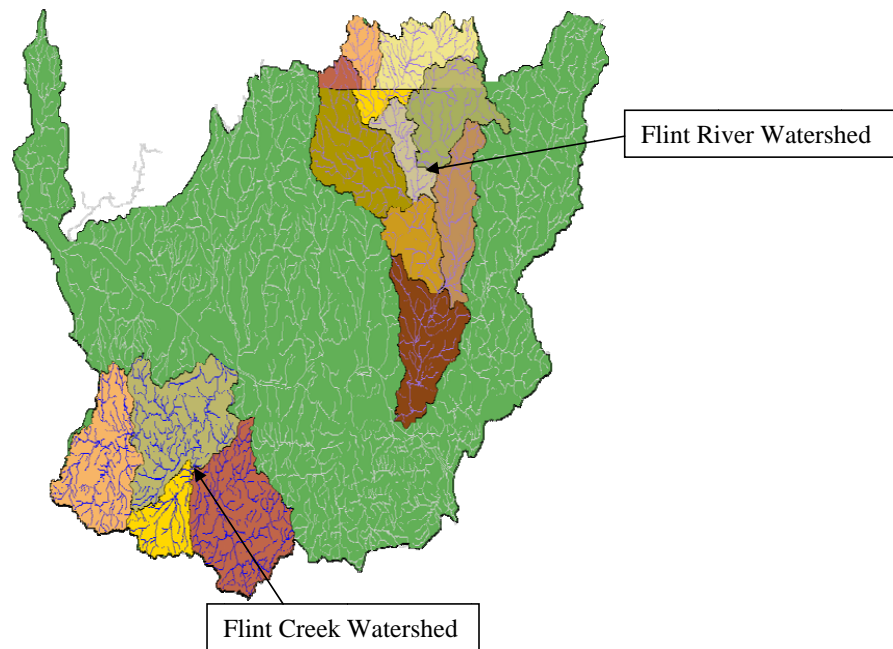


Fig. 1. Map of Wheeler Lake basin showing the Flint Creek and Flint River watersheds.

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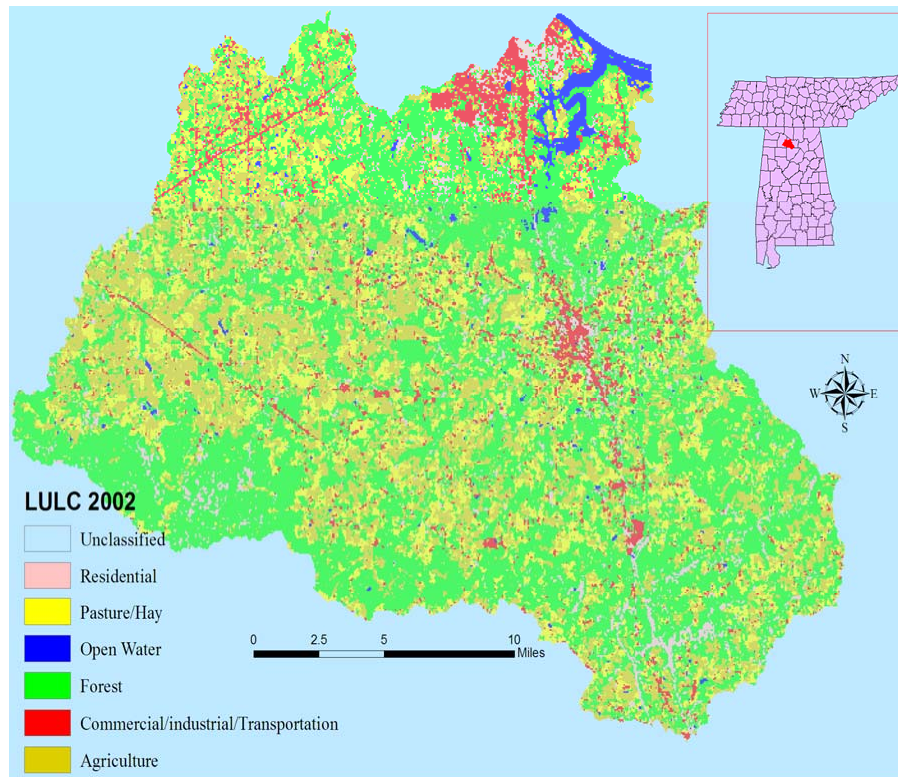


Fig. 2. Land use classification map of the Flint Creek watershed for the year 2002.

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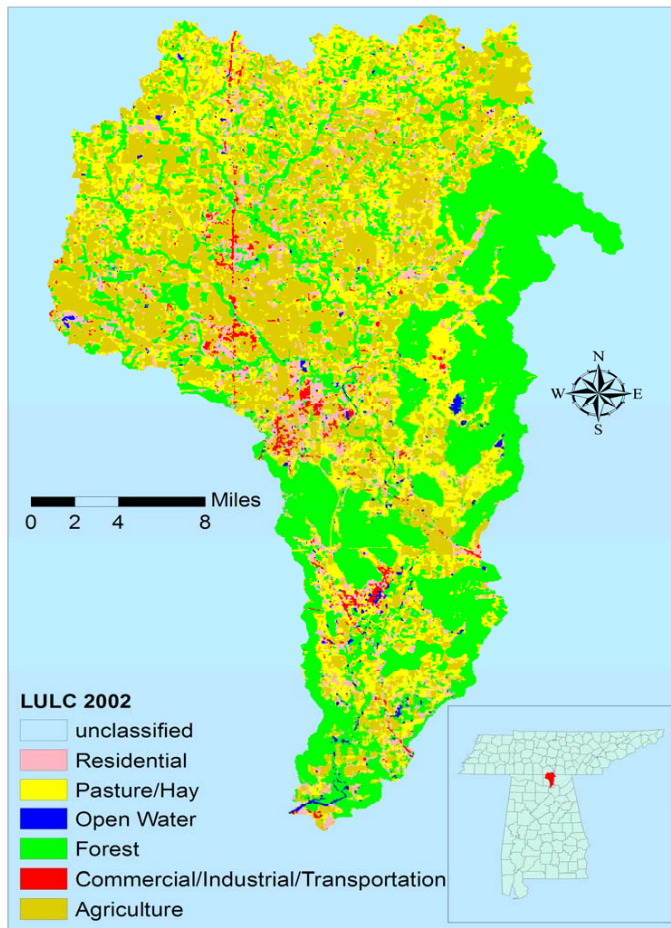


Fig. 3. Land use classification map of the Flint River watershed for the year 2002.

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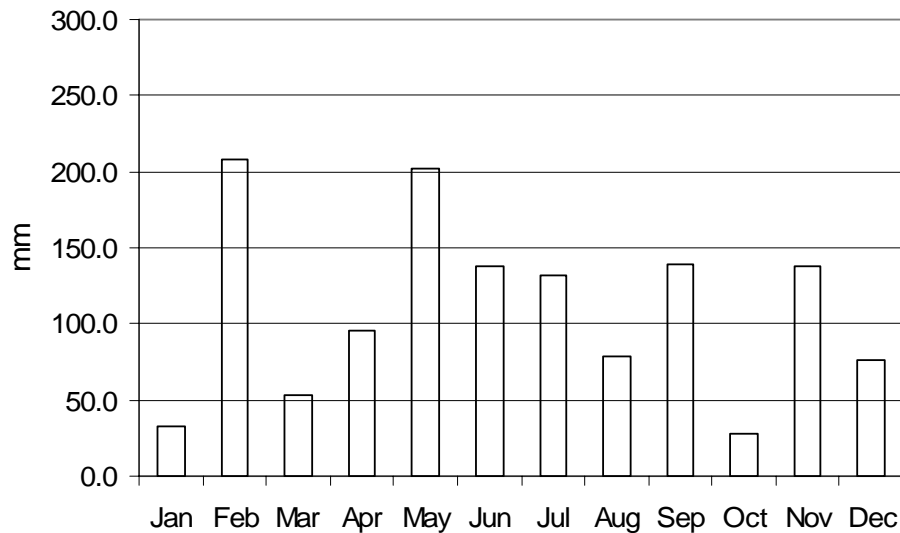


Fig. 4. Monthly rainfall data at the Flint Creek watershed for 2003.

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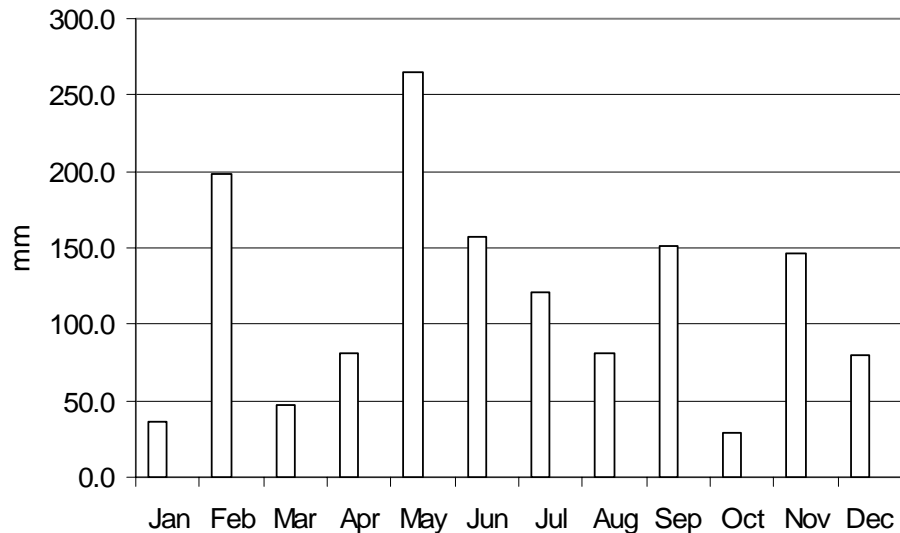


Fig. 5. Monthly rainfall data at the Flint River watershed for 2003.

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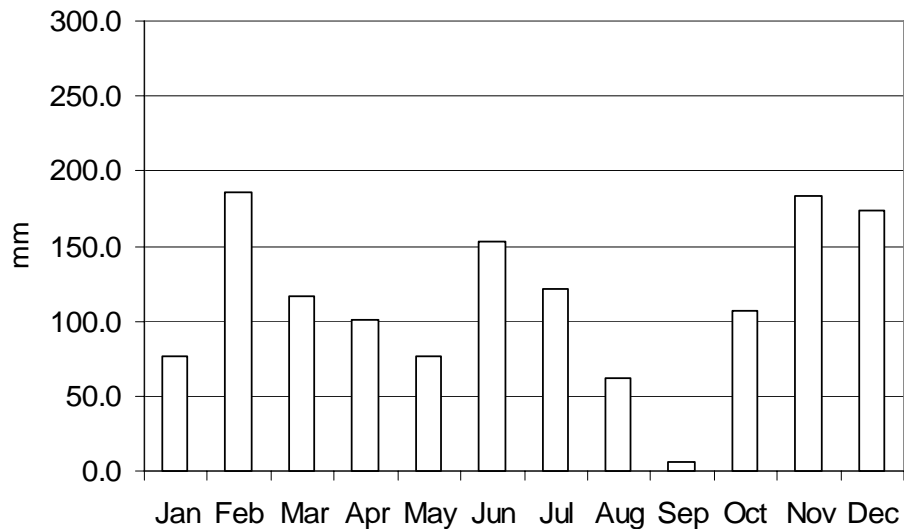


Fig. 6. Monthly rainfall data at the Flint Creek watershed for 2004.

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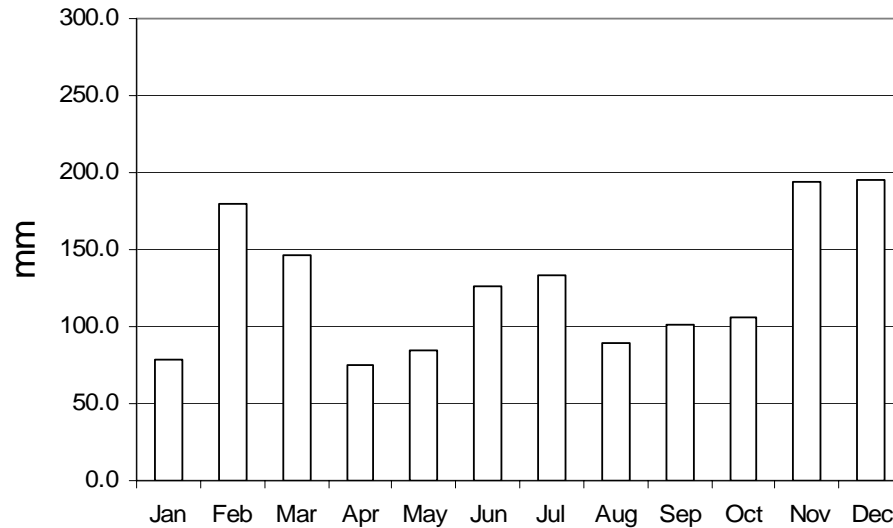


Fig. 7. Monthly rainfall data at the Flint River watershed for 2004.

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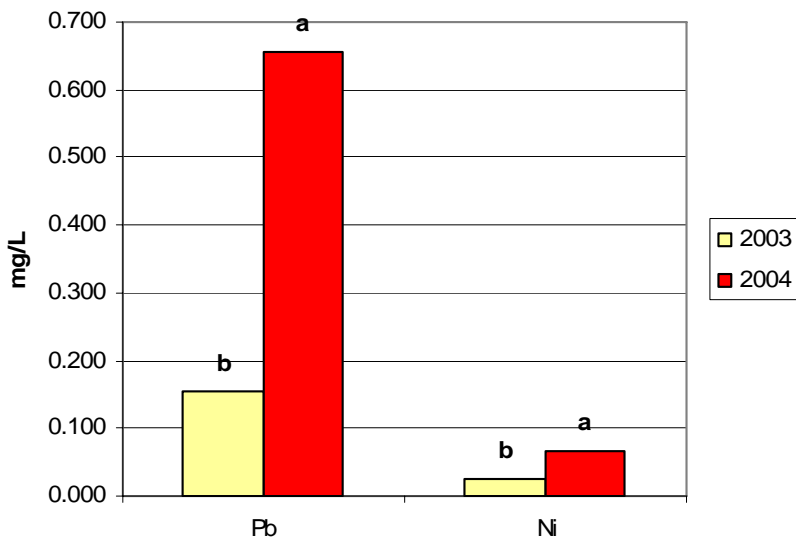


Fig. 8. Effect of year on the average heavy metals during the 2003 and 2004 monitoring period.

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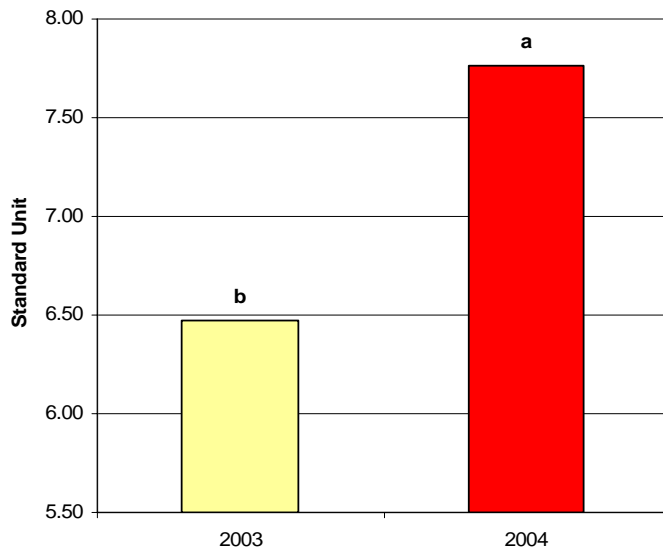


Fig. 9. Mean pH levels observed during 2003 and 2004 monitoring period.

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