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Accumulation of Arsenic in Sediments of Lakes Treated with Sodium Arsenite

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■ Sodium arsenite, an aquatic herbicide, has been extensively used in Wisconsin lakes for the control of excessive macrophyte growths. Sediments in several of these treated lakes were investigated for patterns of arsenic accumulation with depth and relationships to iron, manganese, calcium, and organic carbon content. All sediments had substantial accumulations of arsenic, but no predominant correlation was found with other measured components. In these surficial sediments, arsenic appears to be in a nonoccluded form with highly organic sediments being more readily leached of arsenic than highly calcareous sediments.

One of the frequent problems of lakes which receive large amounts of aquatic plant nutrients is the growth of attached algae and macrophytes to the degree where they seriously impair the recreational potential of these lakes. A variety of chemicals have been used for controlling this growth, one of which has been sodium arsenite. The use of sodium arsenite as an aquatic herbicide was extensive through the mid-1960's (1), especially in the state of Wisconsin. In a 20-year span, beginning in 1950, 741 495 kg of arsenic were applied to Wisconsin public waters in various weed control programs (2). Usage prior to 1950 is also known to have been extensive, but few records were kept as to exact amounts. By 1970, sodium arsenite application to public waters was ended in Wisconsin, principally because of its potential accumulation in lake sediments and the ready availability of organic herbicides (2).

This paper reports the results on a study of the arsenic accumulation in sediments of lakes which had received substantial quantities of sodium arsenite. At a time when this herbicide was still being extensively used, Mackenthun (3) stated that knowledge of the depth to which arsenic may penetrate the sediments of treated lakes was missing, and to date it has not been published elsewhere. Additionally within the elapsed interval, there has been an increase in the concern over arsenic concentrations in surface waters from sources such as smelters (4, 5), coal-fired power plants (6), and even detergents (7). For the lakes investigated, the primary source of concern is the arsenic deliberately added to the waters.

Characteristics of Lakes Investigated

The study lakes are located within the southwestern quadrant of the state of Wisconsin. All of the lakes have areas of extensive growth of rooted aquatic plants, both submergent and emergent. Physical details of the lakes as well as arsenic loadings are presented in Table I.

Cores were collected with an 8.9-cm (i.d.) piston-coring device (8). The cores collected were 0.5–1 m in length and were generally taken in the deeper areas of the lake. Where additional cores were obtained, sampling sites were selected from inshore areas of contrasting character. Certain sites were also sampled with an Ekman dredge, while water samples were collected with a Van Dorn sampler.

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Analytical Methods

Each core was extruded in the field and sectioned into 5-cm intervals and placed in plastic bags. The sections were air dried in petri dishes, and portions were ground to less than 60 mesh size for analysis. Aliquots were dried overnight at 105 °C prior to analysis.

Sample decomposition for P, Fe, Mn, Ca, and Mg followed the procedures outlined by Mackereth (9), incorporating HF pretreatment followed by HNO₃–HClO₄ digestion. Phosphorus was measured by the vanadomolybdate method of Jackson (10), and the metals by atomic absorption on a Perkin-Elmer Model 303. Total carbon was determined with a Model 589-400 LECO analyzer equipped with a thermal conductivity detector. Iron chips and Sn granules (LECO) were used to ignite the sediment.

Carbonate carbon was calculated from Ca and Mg determinations by conversion to carbonate equivalents. Bortleson (8) has discussed the appropriateness of this approach for Wisconsin lake sediments containing 30–80% CaCO₃. Organic carbon represents the difference between total carbon and carbonate equivalents.

Decomposition of sediment samples for arsenic determinations utilized sodium hydroxide fusion (11, 12) in nickel crucibles. Approximately 0.25 g of sediment, as a maximum, was fused with 1.5 g of NaOH in 30-mL crucibles. The sediment–hydroxide mixture was first slurried with a few milliliters of distilled water and dried on a hot plate prior to fusion at 500 °C for 20 min. The melt was leached with distilled water and passed through a fiberglass filter (Reeve-Angel 934AH) into a specially modified 25 × 300 mm test tube fitted with a stopcock at the bottom and a side-arm for vacuum connection. Final sample volume was 30 mL. Arsenic was determined by the silver diethyldithiocarbamate procedure (13).

Arsenic recoveries of spiked calcareous samples averaged 95 ± 7%. Because at least one core of the study, i.e., BCL III, was substantially composed of organic matter, the fusion procedure was compared to extended HNO₃ digestion (12) for this material. Comparative recoveries were substantially lower with the fusion procedure, averaging 80.5% of the acid digest recoveries. Arsenic values for the BCL III core reported were obtained using the acid digestion procedure.

Results

Core analyses are presented in Tables II–V, and arsenic profiles are shown in Figures 1–3. Correlation coefficients between arsenic and other measured parameters are tabulated in Table VI.

Arsenic profiles show similarities in accumulation based on both chemical composition and lake morphology. In lakes having deep basins, such as Big Cedar Lake (BCL) and Pewaukee Lake (PwL), arsenic accumulation is at the immediate surface and may be largely a function of sedimentation rate. In core BCL I (Figure 3) the chemical transition which is often associated with the onset of the postcultural period (8) is at the 30–35-cm interval, but the arsenic rise is extremely sharp from the 5–10-cm section to the surface. A sedimentation rate of approximately 5 mm/yr, as that found by Bortleson (8) for productive lakes in southern Wisconsin, would account for this profile. In core PwL I (Figure 1), the chemical transition is just discernible at the 40–45-cm interval, with the arsenic rise beginning at the 15–20-cm section. Pewaukee Lake began receiving chemical treatment in 1946

Table I. Physical Parameters of the Study Lakes

lake	surface area (ha)	mean depth (m)	max depth (m)	vol (km ³)	drainage area (ha)	water retention time (yr)	arsenic applied (1950–1969) (kg)
Browns (BL)	160	2.4	13.4	0.004	530	... ^a	19 445 ^b
Big Cedar (BCL)	377	10.4	32.0	0.040	2 295	5.5	61 551
Pewaukee (PwL)	1009	3.0	13.7	0.046	7 156	4.2	107 498 ^c
Monona (LMo)	1350	8.4	22.6	0.12	79 320	1.1	9 402 ^d
Mendota (LM)	3940	12.1	25.6	0.48	68 600	5	3 081 ^d

^a ... : unknown. ^b Arsenite application ended after 1961. ^c Arsenite application ended after 1967. ^d Arsenite application ended after 1964.

Table II. Chemical Characteristics of Sediment Cores

All values as mg/g dry wt except arsenic which is reported as µg/g

depth (cm)	As	P	Fe	Mn	Ca	org C
core: BL I, Browns Lake, Mid North Basin water depth at core location 2.3 m; collected 7/8/68						
5	307	1.16	8.4	0.35	179	... ^a
10	194	1.07	8.2	0.35	190	...
15	204	1.08	7.9	0.34	187	...
25	241	0.85	7.2	0.33	208	...
30	143	0.64	5.3	0.26	231	...
37	72	0.44	3.6	0.19	253	...
47	29	0.40	3.1	0.21	250	...
67	6	0.53	4.4	0.24	208	...
87	4	0.41	3.0	0.15	242	...
core: BL II, Browns Lake, East Basin water depth at core location 11.8 m; collected 7/8/68						
5	226	1.23	10.3	0.34	132	159
10	188	0.96	9.2	0.31	128	174
15	212	1.21	9.8	0.32	142	164
20	149	1.29	7.3	0.28	147	197
25	123	1.20	6.9	0.23	105	227
30	100	1.15	6.2	0.18	94	255
35	71	1.11	6.1	0.20	93	286
40	33	1.09	6.1	0.21	95	283
50	7	1.01	8.5	0.20	30	270
60	5	1.11	9.3	0.25	86	256
70	3	1.31	9.6	0.16	62	295
core: BL III, Browns Lake, North Basin water depth at core location 1.7 m; collected 4/19/69						
5	99	0.49	6.26	0.31	237	59
10	40	0.35	3.62	0.30	266	97
15	36	0.30	3.00	0.32	266	87
20	27	0.30	3.27	0.33	260	89
25	16	0.29	2.95	0.28	279	77
30	13	0.29	2.85	0.24	299	74
35	8	0.33	2.55	0.31	261	82
40		0.30	3.31	0.29	272	78
45	3	0.21	2.32	0.28	271	86
55	1	0.23	1.90	0.24	295	70
65	1	0.25	2.63	0.21	279	71

^a ... : Not determined.

Table III. Chemical Characteristics of Sediment Cores of Lakes Monona and Mendota

All values as mg/g dry wt except arsenic which is reported as µg/g

depth (cm)	As	P	Fe	Mn	Ca	org C
core: LMo-I, Monona Lake, North Central Basin water depth at core location 15.9 m; collected 6/30/66						
5	27	1.83	18.5	0.76	113	... ^a
10	33	1.71	17.0	0.72	114	...
15	40	1.51	17.4	0.67	122	...
20	45	1.60	20.3	0.72	121	...
25	48	1.66	20.0	0.74	114	...
30	68	1.77	20.1	0.81	108	...

Table III. Continued

depth (cm)	As	P	Fe	Mn	Ca	org C
35	79	1.82	19.3	0.88	106	...
40	30	1.44	16.5	0.78	144	...
45	7	1.34	13.2	0.70	172	...
50	3	1.33	12.0	0.71	157	...
55	4	1.17	10.9	0.49	172	...
65	4	1.22	7.7	0.46	239	...
75	5	1.12	7.3	0.41	223	...
85	3	0.91	6.5	0.37	212	...
95	3	0.76	5.5	0.38	191	...
105	3	0.76	5.7	0.38	191	...
core: LMo-II, Monona Lake, South Central Basin						
water depth at core location 21.0 m; collected 6/10/69						
5	23	1.74	18.2	0.81	118	101
10	25	1.76	17.8	0.81	113	99
15	27	1.67	17.6	0.74	98	102
20	29	1.71	17.4	0.71	97	107
25	29	1.67	15.6	0.71	107	107
30	27	1.65	16.6	0.74	112	102
35	36	1.60	17.8	0.75	102	99
45	49	1.64	15.9	0.81	99	95
55	51	1.70	17.2	0.85	96	98
65	70	1.51	16.2	0.82	98	96
70	95	1.77	17.5	0.88	90	93
75	97	1.83	17.8	0.88	90	86
80	26	1.48	17.4	0.79	96	92
85	9	1.32	16.7	0.70	102	86
95	7	1.69	13.1	0.87	120	97
100	5	1.43	10.9	0.73	118	101
core: LMo-III, Monona Lake, West Basin						
water depth at core location 13.3 m; collected 6/10/69						
5	24	1.89	18.4	1.03	136	...
10	28	1.69	21.1	0.98	142	...
15	37	1.38	19.1	0.71	129	...
20	48	1.48	19.4	0.78	129	...
25	65	1.54	19.6	0.77	114	...
30	69	1.72	19.4	0.84	124	...
35	29	1.26	15.9	0.70	150	...
40	15	1.34	14.7	0.70	165	...
45	12	1.45	14.6	0.70	170	...
50	6	1.50	10.0	0.58	216	...
55	3	1.32	9.1	0.54	223	...
core: LMo-IV, Monona Lake, East Basin						
water depth at core location 6 m; collected 6/10/69						
5	29	1.79	79.5	0.97	138	...
10	37	1.53	20.2	0.84	145	...
15	44	1.50	19.9	0.78	123	...
20	58	1.46	19.2	0.79	122	...
25	86	1.44	18.3	0.76	137	...
30	57	1.57	17.0	0.85	146	...
35	13	1.56	14.4	0.70	171	...
40	19	1.53	15.0	0.78	181	...
45	9	1.55	16.3	0.72	178	...
60	5	1.68	8.8	0.59	225	...
75	6	1.60	9.0	0.62	239	...
core: LM I, Mendota Lake, Central Basin						
water depth at core location 21.2 m; collected 2/18/69						
5	10	1860	20.7	1.36	131	...
10	9	1560	21.6	1.25	109	...
20	12	1390	25.5	1.10	88	...
30	8	1440	20.4	0.97	127	...
45	6	918	16.1	0.50	114	...
60	3	89	6.9	0.46	261	...
75	3	66	5.4	0.40	256	...
90	4	697	8.4	0.33	250	...

^a ... : Not determined.

(3), approximately 10 years before Big Cedar Lake. Pewaukee Lake is also not as deep as Big Cedar, having a maximum depth of 13.7 m as compared to 32.0 m. Downward mixing of

surface sediments would be expected to be more vigorous in the shallower basin.

Deep mixing of surficial sediments appears to be most

Table IV. Chemical Characteristics of Sediment Cores of Big Cedar Lake

All values as mg/g dry wt except arsenic which is reported as $\mu\text{g/g}$

depth (cm)	As	P	Fe	Mn	Ca	org C
core: BCL I, Big Cedar Lake, South Basin water depth at core location 29.8 m; collected 9/29/68						
5	549	0.81	18.5	0.71	129	71
10	47	0.67	21.2	0.66	104	63
15	11	0.71	20.8	0.68	118	46
20	12	0.77	22.5	0.56	106	41
25	9	0.76	17.9	0.47	115	54
30	5	0.82	12.6	0.49	189	57
35	3	0.79	6.7	0.47	305	49
40	3	0.73	6.9	0.42	292	52
core: BCL II, Big Cedar Lake, North Basin water depth at core location 8.2 m; collected 9/29/68						
5	316	1.08	11.5	0.54	165	99
10	291	0.99	11.7	0.54	161	102
15	304	0.98	13.3	0.51	147	109
20	319	0.99	13.5	0.50	146	103
25	255	0.96	13.1	0.46	139	107
30	166	0.92	14.3	0.51	144	104
35	115	0.84	15.2	0.49	163	90
40	70	0.78	14.5	0.44	137	105
50	15	0.66	5.9	0.35	263	81
60	8	0.55	4.7	0.31	281	62
69	5	0.54	4.7	0.28	273	56
core: BCL III, Big Cedar Lake, North Bay water depth at core location 1.0 m; collected 5/24/69						
5	150	1.49	9.8	0.23	33	320
10	76	1.31	10.2	0.16	24	341
15	74	1.30	10.9	0.16	23	317
20	70	1.31	9.6	0.15	26	330
25	73	1.29	10.1	0.15	28	341
30	149	1.17	4.6	0.10	15	458
35	67	1.26	9.3	0.14	23	328
40	50	1.30	10.1	0.16	27	311
50	29	1.14	10.6	0.14	38	291
60	26	1.20	9.6	0.14	29	301
70	23	1.19	9.8	0.14	27	310
80	19	0.93	9.7	0.12	28	309
90	12	0.91	8.7	0.14	70	275
core: BCL IV, Big Cedar Lake, Lindem Point water depth at core location 1.0 m; collected 5/24/69						
5	191	0.68	5.7	0.50	259	60
10	142	0.65	5.2	0.49	264	65
15	83	0.55	5.6	0.42	276	50
20	57	0.47	4.9	0.40	282	44
25	50	0.47	4.5	0.39	298	39
30	27	0.40	4.2	0.36	305	35
35	11	0.34	4.0	0.37	315	30
40	5	0.23	3.7	0.39	323	22
core: BCL V, Big Cedar Lake, South Bay water depth at core location 1.9 m; collected 5/24/69						
5	659	0.84	9.3	0.43	175	66
10	186	0.71	7.6	0.38	222	48
15	22	0.48	6.3	0.31	262	37
20	13	0.44	5.3	0.26	257	25
25	5	0.45	5.5	0.29	273	21
30	5	0.31	5.2	0.31	310	22
35	3	0.25	4.1	0.22	318	59
40	3	0.26	4.3	0.30	328	53

prominent in the cores taken from the deep areas of relatively shallow basins, as in the case of cores BCL II and Browns Lake (BL) I and II. Each core shows high arsenic concentrations to depths of 20–25 cm, and profiles which tend to parallel their

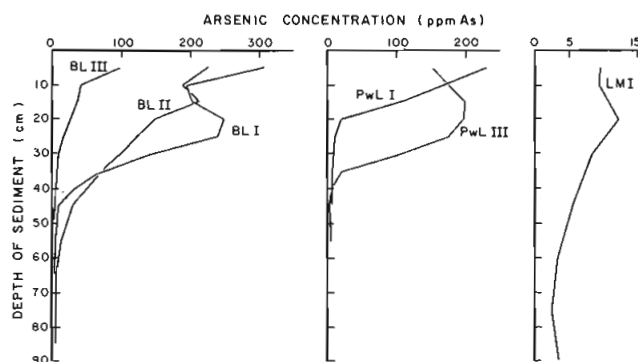


Figure 1. Arsenic profiles for Brown's Lake, Pewaukee Lake, and Lake Mendota sediment

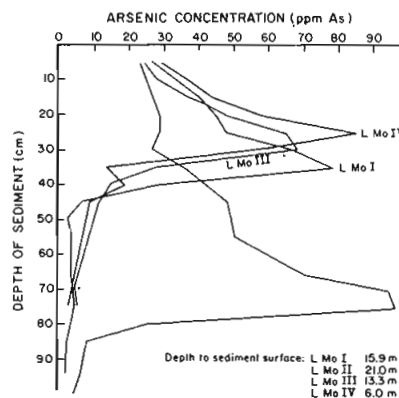


Figure 2. Arsenic profiles for Lake Monona sediment

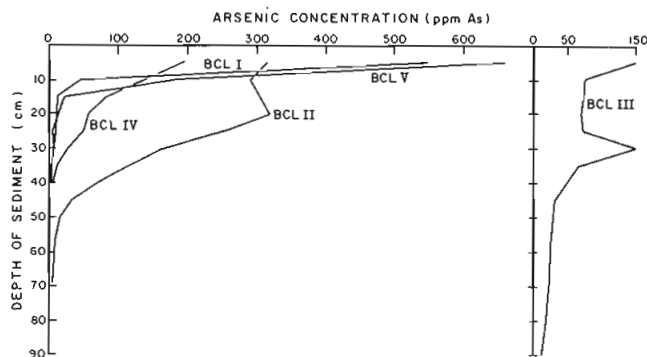


Figure 3. Arsenic profiles for Big Cedar Lake sediment

chemical transitions. The BL II core had no underlying marl. Core BL I was taken at a water depth of only 2.3 m, but it is located in the deepest area of its basin. In addition to mixing, sedimentation should be intensified by the transfer of resuspended sediment to the deepest area in the basins.

The flow of resuspended sediment to the deeper areas of a lake is best indicated by Lake Monona (L Mo) core profiles (Figure 2). Large-scale applications of sodium arsenite to the lake ended in 1944 (14), and the arsenic peaks appear at sediment depths relative to the depths of the water column above the sediment. Arsenic concentrations in the surface sediments have steadily declined due to the mixing with the annual sediment loads since the end of arsenite applications.

Arsenic accumulation is limited to the upper 15 cm for BCL III, BCL IV, and BCL V (Figure 3), and showed increases in

Table V. Chemical Characteristics of Sediment Cores of Pewaukee LakeAll values in mg/g except arsenic which is reported as $\mu\text{g/g}$

depth (cm)	As	P	Fe	Mn	Ca	org C
core: PwL I, Pewaukee Lake, West Basin water depth at core location 14.5 m; collected 7/22/68						
5	232	0.93	17.2	0.79	102	80
10	172	0.96	18.4	0.66	96	76
15	106	0.92	19.8	0.71	90	73
20	18	0.79	20.8	0.64	86	62
25	11	0.76	21.5	0.57	91	57
30	8	0.82	20.8	0.66	89	57
35	7	0.86	21.9	0.54	84	...
40	6	0.66	24.1	0.52	85	...
45	6	0.65	23.9	0.47	138	...
core: PwL III, Pewaukee Lake, East Basin water depth at core location 2.5 m; collected 7/22/68						
5	157	1.01	13.4	0.39	88	...
10	181	0.97	13.7	0.38	77	...
15	199	1.04	12.6	0.34	67	...
20	197	0.91	11.5	0.29	66	...
25	175	1.01	11.6	0.27	78	...
30	106	0.75	20.4	0.29	72	...
35	19	0.85	3.0	0.12	33	...
40	6	0.77	2.3	0.08	33	...
45	3	0.75	2.1	0.05	33	...
55	5	0.67	2.3	0.04	33	...

^a ... : Not determined.**Table VI. Coefficients of Correlation for Total Cores**

core	phosphorus (% P)	iron (% Fe)	manganese (ppm)	calcium (% CaCO ₃)	org C (%)
with arsenic					
BL I	0.25	0.32	0.90	-0.75	...
BL II	0.04	0.10	0.84	0.90	-0.93
BL III	0.26	0.34	0.56	-0.76	-0.29
LMo I	0.23	0.30	0.80	-0.82	...
LMo II	0.13	0.15	0.64	-0.78	-0.41
LMo III	0.09	0.27	0.41	-0.85	...
LMo IV	-0.15	0.23	0.44	-0.76	...
BCL I	0.10	0.07	0.59	-0.24	0.76
BCL II	0.27	0.22	0.86	-0.74	0.77
BCL III	0.18	-0.16	0.40	-0.45	0.73
BCL IV	0.26	0.31	0.96	-0.98	0.93
BCL V	0.24	0.31	0.86	-0.84	0.60
PwL I	0.25	0.34	0.81	0.90	0.96
PwL III	0.25	0.27	0.92	0.89	...

^a ... : Not determined.

concentration uniformly to the surface. The dilution by the calcium carbonate of all other parameters is apparently sufficient to yield the good correlations found, especially with iron. These nearshore areas are approximately a meter in depth, and there is no depletion of dissolved oxygen in the water column. Coprecipitation of arsenic with both hydrous ferric oxides and calcium carbonate would be likely.

Core BCL III is highly organic and unconsolidated, having a total solids content ranging from 6 to 11%. It was taken from an area of the lake which is essentially marsh. Distilled water extraction of these sediments yielded 62% of the total arsenic in 75 days. This may account for the relatively low arsenic concentrations in the core, for an area which received direct

spraying with sodium arsenite.

Core PwL III was taken from the east basin of Pewaukee Lake, which was formed by the impoundment of a stream from the west basin. The lower 20 cm of the core consists of vegetation submerged by the impounding and since been overlaid with silt. The sediment is not calcareous, and is relatively uniform in composition through the upper 25 cm. A high deposition rate, i.e., exceeding 5 mm/yr, would not be expected in this very productive basin. A 77-day distilled water extraction of this material yielded 42% of the total arsenic.

In all the cores, sediment arsenic concentrations fall to approximately 10 ppm As or less. That this level represents background concentrations is indicated by the Lake Mendota core which has an average value of 6.8 ppm As, with a rise in the upper 20 cm of core to 10 ppm As. These values agree well with those of Bors (15) for Mendota cores and those of Ruch et al. (16) for Lake Michigan.

Discussion

Arsenic in the Lake Mendota core shows positive correlation with Fe, Al, and K. Fe has generally been considered a primary control for arsenic in the water column, principally through sorption of arsenate forms on hydrous ferric oxides and their subsequent sedimentation (17). Using jar studies of water and sediment from an oligotrophic lake, Kanamori (18) found that arsenic sorbed to hydrous ferric oxides to an As/Fe equal to 1.3×10^{-3} . Much earlier, Keaton and Kardos (19) had added Fe_2O_3 to clay and studied arsenic sorption. They obtained an As/Fe value of 3.6×10^{-3} . Kanamori (18) found an As/Fe value in Lake Kizaki-Ko sediments of 0.46×10^{-3} , indicating undersaturation of the hydrous ferric oxides. In the Lake Mendota core, As/Fe ratios average 0.44×10^{-3} . Similar results were reported by Bors (15).

Sediments from treated lakes have As/Fe ratios considerably larger than these and require other sorption sites, such as aluminum hydrous oxides, sulfides, and clay minerals (20). Correlation of As with Al and K may indicate sorption with clays or simply the erosion of Lake Mendota basin soils. Bors (15) reported an As/Fe ratio of 0.25×10^{-3} for Lake Mendota sediments.

The productive, hard water lakes studied also suggest that arsenic may be bound in sediments in the form of a hydroxylapatite or a carbonatoapatite, analogous to phosphate (21).

Conclusions

High loadings of arsenic, in the form of sodium arsenite, to recreational lakes in southeastern Wisconsin show appreciable arsenic retention within the sediments over an extended period. While core data show considerable increases of arsenic concentrations toward the surface of the sediments, extraction results indicate that virtually all the arsenic is in a nonoccluded form. Extended extractions with distilled water indicated that highly calcareous sediments under oxic conditions yield the least amount of arsenic (25% of the total), and that highly organic sediments, again under oxic conditions, yield the most (in excess of 60%). Deep sediments, under anoxic conditions, yielded 35–49% of the total arsenic present. Arsenite chemistry is similar in the process of iron control, but sulfides would play a more direct role through actual compound formation as well as coprecipitation, than in the case of arsenates. With the latter, sulfide control is indirect through the formation of ferrous sulfides, releasing arsenates. Furthermore, arsenite would largely exist in an undissociated form at sediment pH levels and would not participate in exchange processes.

Lack of any predominant correlating parameter with arsenic is another substantiating factor. For some cores the

greatest correlation appeared to be between arsenic and manganese, although this element would be expected to exert a minimal control on arsenic (20). However, Sevast'yanov (22) points out that arsenic is one of the most mobile elements in marine sediments and closely resembles manganese in this capacity. The existence of two oxidation states is cited as the reason for the mobility.

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