

ARSENIC AND ARSENIC SPECIES IN THE RIO GRANDE,
AND THE EFFECT ON IRRIGATED LANDS

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TECHNICAL COMPLETION REPORT

Account Number 01345693

February 2002

New Mexico Water Resources Research Institute

in cooperation with the

Department of Earth and Environmental Science
New Mexico Tech

The research on which this report is based was financed in part by the U.S. Department of the Interior, Geological Survey, through the New Mexico Water Resources Research Institute.

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ACKNOWLEDGMENTS

This research work could not have been completed without the assistance of several individuals. Special thanks to Greg Miller for his assistance in writing the original proposal. Patti Fritsch conducted field sampling and assisted with instructing additional field personnel in field and laboratory technique. Additional field samplers included Karen Karen and Reyna Abeyta. Reyna also made sure that the chemical analysis was completed during the last part of the project. Special thanks to the New Mexico Bureau of Mines Water Laboratory managed by Lynn Brandvold and Terry Thomas. Finally, thank you to Ron Wielkopolski who assisted with creating the figures and tables for this report.

ABSTRACT

The Rio Grande was used for centuries and continues to be used for irrigation in New Mexico. In the Middle Rio Grande Valley arsenic concentrations of between 2 and 16 ppb have been reported in river water. Of concern is build up of arsenic in irrigated fields. To determine if arsenic is being adsorbed in the fields, irrigation water and drain water were measured weekly. The chemical behavior and mass balance of arsenic were evaluated to determine changes in the arsenic flux on a spatial and time basis. Waters were sampled and tested for arsenic species arsenite (3^+), and arsenate (5^+). Arsenic levels in waters used for irrigation had arsenate values as high as 13.4 ppb. For the drain, arsenite values as high as 14.3 ppb were obtained. We estimate that greater than 80 percent of the arsenic in irrigation waters remains in the irrigated fields. Preliminary analysis of irrigated soils indicates they contain 1.6 to 2.7 ppm labile arsenic, which is consistent with a century of arsenic accumulation. A major implication is that irrigated soils in the Middle Rio Grande Valley are an arsenic time bomb that could catastrophically release their arsenic in the future.

Keywords: arsenic, irrigation, water quality, monitoring

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INTRODUCTION

The Middle Rio Grande Valley has been irrigated for centuries using water from the Rio Grande. Arsenic concentrations within the Rio Grande have been reported as between 2 to 16 parts per billion (ppb) (Chapin and Dunbar, 1995; Norman unpublished analyses). Most rivers and streams in the U.S. contain < 1 ppb arsenic concentrations (Lettenmaier et al 1991). The higher concentrations of arsenic in surface water are generally associated with geothermal areas, mineralized zones, or young volcanic sediments in other regions of the U.S.(Welch et al 2000). The Rio Grande Valley of central New Mexico shares these characteristics. Geologically recent volcanic activity is responsible for the presence of young volcanic sediments and contributes to some of the geothermal activity. However, many of the mineralized zones and geothermal areas in the region are due to regional groundwater flow systems (Witcher 1995). The Rio Grande accumulates arsenic as it transverses these areas in central New Mexico. Arsenic is found in water and sediments as inorganic species arsenite As (III), arsenate As(V), and as the organic compounds monomethylarsonic acid (MMA), and dimethylarsinic acid (DMA). Arsenite is 10 times more toxic than arsenate, which is about 1000 times more toxic than the methylated arsenic species.

Environmental pathways are complex due to the large number of interactions and transformations that arsenic undergoes (National Academy of Sciences 1977). The general arsenic cycle involves not only natural sources but also manmade sources including fertilizers, pesticides, and other industrial chemical compounds. The purpose of this study was to understand the environmental pathways that arsenic follows in the irrigation system. On a weekly basis, arsenic concentrations in the inflow conveyances of the irrigation system and in the drains were evaluated in order to estimate the amount, if any, arsenic that would accumulate within the irrigated fields.

Objectives

The objectives of the project were met; however there are a number of important follow-up studies that could be conducted based on this research. This research was considered a baseline study in order to obtain data for a specific period. The follow on studies would assist in understanding the questions that were raised from this work. The primary objective was to sample the inflow and drainage areas on a weekly basis and to analyze the water samples for arsenic concentrations to determine if there was a variability in the concentrations throughout the year. A secondary objective was to differentiate between the inorganic arsenic species, arsenite and arsenate concentrations. In addition to the arsenic concentrations, bulk water chemistry was analyzed to evaluate the overall chemical system. Daily variations in the arsenic concentrations for both the inflow and drainage were also studied by conducting

three diurnal sampling events. Based on the data obtained, the mass balance for arsenic within the irrigation system was developed.

Project Location

San Acacia is located approximately 75 kilometers south of Albuquerque, New Mexico and was chosen as the sampling site. Figure 1 presents an aerial photograph of the San Acacia area and the relationship between the sampling locations and the Rio Grande and irrigated fields. Two inflow water sampling points were used since the water did not flow in the inflow channels between October 14, 2000 and March 10, 2001, since irrigation of the fields was stopped for the winter. During this period, water samples representing the inflow were collected directly from the river. Water samples were collected from two different drain sampling locations as well. The first drain sampling site was to the south of the I-25 off ramp to San Acacia. On September 10, 2000, the sampling location was moved upstream since the original sampling site became inaccessible due to reconstruction of a barbed wire fence.

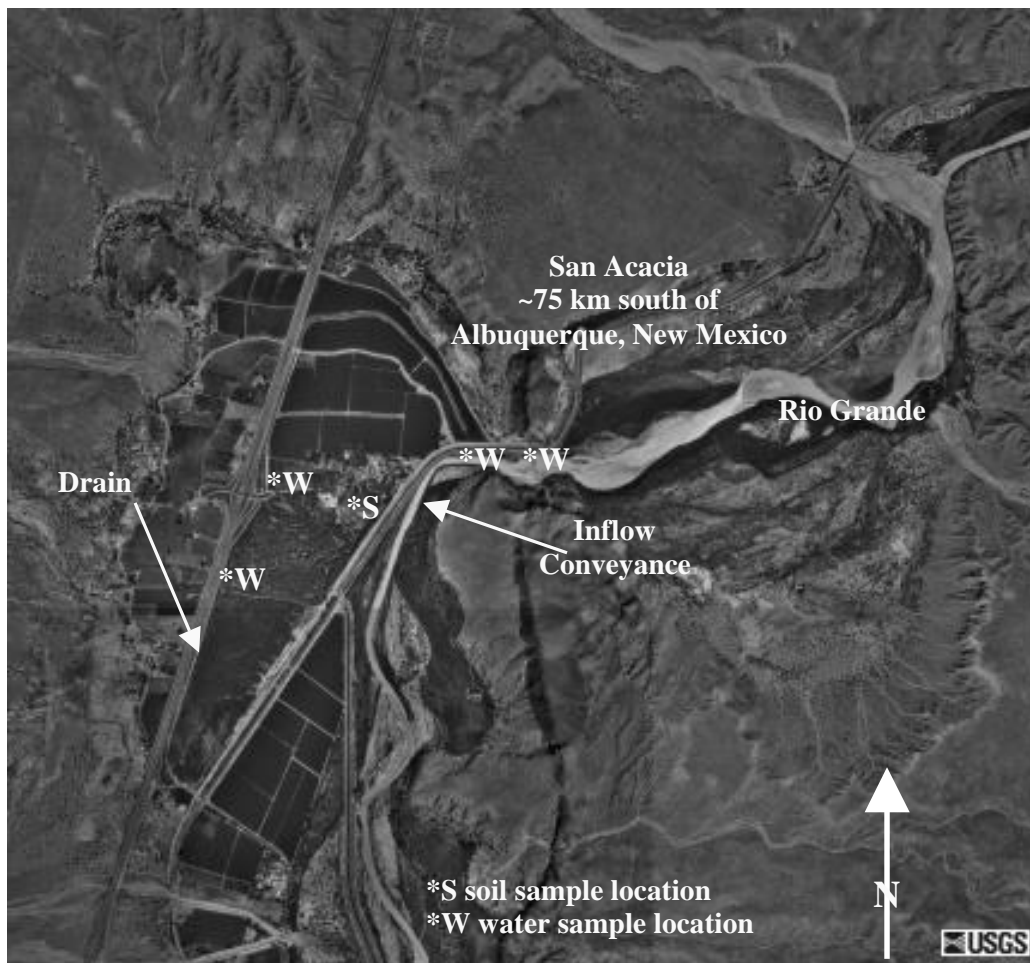


FIGURE 1. Aerial photograph of sampling locations. The Rio Grande is located on the eastern side of Interstate I-25 and the irrigated fields (USGS 1999).

METHODS

The project required both a set field regime and consistent laboratory methods. The field regime was rigorously adhered to since variability in sampling methods could potentially corrupt the data obtained. Laboratory methods were supervised by the senior chemist for the New Mexico Bureau of Mines water chemistry laboratory. Sampling events occurred weekly from March 3, 2000 to May 25, 2001.

Six water samples were collected at each sampling location weekly in order to determine arsenic concentrations, speciation, and bulk water chemistry. Samples were tested for the following:

- 1) S –total dissolved solids/total suspended solids, 500 ml bottle
- 2) T –total arsenic – filtered, acidified, 125 ml bottle
- 3) C –cations – unfiltered, acidified, 125 ml bottle
- 4) A –anions – unfiltered, no acid, 125 ml bottle
- 5) As (III) –arsenic species 3^+ , 60 ml bottle
- 6) As (V) –arsenic species 5^+ , 60 ml bottle.

Samples were collected using a sampling beaker cleaned with river water. The beaker was lowered into the river/irrigation drain and approximately 750 ml of sample was obtained. The water was poured into the S sample container first, followed by the T, C, and A sample containers. Additional water or these samples were obtained from the sampling beaker. The beaker was not cleaned between the collection of these samples except for flushing with river water. The T sample was returned to the laboratory for filtering through a 0.45-micron filter. Acidified samples were obtained by using ultra pure HNO_3 and acidifying the sample to a pH of between 2 to 3. The acid was applied to the samples immediately after collection for the unfiltered samples and after filtration for the T samples.

Speciation was conducted in the field as soon as the water samples were collected. A constructed, validated acetate resin column was used to separate the arsenic species. A 25-ml sample of acidified water was filtered through a 0.45-micron filter into the resin column. The water drained through the resin column into a 60 ml bottle labeled for As (III). After the collection of this water, 25 ml of 0.12 M HCl was drained through the resin column into the same bottle. The bottle was capped after all of the acid had drained into the bottle. Using the same resin column, 50 ml of 0.12 M HCl was drained through the column into a prelabeled As (V) bottle.

Samples were labeled according to the inflow (SI) or drain (SD), the date and time and the type of sample using the lettering scheme presented above. For example, a SI01160114-C is a sample collected from the San Acacia inflow on January 16, 2001 at 1400 hours. The C sample would be a 125 milliliter (ml) bottle, unfiltered and acidified. The sample was used for cation analysis and included total arsenic in unfiltered water.

Eh, pH, conductivity, temperature, and alkalinity were measured from the sampling beaker. Eh, pH, and conductivity were measured using hand-held meters according to standard methods. Alkalinity measurements were obtained by using a color sensitive solution to estimate the alkalinity. The sampling beaker, meters and other reusable tools were rinse with RO water prior to collection of the next sample. The sampling beaker and other tools were rinsed with the sampling water prior to the collection of the next sample. General field conditions were noted in the logbook.

Laboratory work included the use of graphite furnace and flame atomic absorption, and ion chromatography in order to determine the concentrations of arsenic, major anions and cations, and metals within the samples. Total dissolved solids and total suspended solids were also determined for each sample. Appendix A presents tables of all the data obtained. A Varian graphite furnace was used to obtain arsenic concentrations. The furnace was generally calibrated prior to each run to 1 part per billion (ppb) detection limit. In some instances this was difficult to obtain and a 3 ppb, detection limit was set.

Duplicate samples were created from both the inflow and drain samples for all tests. In general, the duplicate values ranged from about three to eight percent of each other. Bromine, fluorine, and phosphorous had larger ranges. In addition to the duplicate samples, calibration samples were spaced throughout a test run in order to estimate the machines variability. In general, the calibration samples were within a percent of standards used. When a large variation occurred (greater than 3 percent), the machines were recalibrated and the samples were retested.

The acetate resin columns were created in the laboratory and were validated prior to use. Validation consisted of draining a prepared As (III) solution through the column as described for the field procedures and draining a prepared As (V) solution through a separate column as described for the field procedures. Ten, 10-ml samples were obtained from each column and analyzed using the graphite furnace atomic absorption.

Two soil samples collected were analyzed using nitric acid digestion and the graphite furnace to determine the arsenic concentrations. Three separate samples were analyzed for each soil sample collected.

DISCUSSION

Eighty-two inflow samples and seventy-three drainage samples were collected over the course of 14 months. Arsenic species, arsenite and arsenate, as well as total arsenic, cations, anions, and bulk water chemistry were analyzed. Table 1 presents the summary statistics for these analyses. It can be seen that the mean concentrations in the inflow water, for arsenite was 1.00 ppb and for arsenate, 1.87 ppb. Values as high as 4.2 ppb and 13.4 ppb, respectively, were obtained for the inflow. For the drain, the mean concentration of arsenite was 1.91 ppb and for arsenate, 1.04 ppb. Values as high as 14.3 ppb and 4.16 ppb were obtained.

Bulk water chemistry indicated that the pH from the inflow to the drain was slightly lower and the Eh was relatively the same. The pH values indicate slightly alkaline waters from a mean of 8.26 for the inflow to 7.98 in the drain with a maximum of 9.2. If these waters were plotted on a Eh/pH diagram, they lie on the boundary between arsenate and arsenite (Wilson and Hawkins 1978). The arsenite is more stable in waters that are slightly more neutral. The inflow water had a mean temperature of 16.5 °C, while the drain water was at an average temperature of 19.1 °C. The higher temperature is related to the drain channels being shallower and therefore the water was likely to heat up. The cooler waters were located in the deeper channels of the inflow conveyance channels. Alkalinity was higher in the drain at 255.08 parts per million (ppm) versus 173.56 ppm for the inflow. The total dissolved solids were about three times as high in the drain as oppose to the inflow. The total suspended solids were about 3.5 times higher in the inflow compared to the drain. This is also reflected in the conductivity measurements with about twice the value in inflow versus the drain.

TABLE 1. Summary Statistics for Analysis performed on Inflow and Drainage Samples

<i>INFLOW</i>	<i>pH</i>	<i>Eh</i>	<i>Temp</i>	<i>Cond</i>	<i>Alkalinity</i>	<i>TDS</i>	<i>TSS</i>	<i>As3+</i>	<i>As5+</i>	<i>As C</i>	<i>As T</i>
		mV	C	μS	ppm	ppm	ppm	ppb	ppb	ppb	ppb
Mean	8.26	128.41	16.48	557.23	173.56	561.34	205.80	1.00	1.87	1.91	3.07
Median	8.40	115.00	17.00	561.50	176.00	396.00	158.00	0.58	1.85	2.04	2.40
Stan. Dev.	0.99	51.82	7.14	116.67	41.02	499.12	206.50	1.10	1.88	0.64	1.92
Range	9.20	280.00	27.60	760.00	304.00	2290.00	1139.00	4.20	13.40	2.59	11.20
Minimum	0.00	0.00	0.00	0.00	0.00	87.00	4.00	0.00	0.00	0.73	0.00
Maximum	9.20	280.00	27.60	760.00	304.00	2377.00	1143.00	4.20	13.40	3.32	11.20

<i>DRAIN</i>	<i>pH</i>	<i>Eh</i>	<i>Temp</i>	<i>Cond</i>	<i>Alkalinity</i>	<i>TDS</i>	<i>TSS</i>	<i>As3+</i>	<i>As5+</i>	<i>As C</i>	<i>As T</i>
		mV	C	μS	ppm	ppm	ppm	ppb	ppb	ppb	ppb
Mean	7.98	125.02	19.13	247.58	255.08	1498.63	63.34	1.91	1.04	0.75	1.06
Median	8.10	115.00	18.60	255.00	256.00	1656.50	45.00	1.00	0.72	0.77	0.89
Stan. Dev.	1.01	61.83	6.89	60.94	58.35	430.08	67.34	2.78	1.08	0.40	0.72
Range	9.20	300.00	33.00	568.00	336.00	1546.00	316.00	14.30	4.16	1.53	2.80
Minimum	0.00	0.00	0.00	0.00	0.00	297.00	0.00	0.00	0.00	0.00	0.00
Maximum	9.20	300.00	33.00	568.00	336.00	1843.00	316.00	14.30	4.16	1.53	2.80

<i>INFLOW</i>	<i>Ca</i>	<i>Mg</i>	<i>Na</i>	<i>K</i>	<i>Fe</i>	<i>Mn</i>	<i>Cl</i>	<i>SO4</i>	<i>NO3</i>	<i>F</i>	<i>PO4</i>
	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
Mean	63.44	12.19	54.68	5.50	10.08	40.05	46.69	109.54	0.12	0.38	0.06
Median	67.04	10.25	58.65	5.84	0.09	0.04	35.00	101.68	0.09	0.41	0.02
Stan. Dev.	17.02	15.53	18.65	1.69	15.39	52.28	55.07	69.74	0.21	0.17	0.11
Range	86.34	88.29	90.49	6.94	40.08	121.13	395.10	598.78	1.57	1.00	0.50
Minimum	0.00	0.00	0.00	0.00	0.00	0.00	20.38	0.14	0.00	0.00	0.00
Maximum	86.34	88.29	90.49	6.94	40.08	121.13	415.48	598.92	1.57	1.00	0.50

<i>DRAIN</i>	<i>Ca</i>	<i>Mg</i>	<i>Na</i>	<i>K</i>	<i>Fe</i>	<i>Mn</i>	<i>Cl</i>	<i>SO4</i>	<i>NO3</i>	<i>F</i>	<i>PO4</i>
	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
Mean	177.79	41.54	340.01	10.49	0.08	21.78	297.97	467.79	0.23	0.52	0.74
Median	181.23	42.94	344.21	10.55	0.04	0.12	329.35	540.25	0.28	0.66	0.02
Stan. Dev.	35.04	8.96	24.41	0.73	0.10	108.28	127.32	181.38	0.13	0.26	2.22
Range	150.19	47.61	96.72	2.87	0.36	541.53	439.27	666.36	0.41	0.76	9.40
Minimum	103.40	0.00	292.98	9.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Maximum	253.59	47.61	389.70	11.90	0.36	541.53	439.27	666.36	0.41	0.76	9.40

Figures 2 and 3 present the arsenic variations throughout the sampling period of March 3, 2000 to May 25, 2001, for both the inflow and drain, respectively. There is a variation in both the inflow and drain in the concentrations of total arsenic, and in the species, arsenite and arsenate.

For the inflow, the lowest concentrations appear to occur in the summer from about May to August. This is the period of intensive irrigation. The concentrations of the species also vary, with little to no arsenite occurring in the spring and in the winter months. In the drain, the lowest concentrations occur in May 2000, August, and December. Both species occur throughout the year. There appears to be an anomalous result in July 17, 2000. There are also high concentration of arsenic in April and May 2001. The inflow values are low, generally below 3 ppb. This high out flux of arsenic in the drain can either be caused by the use of fertilizers on the soils releasing a greater amount of arsenic in the drain, an introduction of pesticides containing arsenic, or some other unidentified cause.

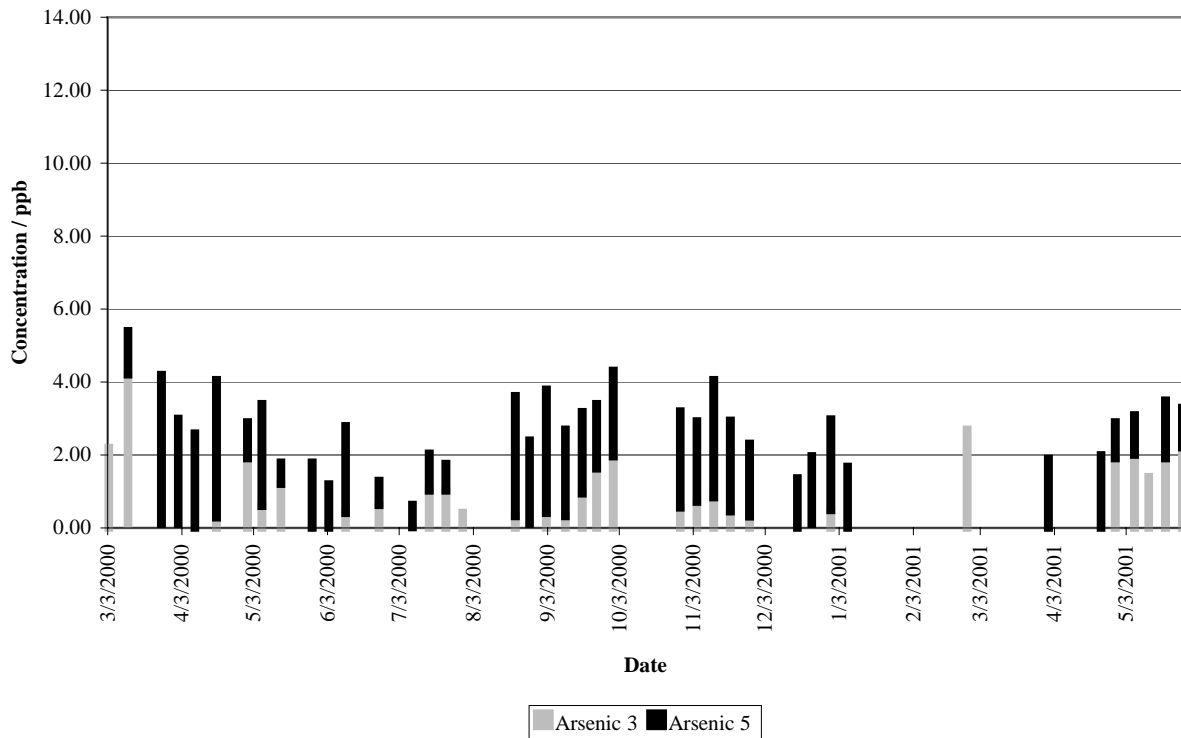


FIGURE 2. Arsenic concentrations in the inflow water samples throughout the sampling period.

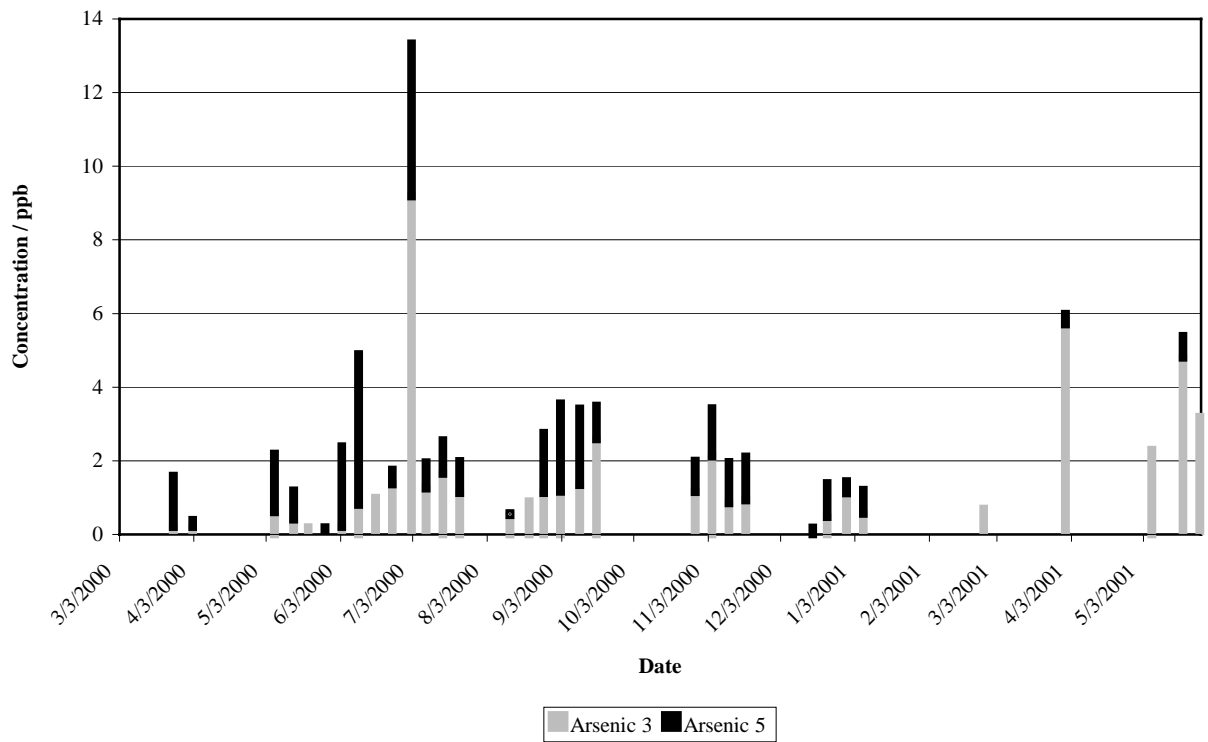


FIGURE 3. Arsenic concentrations in the drain channel water samples throughout the sampling period.

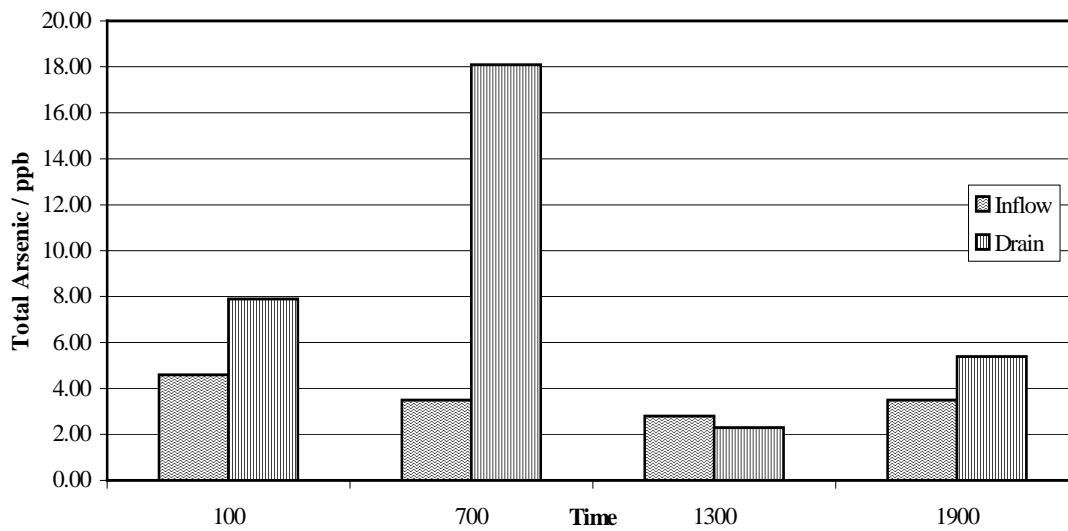


FIGURE 4. Daily variations in the arsenic concentrations.

The daily variation in the arsenic concentrations can be seen in Figure 4. The highest concentrations occurred around midnight and early in the morning in the inflow water samples. The late afternoon to evening samples had lower concentrations in both the inflow and drain.

The cations and anions average concentrations increase from the inflow to the drain as shown in Figure 5. This increase from the inflow to the drain in concentrations ranges from about seven times for chloride (Cl⁻) ion and six times for sodium (Na⁺) to only about 1.5 times for potassium (K⁺). Generally, the ion concentrations increase about three to four times. There is also a high concentration in sulfate (SO₄²⁻). Total dissolved solids also are increased about 3 times from the inflow to the drain. Arsenite and arsenate concentrations for the inflow versus the drain are presented in Figure 6. The arsenite concentration increased about two fold. The arsenate concentration decreased slightly from 1.87 ppb to 1.04 ppb.

The increase of the ion concentrations can be related to two different mechanisms either acting separately or in conjunction. First, the evaporation of the inflow water will tend to concentrate the ions. Secondly, the ion concentration increase may be due to release of ions from the soils into the drain. The arsenate ion concentration remains about equivalent or slightly decreases. This indicates that the arsenate remains in the soil column or is taken up in the vegetation.

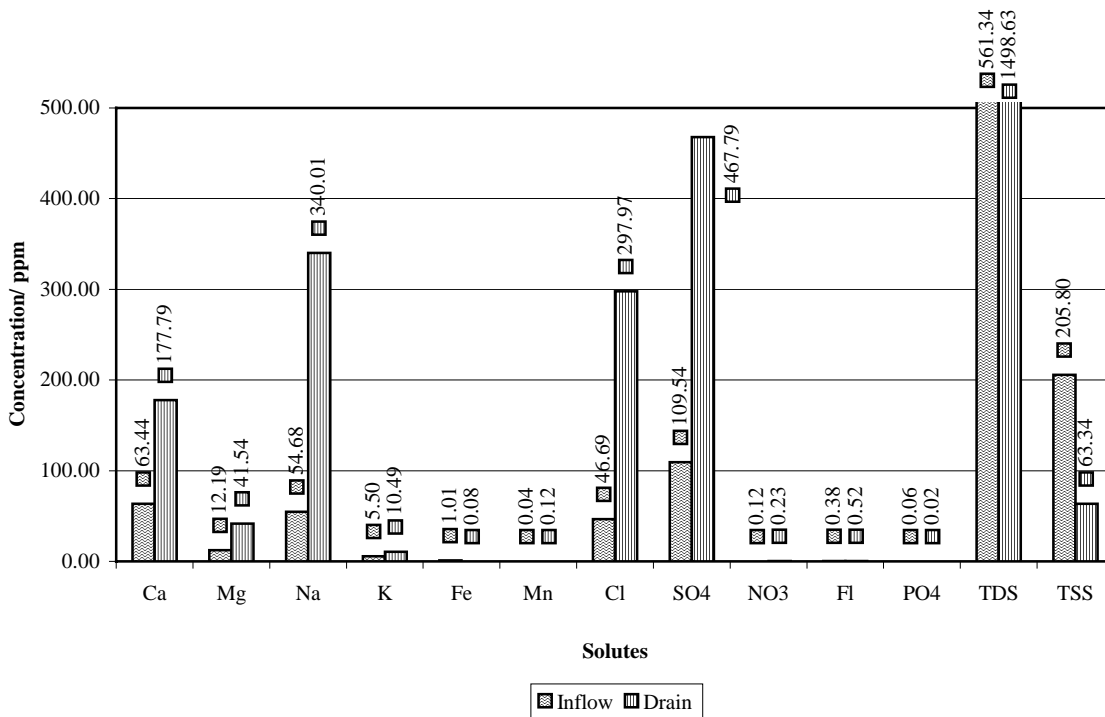


FIGURE 5. Mean concentrations of cations and anions for the inflow and drain.

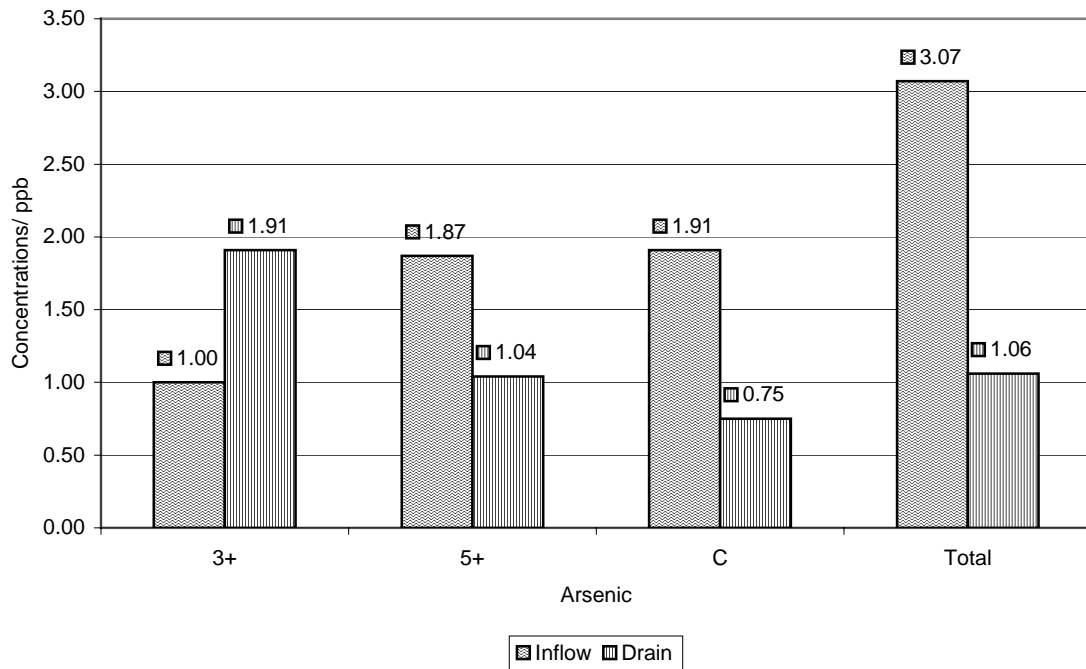
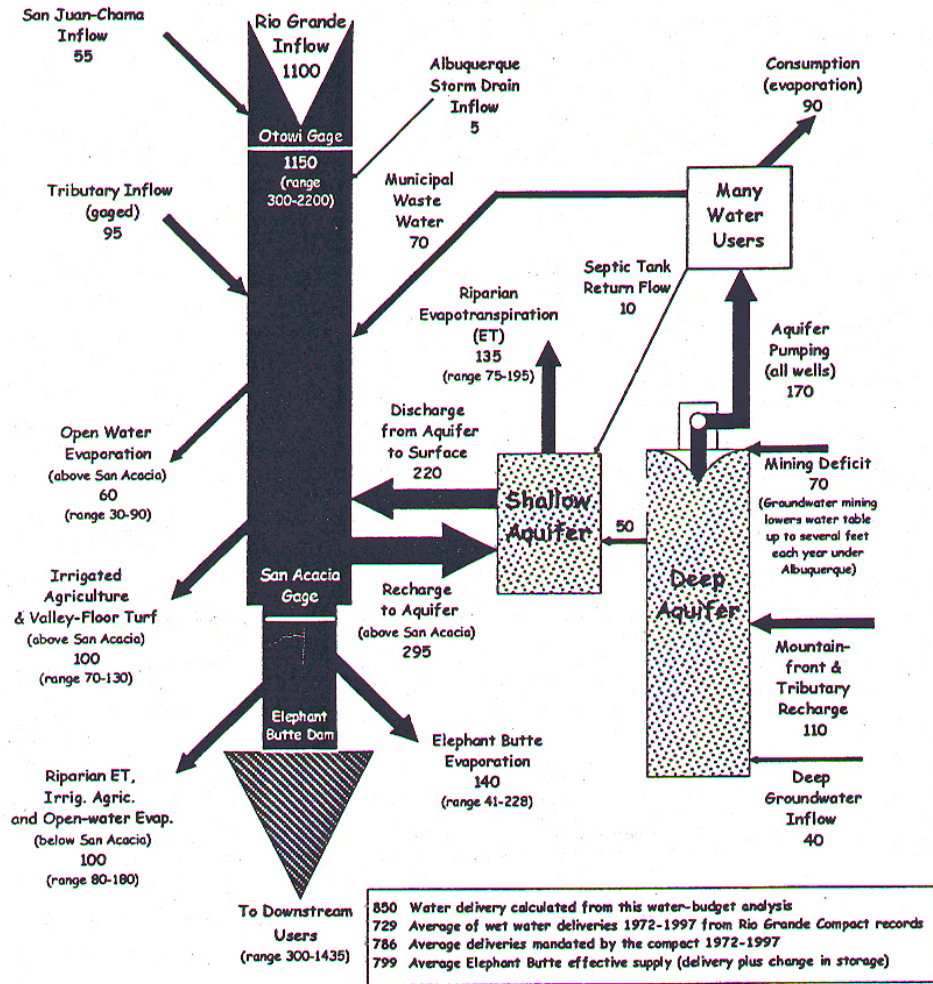


FIGURE 6. Mean concentrations for arsenic species and totals in the inflow and drain.

Figure 7 presents the Middle Rio Grande water budget based on average values for 1972 to 1997 (Middle Rio Grande Water Assembly 1999). Irrigated agriculture and open-water evaporation and riparian evapotranspiration below San Acacia averages about 100 units out of 1100 units of inflow or about 10 percent of the inflow into the area. It can range as high as about 18 percent. About 20 percent of the inflow is recharged into the aquifers. Above San Acacia, approximately 10 to 15 percent of the inflow is applied to irrigation and valley-floor turf and a little over 10 to 20 percent is observed as riparian evapotranspiration. The increase in the sodium and chlorine contents correspond to an increase in the salt contents, which as well as the evaporation rates in the water budget suggest evaporation as a mechanism for concentration.

Soil samples were obtained after preliminary results were determined suggesting that arsenic concentrations did not increase in the drain waters. Preliminary analysis of irrigated soils indicates they contain 1.6 to 2.7 parts per million (ppm) labile arsenic. These high arsenic values within the soils are consistent with arsenic remaining in the soils and accumulating over the centuries of irrigation of these fields.

MIDDLE RIO GRANDE WATER BUDGET
(Annual Values Typical for 1972-1997)



Values are annual average (rounded). Natural variability is large for most. Some but not all variabilities are shown.

Line widths are drawn in relative scale of magnitude
Action Committee of the Middle Rio Grande Water Assembly
10/7/99 Version

FIGURE 7. Middle Rio Grande water budget.

CONCLUSIONS AND RECOMMENDATIONS

Conclusions reached from this research meet the objectives of understanding the environmental pathway of arsenic in a surface water/irrigation scenario. Arsenic within this irrigation system is brought in from the inflow waters and is applied to the fields. Once the water is on the fields, evaporation operates to concentrate major anions and cations within the water. The pH and the Eh of the waters remain relatively the same and there does not appear to be a radical shift in the acidity or oxidizing potential of the water. Therefore, evaporation appears to be the main mechanism for concentrating the ions. As the water flows through the soils, arsenic remains within the soil column. Arsenate appears to be the primary species that remains within the soils. Arsenite is the more toxic of the inorganic species and is in higher concentrations in the drain.

Arsenic species vary in concentration throughout the year and there appears to be a diurnal variation. The arsenic species also do not seem to vary at the same rate or same time throughout the sampling period. Arsenic concentrations in the inflow water are generally higher in March through April, and September through October. Anomalous concentrations in the drain occur in July, March and May, which may correspond to application of fertilizers or insecticides into the soils or may be related to another cause. The arsenic concentrations in the drain decrease in December through beginning of March, when the fields are not being used and irrigation is not used.

Preliminary analysis of irrigated soils indicates they contain 1.6 to 2.7 ppm labile arsenic. We estimate that greater than 80 percent of the arsenic in irrigation waters remains in irrigated fields. This is consistent with a century of arsenic accumulation. A major implication is that irrigated soils in the Rio Grande Valley are an arsenic time bomb that could catastrophically release their arsenic in the future. Arsenic can be released from iron surfaces by mildly acid solutions, or replaced by another ion that is preferably sorbed (Pierce and Moore, 1980; Livesey and Huang, 1981; Wauchope and McDowell, 1984; Voigt, et al., 1996). The phosphate ion replaces the arsenate ion on iron oxide surfaces hence an application of phosphate fertilizer could result in a massive release of arsenic. When oxide surfaces become saturated, arsenic break out will occur, which may also release waters with high arsenic concentrations. This may explain the seasonal variation observed in the drain where high concentrations were observed in March and May as well as July, which is generally the growing season and associated with the use of fertilizers.

Additional Research

Additional research is needed to further refine conclusions about how much arsenic is within the soil column and how it may be released. This baseline study provided information on the potential for arsenic variation and accumulation within the soil column. Soil studies should be conducted to measure the amount of arsenic within the soils. As part of a follow-up study, samples at different levels within the soil column should be collected to estimate what soil horizons accumulate the arsenic. Additional water sampling should be collected to evaluate the variability that is observed and the anomalous concentrations observed in specific months. Farmers should be interviewed to evaluate the application of fertilizers and pesticides to the fields. More diurnal sampling should also be conducted to verify the variation in arsenic concentrations throughout the day. A detailed Eh/pH diagram should be developed to aid in understanding the behavior of the arsenic species within the waters.

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APPENDIX A
DATA TABLES