5

6

7

8

9 10

11

12

13

14

15

16

17

18

19

20

21

22

49

50

51

Effectiveness and Reliability of Arsenic Field Testing Kits: Are the Million Dollar Screening Projects Effective or Not?

MOHAMMAD MAHMUDUR RAHMAN, [†]
DEBAPRIYO MUKHERJEE, [‡]
MRINAL KUMAR SENGUPTA, [†]
UTTAM KUMAR CHOWDHURY, [†]
DILIP LODH, [†] CHITTA RANJAN CHANDA, [†]
SHIBTOSH ROY, [§] MD. SELIM, [§]
QUAZI QUAMRUZZAMAN,§
ABUL HASNAT MILTON,#
S. M. SHAHIDULLAH, [#]
MD. TOFIZUR RAHMAN, # AND
DIPANKAR CHAKRABORTI*,†
School of Environmental Studies Jadavnur University
Calcutta India Central Pollution Control Board
Calcutta, India, Ochina Fondion Control Doard,
Baro Moghbazar Dhaka Community Hospital,
NCO Forum for Drinking Water Supply & Sanitation
I almatia Dhaka Bandladash
Lainatia, Dilaka, Dangiautsii

The exposure of millions to arsenic contaminated water 23 from hand tube wells is a major concern in many Asiatic 24 countries. Field kits are currently used to classify tube wells 25 as delivering arsenic below 50 μ g/L (the recommended 26 limit in developing countries) as safe, painted green or above 27 50 μ g/L, unsafe and painted red. More than 1.3 million 28 tube wells in Bangladesh alone have been tested by field 29 kits. A few million U.S. dollars have already been spent 30 and millions are waiting for the ongoing projects. However, 31 the reliability of the data generated through field kits is 32 now being questioned. Samples from 290 wells were tested 33 34 by field kits and by a reliable laboratory technique to ascertain the reliability of field kits. False negatives were 35 as high as 68% and false positives up to 35%. A statistical 36 analysis of data from 240 and 394 other wells yielded similar 37 rates. We then analyzed 2866 samples from previously 38 labeled wells and found 44.9% mislabeling in the lower range 39 (<50 μ g/L) although mislabeling was considerably 40 reduced in the higher range. Variation of analytical 41 results due to analysts and replicates were pointed out 42 adopting analysis of variance (ANOVA) technique. Millions 43 of dollars are being spent without scientific validation of 44 the field kit method. Facts and figures demand improved, 45 environmentally friendly laboratory techniques to produce 46 47 reliable data. 48

Introduction

Among the 21 countries in different parts of the world affected by groundwater arsenic contamination, the largest population at risk is in Bangladesh (1-4) followed by West Bengal in India (5-10). In recent years evidence of arsenic groundwater contamination has emerged from other Asiatic countries including Lao PDR, Cambodia, Mayanmar (11), and Nepal (12). In Vietnam (13), several million people consuming untreated groundwater may run a considerable risk of chronic arsenic poisoning.

From our analysis of more than 100 000 hand tube wells from West Bengal in India and 34 000 from Bangladesh, we found 9 out of the total 18 districts in West Bengal, India and 50 out of the total 64 districts of Bangladesh that carry arsenic levels in groundwater above $50 \mu g/L$, the recommended value of arsenic in drinking water for India and Bangladesh although the World Health Organization (WHO) recommended value of arsenic in drinking water is $10 \mu g/L$. The area and population of the 50 districts of Bangladesh are 118 849 km² and 104.9 million, respectively, and of the 9 districts of West Bengal are 38 865 km² and 50.0 million, respectively, but the actual number at risk is still under evaluation.

The British Geological Survey (BGS), on the basis of 3 534 hand tube well water samples from throughout Bangladesh excluding the Chittagong Hill Tracts, estimated that 35 million inhabitants of Bangladesh drink arsenic contaminated water (above 50 μ g/L) (14). The magnitude of arsenic contamination in Bangladesh surfaced only after the international conference (15) on arsenic in Dhaka, Bangladesh in February 1998. In a post-conference report (16) the World Bank's local chief estimated that tens of millions of people are at risk for health effects and that 43 000 villages of 68 000 were presently at risk or could be at risk in future. In the same report (16), the World Health Organization (WHO) predicted that, within a few years, death across much of southern Bangladesh (1 in 10 adults) could be from cancers triggered by arsenic.

Beginning in 1997 the World Bank, UNICEF, WHO and other international aid agencies came forward to combat the situation. A unanimous decision was taken to test all the hand tube wells to gauge the magnitude of the calamity. Both in Bangladesh and West Bengal, India, it was decided to use field test kits to determine the arsenic concentration in hand tube wells and to color tube wells having arsenic less than 50 μ g/L green and those above 50 μ g/L red. Based on the available published reports (17-22), at least 1.3 million water samples from hand tube wells were analyzed by field kits and an estimated 1 million hand tube wells were colored red (>50 μ g/L) or green (<50 μ g/L) based on the Merck kit which has 100 μ g/L of arsenic as the minimum detection limit. Using the same kit, the Bangladesh Arsenic Mitigation Water Supply Project (BAMWSP) analyzed 617 366 hand tube wells (17), and DPHE-UNICEF (18) analyzed 403 651 tube wells. The Dhaka Community Hospital (DCH) analyzed 137 971 samples by Merck kit and 19 436 samples by National Institute of Preventive and Social Medicine (NIPSOM) kit (19). Other organizations from Bangladesh such as the NGO Forum (20), Grameen Bank (21), CARE (21), BRAC (22) also used the Merck kit and NIPSOM kit for testing. Only a few thousand samples were analyzed by the General Pharmaceuticals Limited (GPL) kit. By 2001, the Merck doubling kit (minimum detection limit 10 μ g/L) and the Hach kit (minimum detection limit $10 \,\mu g/L$) were considered for use in the ongoing projects by many organizations. Wide scale use of the Arsenator field kit has not been reported. The government of West Bengal and UNICEF launched a 1.1 million dollar project (23) in 1999 to test the hand tube wells with the All India Institute of Hygiene and Public Health (AIIH&PH) field kit.

52

53

^{*} Corresponding author phone: +91-33-4146233; fax: +91-33-4146266; e-mail: dcsoesju@vsnl.com.

 [†] Jadavpur University.
 [‡] Central Pollution Control Board.

[§] Dhaka Community Hospital.

[#] NGO Forum for Drinking Water Supply & Sanitation.

TABLE 1. Details of Three Field Kits (NIPSOM, GPL and Merck)							
parameter	NIPSOM (National Institute of Preventive and Social Medicine) kit	GPL (General Pharmaceuticals Ltd.) kit	Merck kit				
design	box oversize and heavy; contents scatter during transit	box oversize and heavy; contents scatter during transit	compact and light; contents intact during transit				
kit capability minimum detection limit (µg/L)	semiquantitative 10	semiquantitative 10	semiquantitative 100				
arsenic detection range (µg/L)	10, 20, 50, 100, 200, 300, 400, 500 & 700	10, 50, 100, 200, 400, 500, 600, 800, 900, 1000 & 1500	0, 100, 500, 1000, 1700 & 3000				
chemicals used	HCI (1:1), KI (reagent 1), Zn powder (reagent 2), SnCl ₂ (reagent 3), bromide paper (test strip)	HCI (1:1), KI (reagent 1), SnCl ₂ (reagent 2), Zn powder (reagent 3), bromide paper (test strip), lead acetate cotton wool	mixture of KI, SnCl ₂ and Zn as reagent 1 and HCl as reagent 2				
procedure	Place the disk paper between flattened surface of two glass flanges and secure the flanges with the clip. Take 15 mL of test solution in a test tube and add 0.1 g of reagent 1, 0.5 g of reagent 2 and 0.1 g of reagent 3. Now add 4 mL of 1:1 HCI and insert the end of the flange in the test tube and shake gently. Allow to stand for 5 min. Remove the clipped flanges from the test tube and compare the color change obtained on disk paper with the color scale.	Place 15 mL of water in the reaction test tube and add 4 mL of HCI, one level spoonful of reagent 1 and same amount of reagent 2. Shake gently and wait for 10 min. While waiting 10 min, use forceps to place bromide paper between the two sections of flange unit and secure with clamp. After 10 min, add one level spoonful of reagent 3 to the reaction mixture in the test tube. Immediately fit the flange unit into the mouth of the test tube. Gently shake the test tube and wait for 10 min. Using forceps, remove bromide paper and compare to color chart.	Insert a test strip into the slot in the cap of reaction vessel. Place 5 mL of test solution in the reaction vessel and add 1 measuring spoonful of reagent 1 and shake. Add 10 drops of reagent 2 and close the reaction vessel immediately with cap. Allow to stand for 30 min and shake gently for 2–3 times. Immerse briefly in water and compare with color scale.				
time required for test	5 min	20 min	30 min				
health hazard	 (i) accidental spillage of acid (ii) toxic arsine gas may cause bealth bazard 	(i) accidental spillage of acid (ii) toxic arsine gas may cause bealth bazard	(i) accidental spillage of acid (ii) toxic arsine gas may cause bealth bazard				
cost per test in U.S.\$ (approximate)	0.3	0.4	0.5				

118 119 120

121

122

123

124

125

126

127

128

129

130

149

All field kits used to date are based on the principle of mercuric bromide stain method (24). The accuracy of the bromide stain method was evaluated in a round robin exercise, and it was reported that the arsenic concentrations for a solution of arsenite at 50 μ g/L in distilled water had a relative standard deviation (RSD) of 75% (24). It was further reported (24) that the mercuric bromide stain method was incapable of producing quantitative meaningful results with RSD \leq 10% below a concentration 150 μ g/L of arsenic. A recent publication (25) notes that the Merck field kit "is not only inadequate to screen water samples containing less than 100 ppb of arsenic, but also provides toxic arsine gas that may be a health hazard".

131 We have been working in the villages of Bangladesh for the last 7 years to know the magnitude of the contamination 132 and the number of people affected. While working in the 133 field during last 4 years, we found many tube wells were 134 135 colored either 'red' or 'green' by field workers after testing by field kits. During our survey we collected samples from 136 time to time from tube wells colored red/green and found 137 138 after analyzing these samples by flow injection hydride generation atomic absorption spectrometry (FI-HG-AAS) 139 technique that a good percentage of the tube wells were not 140 correctly colored. This discrepancy initiated us to study the 141 142 efficiency of the field kits.

Mode of Evaluation 143

This paper will evaluate the efficiency of field kits and show 144 how far the field kits are justified to be used in villages to 145 color hand tube wells red (>50 μ g/L) or green (<50 μ g/L) to 146 indicate unsafe or safe and whether the adopted millions of 147 dollar in projects using field kits are justified or not. 148

Our field kit evaluation had 3 phases.

First Phase. When we started field kit evaluation in Bangladesh in 1998, 3 kits, NIPSOM, GPL and Merck (see Table 1), were widely used, but the Merck (doubling method) kit was not available at that time. In the first phase, we analyzed 290 samples by NIPSOM, GPL and Merck kits and compared the results with flow injection hydride generation atomic absorption spectrometry (FI-HG-AAS) to test the accuracy of these kits. Several organizations used the Merck kit for coloring hand tube wells red (>50 μ g/L) or green (<50 μ g/L). Since the Merck kit has a minimum detection limit for arsenic of 100 μ g/L, we excluded Merck kit results below 100 μ g/L from our first phase evaluation study. After evaluation of the field kit, we collected 2866 field samples from hand tube wells colored red/green on the basis of field kit testing and analyzed these samples by FI-HG-AAS.

150

151

152

153

154

155

156

157

158

159

160

161

162

163

164

165

166

167

168

169

170

171

172

173

174

175

176

177

178

179

180

181

182

183

Second Phase. From 2000 onward the Merck (doubling method) kit was used in the field for minimum detection of 10 µg/L level of arsenic. In August 2000, BRAC-Bangladesh reported (22) analytical results of 240 water samples from Panisara Union, Jikargachha police station of Jessore district. The samples were analyzed by the Asia Arsenic Network (AAN) field kit and by the Merck (doubling method) kit and cross-checked by continuous flow hydride generation AAS by Intronics Technology Center (ITC), Bangladesh. We made a statistical evaluation of the results of all 240 samples that were reported by BRAC in their report (22) to compare the accuracy of the AAN kit and the Merck (doubling method) kit.

Third Phase. The AIIH&PH field kit results from hand tube wells of Murshidabad and Malda districts were also compared with FI-HG-AAS in our laboratory. UNICEF, Calcutta sent 394 field samples to our laboratory, which were simultaneously measured by the AIIH&PH field kit for comparison with the field kit results.



FIGURE 1. Adjoining red (unsafe) and green (safe) colored tube wells of the same depth as defined by field kit testing (Village-Belghor, Police Station-Haziganj, District-Chandpur, Bangladesh).



AFFECTED DISTRICTS OF BANGLADESH

27 Shariatpur

29 Mymensingh

30 Narayanganj

32 Lakshmipur

34 Kishoregani

36 Brahmanbaria

33 Netrokona

35 Narsingdi

37 Comilla

39 Feni

42 Bogra

43 Rangpur

44 Gazipur

46 Habiganj 47 Kurigram

49 Moulavibazar

50 Gaibandha

45 Sylhet

48 Dhaka

38 Noakhali

40 Sunamganj

41 Chittagong

28 Sherpur

31 Candpur

FIGURE 2. The distribution of groundwater arsenic contamination in Bangladesh and the districts where we collected red/green tube well water samples for FI-HG-AAS analysis (*).

184 Experimental Section

185 Study Area. For the first phase, 290 hand tube wells were
186 selected from 6 police stations (Songargaon, Moulavibazar
187 sadar, Rajnagar, Srimangal, Kamalganj and Baro Lekha) of
188 2 districts (Narayanganj and Moulavibazar) of Bangladesh

to compare arsenic determination by the field kits (NIPSOM, GPL, Merck) with FI–HG–AAS analysis. In the first phase, the accuracy of the field kits was evaluated. We then analyzed 2 866 samples from hand tube wells by FI–HG–AAS already colored red or green (Figure 1) on the basis of field kit testing





FIGURE 3. The color chart of arsenic concentrations as provided by the NIPSOM, GPL and Merck kits.

from 60 villages of 20 police stations in 10 districts of 194 195 Bangladesh. Figure 2 shows the distribution of groundwater contamination by arsenic in Bangladesh and the districts 196 from where we collected red/green hand tube well water 198 samples for arsenic analysis.

197

199

200

201

202

203

204

205

206 207

208

209

210

211

212

213

214

215

216

217

218

219

220

221

222

223

224

225

226

227

228

229

230

Selection of Tube Wells and Sampling. The 290 water samples from hand tube wells were collected at random after pumping for at least 10 min. About 1 L water from each tube well was collected in a pre-cleaned and acid washed plastic container with 1 mL of concentrated nitric acid, thoroughly mixed and equally distributed among 10 pre-cleaned plastic containers of 50 mL capacity. Six containers were tested by field kit at a central site in each village; four went to the School of Environmental Studies (SOES) for analysis by FI-HG-AAS.

Estimation of Arsenic using Field Kits, Spectrophotometric Method and FI-HG-AAS Method. The 290 samples were evaluated by NIPSOM, GPL and Merck kits (Table 1) utilizing the color charts provided by the manufacturers (Figure 3).

Spectrophotometric Method. A Shimadzu (Japan) double beam Spectrophotometer (Model 1601) was used. Total arsenic in water was determined with Ag-DDTC-chloroformhexamethylenetetramine as absorbing solution (26-27). The determination limit of arsenic by the spectrophotometric method with 95% confidence is 30 μ g/L.

Flow Injection Hydride Generation Atomic Absorption Spectrometry Method (FI-HG-AAS). A FI-HG-AAS system was assembled from commercially available instruments and accessories in our laboratory. A Perkin-Elmer Model 3100 atomic absorption spectrometer equipped with a Hewlett-Packard Vectra Computer with GEM software, Perkin-Elmer EDL system-2, arsenic lamp (lamp current 400 mA) and Varian AAS Model Spectra AA-20 with Hollow Cathode As lamp (lamp current 10 mA) were used. The flow injection assembly consists of an injector, Teflon T-piece, tigon tubings and other parts for the FI system from Omnifit, UK. The peristaltic pump (VGA-76) from Varian and Minipuls 3 from 231 Gilson were incorporated into the FI system. In the FI-HG-232 AAS system the sample was injected into a carrier stream of 233 5 M HCl by means of a six-port sample-injection valve fitted 234 with 50 μ L sample loop. The injected sample, together with 235 carrier solution, met subsequently with a continuous stream 236 of sodium borohydride (1.5%) dissolved in sodium hydroxide 237 (0.5%). Mixing with sodium borohydride generated arsenic 238 hydride (arsine), which subsequently entered into the ice 239 water bath and then into the gas-liquid separator apparatus, 240 which was cooled with ice-cold water. Carrier gas nitrogen 241 transported the arsine to the quartz tube mounted in the 242 air-acetylene flame for AA measurement. The minimum 243 detection limit with 95% confidence level was 3 μ g/L of 244 arsenic. Details of the instrumentation, analytical procedures 245 and analytical performances were reported in our earlier 246 publications (27-29). 247

Statistical Analyses. Standard statistical techniques such as mean, standard deviation (SD), minimum, maximum and ANOVA were applied to study the variation among the values. Correlation coefficients and linear regression were utilized for the relationship of values obtained by two methods.

248

249

250

251

252

253

254

255

256

257

258

259

260

261

262

263

264

265

266

267

268

269

270

271

272

273

274

275

276

277

278

279

280

281

282

283

284

285

286

287

288

289

290

291

292

293

294

295

296

297

298

299

Interlaboratory Comparison and Analysis of Standard Reference Material. Aliquots of the field samples were sent to NGO Forum, Bangladesh for analysis of arsenic spectrophotometrically. After the analysis, the residual samples were sent by courier to the SOES laboratory, Calcutta for FI-HG-AAS Analysis. Three such batches (total 62 samples) were analyzed within 3-7 days for interlaboratory comparison. A similar interlaboratory study was done with the Environment and Public Health Organization (ENPHO), Nepal (30) which also used FI-HG-AAS. Standard Reference Materials (SRM) and an EPA water standard for arsenic were used to check the accuracy of FI-HG-AAS method (28, 31).

Analysis of 2866 Hand Tube Wells by FI-HG-AAS, which Were Colored Red/Green after Analyzing with Field Kits by Organizations in Bangladesh. Water samples from 2 866 hand tube wells, previously analyzed by field kits and colored either red or green, were stored in 10 mL plastic bottles with 1 drop of nitric acid added to each bottle as preservative. Around 5% of the samples was collected from the same sources in duplicate to check the variation of arsenic concentration among the two samples. Samples were analyzed by FI-HG-AAS at the SOES laboratory within 3 to 7 days.

Statistical Analysis of the Results of 240 Samples Measured by the Merck (Doubling Method) and AAN Kits **Compared with Continuous Hydride Generation AAS from** ITC Laboratory, Dhaka (Reported by BRAC, Bangladesh). An independent report which compared two field kits with a continuous hydride generation AAS assay was published by BRAC, Bangladesh (22) in August 2000 utilizing 240 groundwater samples from Panisara Union of Jikargachha police station, Jessore district. Merck (doubling method) and AAN kits were used, and the results of these two field kits were compared with continuous hydride generation AAS from Intronics Technology Center (ITC) laboratory, Dhaka, Bangladesh and the results published in tabular form (22). With the permission of BRAC, we made a statistical evaluation of the analytical results of 240 samples.

Validation of AIIH&PH Field Kit Used in Arsenic Affected Villages of Murshidabad and Malda (an UNICEF Project) of West Bengal, India. We then compared the AIIH&PH field kit with our FI-HG-AAS system. Three groups collected 394 samples from arsenic affected districts of Murshidabad (Group A, 192 samples) and Malda (Group B, 104 samples and Group C, 98 samples) as having arsenic below 50 μ g/L and above 50 µg/L. We utilized FI-HG-AAS analysis to test the accuracy of the information generated by the field kit.



FIGURE 4. (a) Correlation of arsenic analyses of groundwater samples by FI-HG-AAS and the spectrophotometric method. (b) Correlation of arsenic analyses by FI-HG-AAS by two laboratories: (i) School of Environmental Studies (SOES), Jadavpur University, India (ii) Environment & Public Health Organization (ENPHO), Nepal.

300 Results and Discussion

301

302

303

304

305

306

307

308

309

310

311

312

313

314

315

We have reported elsewhere (*32*) the effects of various parameters (time, temperature, preservative, types of bottle, etc.) on arsenic assays. If the samples are collected in prewashed (overnight with 1:1 HNO₃) plastic bottles (10 mL capacity) adding one drop of concentrated HNO₃ as preservative to 10 mL of water then the samples do not show any significant variation in total arsenic content when measured within 7 days. The correlation between the output of the spectrophotometric and FI–HG–AAS methods was defined by the least-squares technique to calculate the regression equation and correlation coefficient ($r^2 = 0.983$) as diagrammed in Figure 4a. The correlation (r = 0.985) of 10 samples from arsenic affected villages of Nepal analyzed by FI–HG–AAS by ENPHO laboratory (*30*) and our laboratory are shown in Figure 4b.

Statistical Analysis of 290 Hand Tube Wells Measured 316 317 by 3 Field Kits and FI-HG-AAS. For comparison all the 318 values of FI-HG-AAS corresponding to the value at particular 319 level obtained by field kits were processed to find out the minimum, mean, maximum and standard deviation (SD) to 320 321 have an idea about the degree of deviation. Minimum, mean minus SD, mean, mean plus SD and maximum values 322 323 obtained by FI-HG-AAS were plotted against the corresponding value of field kits (NIPSOM and GPL kits). Figure 324 325 5a,b shows a wide variation at each level of both NIPSOM and GPL field kits. Based on SD it may be mentioned that 326 327 some of the values (calculated) will be negative even if we



FIGURE 5. (a) Minimum, mean –SD, mean, mean +SD and maximum of FI–HG–AAS values corresponding to each category of arsenic concentration determined by the NIPSOM kit. (b) Minimum, mean –SD, mean, mean +SD and maximum of FI–HG–AAS values corresponding to arsenic concentrations by GPL kit. (c) Minimum, mean –SD, mean, mean +SD and maximum of FI–HG–AAS values corresponding to arsenic concentrations by Merck kit.

consider the mean minus one sigma as shown in Figure 5a,b. The values less than zero have no physical entity.

Since the minimum detection limit of the Merck kit is 100 μ g/L, we limited the comparison to values above this minimum. By FI–HG–AAS, arsenic in the range of 100 μ g/L and above was found in 127 of 290 samples. The Merck kit identified 113 (89%) with an 11% failure rate. Figure 5c shows the wide variation at each level of the Merck kit.

The false positives and false negatives of the field test kits were calculated for each category: (a) $3 \mu g/L$ to $50 \mu g/L$, (b) 50.1 $\mu g/L$ to 100 $\mu g/L$ and (c) >100 $\mu g/L$ (3 $\mu g/L$ is the

TABLE 2 Falco Desitive and Falco Negative Desults Obtained by Field Kits Comparing with EL. U.C. AAS

TADLL	2. 10130 10			ins obtained by	neiu kits comp		IIO AAS	
	ranges	falso (_)	false (+)	truo	falso (_)	GPL	truo	no of observations
3—5 50.1 >10	0 μg/L 100 μg/L 0 μg/L	29 (56.9%) 19 ^a (15.0%) 33 ^b (26.0%)	7 (6.2%) 18 (35.3%) NA NA	69 (61.6%) 4 (7.8%) 108 ^a (85.0%) 94 ^b (74.0%)	35 (68.6%) 11 (8.7%) 22 (17.3%)	3 (2.7%) 9 (17.7%) NA NA	43 (38.4%) 7 (13.7%) 116 (91.3%) 105 (82.7%)	112 51 127
^a In	terms of <5	0 μg/L. ^b In terms	of <100 µg/L		. ,			
NIPSOM kit values (ug/L)	400 - 360 - 300 - 250 - 150 - 150 - 100 - 50 -		Y=-61.8+2 r=0.294 n=51	2.34* X	400 - 400 -	Minimum Mean-SD Mean Maximum Maximum	0 80 100 120 bubling method) k	140 160 180 200 it values (μg/L)
G P L kit values (µg/L)		FI-HG-A	200 250 300 A S values (μg/ Y=-176.9+3.5 r=0.440 n=51 200 250 30 A S values (μg/L	- 350 400 L) *X	FIGURE 7. (a) of FI-HG-AA categories d Minimum, me AAS values c determined b kit, this will well red. Above 10	50 100 15 A Minimum, mea AS values corre etermined by ean – SD, mear orresponding to by the AAN kit have no effect	 Minir Mean Mean Mean Mean Maxi Maxi Maxi Maxi 	+ S D + S D mum

fixed.

FIGURE 6. (a) The variance between FI-HG-AAS and NIPSOM kit values at the range of 50.1–100 μ g/L of arsenic (some points are overlapping each other). (b) The discrepancy between FI-HG-AAS and GPL kit values at the range of $50.1-100 \mu g/L$ of arsenic (some points are overlapping each other).

minimum arsenic concentration determined with 95% confidence by our FI-HG-AAS system.) as shown in Table 2.

At arsenic concentrations below 50 μ g/L (Table 2), false positive values were 6.2% for NIPSOM and 2.7% for GPL kits, supporting use of either kit to categorize wells as safe.

In the range of $50.1-100 \,\mu\text{g/L}$, we found 56.9% and 68.6%false negatives and 35.3% and 17.7% false positives for NIPSOM and GPL, respectively. As a result, unsafe tube wells will be colored green i.e., safe. Although false positives at concentrations above 100 μ g/L are not negligible for either



347 348

339

340

341

342

343

344

345

346

F = ENVIRON. SCI. & TECHNOL. / VOL. xx, NO. xx, xxxx

Overall, there was no systematic bias toward under or over reporting, but random error led to false positives and

mislabeled safe, are 15% for NIPSOM and 8.7% for GPL.

Overall false negatives i.e., reporting as less than 100 μ g/L,

are 26% for NIPSOM and 17.3% for GPL. False positives were

not considered in this range since no boundary line was

that only 3 of 113 were false negative plus one sample between

50.1 and 100 μ g/L. Thus, false negatives were not significant.

On comparison of Merck kit results with 127 samples where

FI-HG-AAS found arsenic above 100 µg/L, then 18 samples

(14.1%) tested by Merck kit as below 100 μ g/L. Although the

variation of Merck kit from FI-HG-AAS is not significant,

the Merck kit cannot be used for field testing of hand tube

wells for arsenic below 100 μ g/L.

For the Merck kit, at concentrations < 50 μ g/L, it was found

362

363

364

365

366

367

349 350

Merck				no, of samples			
ranges	false (–)	false (+)	true	false (–)	false (+)	true	analyzed by AAS
1-50 µg/L		47 (25.6%)	127 (69.5%)		47 (25.6%)	128 (70%)	183
50.1-100 µg/L	14 (46.7%)	5 (16.6%)	11 (36.7%)	15 (50%)	4 (13.3%)	11 (36.7%)	30
>100 µg/L	13 ^a (48.2%)	NÁ	14 (51.8%)	19 ^a (70.4%)	NÁ	8 (29.6%)	27
	23 ^b (85.2%)	NA	4 (14.8%)	25 ^b (92.6%)	NA	2 (7.4%)	

TABLE 4. Validation of AIIH&PH Field Kit by FI-HG-AAS

organizations	total samples analyzed by both field kit and AAS	field kit reported 'Yes' [As >50 µg/L]	field kit reported 'No' [As <50 µg/L]	FI-HG- AAS method [As >50 µg/L]	FI-HG-AAS method [As <50 µg/L]	comparable value of field kit with FI-HG-AAS method (true value)	false (+)	false (–)
А	192	138	54	119	73	172 (89.6%)	19 (9.9%)	1 (0.5%)
В	104	86	18	44	60	58 (55.8%)	44 (42.3%)	2 (1.9%)
С	85 ^a	85	b	68	17	68 (80%)	17 (20%)	

^a Actually 98 samples supplied but out of them 13 samples marked as 'confusion'. Statistical analysis reported on 85 samples. ^b None of the samples reported 'No'.

false negatives. Field kits are not only semiquantitative but also produce false values.

 To review the status of false positives and false negatives we have graphically presented 51 samples from Table 2 in the range of $50.1-100 \ \mu g/L$ of arsenic tested by FI-HG-AAS. Values of FI-HG-AAS are plotted on the *X*-axis against the values of field kits on the *Y*-axis and shown in Figure 6a for the NIPSOM kit and Figure 6b for GPL kit. Slope and intercept are given in the form of an equation. The intercept and angle of slope in Figure 6a,b indicate a weak correlation (r = 0.294 for NIPSOM and r = 0.440 for GPL kit). A vertical and a horizontal line at 50 $\mu g/L$ and 100 $\mu g/L$ are drawn to divide values below $50.1 \ \mu g/L$ and above $100 \ \mu g/L$ L:29 and 35 values of 51 are below $50.1 \ \mu g/L$ and 18 and 9 values of 51 are above $100 \ \mu g/L$ for the NIPSOM and GPL kits, respectively.

Statistical Analysis of the Results of 240 Hand Tube wells by the Merck (Doubling Method) and AAN Kits and Checked by Continuous Hydride Generation AAS as Reported by BRAC, Bangladesh. The detection limit reported for both the Merck (doubling method) and AAN kits is 10 μ g/L. The detection limit of continuous hydride generation AAS of ITC is 1 μ g/L (*33*), and the lowest value detected while analyzing 240 samples by ITC was 1.8 μ g/L.

The same procedure as adopted for NIPSOM and GPL kits (Figure 5a,b) was used to compare the analytical results of 240 water samples tested by the Merck (doubling method) kit and the AAN kit with the AAS value as the true value. A wide variation among the AAS values versus the kit values are shown in Figure 7a for the Merck (doubling method) kit and Figure 7b for the AAN kit grouped in 3 categories: $1-50 \mu g/L$, $50.1-100 \mu g/L$ and above $100 \mu g/L$. The minimum detection limit of continuous flow AAS of ITC laboratory was reported to be $1 \mu g/L$, so lowest value was $1 \mu g/L$ in the first group i.e., $1-50 \mu g/L$. The number of false positives and false negatives are shown in Table 3.

In the range $1-50 \mu g/L$, both the Merck (doubling method) and AAN kits have a false positive rate of 25.6%. That means 47 of 183 wells were erroneously colored red despite delivering safe water.

In the range of $50.1-100 \,\mu$ g/L, the Merck kit shows 46.7% false negatives i.e., 46.7% of the hand tube wells were colored green (safe) despite containing arsenic above $50 \,\mu$ g/L. For the AAN field kit, the false negative rate was 50%.

On analysis of the 27 samples containing more than 100 μ g/L, 48.2% of the samples tested by the Merck kit were read

as less than 50 μ g/L falsely indicating safe water. The AAN kit reported 70.4% below 50 μ g/L when the actual value was above 100 μ g/L by AAS. For all samples above 100 μ g/L by AAS, 85.2% samples were read as below 100 μ g/L by the Merck kit and 92.6% by the AAN kit.

The graphical presentations in Figure 8a–d of two sets of results from Table 3 i.e., (i) 30 samples in the range 50.1–100 μ g/L and (ii) 27 samples above 100 μ g/L of arsenic by AAS and the corresponding Merck (doubling method) kit and AAN kit values use parameters similar to Figure 6a,b. Figure 8a,b indicates that in the range of 50.1–100 μ g/L, 14 and 15 of 30 values are below 50.1 μ g/L while 5 and 4 values are above 100 μ g/L for the Merck (doubling method) and AAN kits, respectively. At concentrations above 100 μ g/L (Figure 8c,d) 13 and 19 values of 27 are below 50 μ g/L while 23 and 25 values are below 100 μ g/L by the Merck (doubling method) and AAN kits, respectively.

Validation of AIIH&PH Field Kit Results by FI–HG– AAS. The field kit of AIIH&PH has a 'Yes'/'No' system. A color on the bromide paper means the water sample contains arsenic above 50 μ g/L ('Yes') while 'No' means no visible color i.e., the sample contains less than 50 μ g/L. UNICEF, Calcutta sent us 394 samples from Murshidabad and Malda, two arsenic affected districts of West Bengal. Three separate groups worked in the field to generate field data utilizing the AIIH&PH kit. Group A sent us 192 samples marked red (above 50 μ g/L) and blue (<50 μ g/L), Group B sent us 104 samples reporting Yes/No and Group C sent us 85 samples marked 'Yes' and 13 samples marked 'Uncertain' which were excluded with statistical analysis of 85 samples. Table 4 compares the results with the FI–HG–AAS values.

Each of the 3 groups had been trained separately in use of the AIIH&PH field kit, worked independently, and had no connection with the other groups. As shown in Table 4, group A generated 89.6% reliable data, whereas group B and group C generated 55.8% and 80% reliable data. It appears that the performance of field kit is not the only factor affecting accuracy. UNICEF, Calcutta no longer uses the AIIH&PH field kit to color hand tube wells safe or unsafe in the villages of West Bengal.

Operation of Field Kit by Skilled and Trained Person. The ANOVA technique was applied to breakdown the total precision into its component e.g. between replicates and between persons trained for using field kits. These two components have an influence on the performance characteristics. These are considered as the optimization criterion

keplicates					
	true value ^a	mean		level of s	significance
	(µg/L)	(µg/L)	SD	analyst ^c	replicates ^c
GPL	67	50	0	NS	NS
NIPSOM	34	10	0	NS	NS
GPL	34	<50	0	NS	NS
NIPSOM	56	30	15	NS	S
GPL	56	50	0	NS	NS
NIPSOM	63	75	26	NS	S
GPL	63	50 or less	b		
NIPSOM	83	110	65	NS	S
GPL	83	50	0	NS	NS
NIPSOM	119	127	78	NS	S
GPL	119	100	0	NS	NS
NIPSOM	150	210	74	NS	S
GPL	150	100	0	NS	NS
NIPSOM	214	303	67	NS	S
a Magazir		AAC bCalaul	otion	not dono o	o o o mo o o f the

TABLE 5. Study of Variation among Analysts and among

^a Measured by FI–HG–AAS. ^b Calculation not done as some of the values are $<50 \ \mu g/L$. ^c NS = not significant, S = significant.

in evaluating the performance of field kits. The two field kits (NIPSOM and GPL) were considered.

Table 5 reveals that there is no significant variation among the analysts whereas variation is significant among the replicates in most of the cases. The analyst variability defined in the UNICEF samples utilizing the AIIH&PH kit by groups trained under variable conditions was not confirmed in the NIPSOM and GPL results obtained by skilled analysts but inconsistency of the test kits was found in each group.

460

461

462

463

464

465

466

467

468

469

470

471

472

473

474

475

476

477

478

479

480

481

482

483

484

485

486

487 488

489

490

491

492

493

494

Analysis of 2866 Hand Tube Wells by FI-HG-AAS Previously Colored Red (Unsafe)/Green (Safe) after Field Kit Testing by Various Organizations in Bangladesh. To judge the accuracy of well markings, SOES, jointly with DCH, Bangladesh, went to the field and collected water samples from 2 866 hand tube-wells previously colored red or green on the basis of kit testing by field workers. FI-HG-AAS analysis of 1 143 tube wells colored green by field kits confirmed that 1 058 tube wells had arsenic concentrations below 50 μ g/L but found that 85 (7.5%) were unsafe. Of 1 723 tube wells colored red by field kits, only 861 were found unsafe (>50 μ g/L) and the remaining 862 were safe (<50 μ g/L) by our FI-HG-AAS system. The field kits had mislabeled 862 + 85 = 947 (33.04%) of 2 866 hand tube wells. Given the scarcity of uncontaminated water the mislabeling of 50% of safe wells has a major socioeconomic impact.

The FI–HG–AAS results for 2 866 samples were compared with the status (green or red) of tube wells measured by field kits at concentrations ranging from $<3 \mu g/L$ to $<600 \mu g/L$. Table 6 indicates that 1 920 tube wells were safe, although the kits had mislabeled 862 (44.9%) as unsafe. The overall results of 2 866 field sample analyses (Table 6) show that at arsenic concentrations $<3 \mu g/L$ to $<70 \mu g/L$, field kits are not reliable. However from 70 $\mu g/L$ to $<600 \mu g/L$ the false detection by field kits is only 4.17% to 9.59%.

495 From Table 6, it appears that field kit results are mainly unreliable up to $<70 \mu g/L$ of arsenic. British Geological Survey 496 has analyzed from Bangladesh 3 534 hand tube wells (14) by 497 498 reliable instrumental system in the laboratory. We have 499 calculated from BGS result that 2 804 (79.3%) out of 3 534 samples contain arsenic $\,{<}70$ $\mu g/L$. We too have analyzed 500 34 000 hand tube well water samples by FI-HG-AAS from 501 502 64 districts of Bangladesh. Out of these 34 000 samples, 22 000 samples were from 31 districts where we have found arsenic 503 504 patients and 65.1% samples contain out of 34 000 arsenic <70 μ g/L. From West Bengal, we have so far analyzed more 505 than 100 000 samples by the same FI-HG-AAS method and 506 507 79.2% contain arsenic < 70 μ g/L. The average of these results

TABLE 6. Comparative Study of Field Kit with FI-HG-AAS from 2866 Field Samples

range in μg/L	samples analyzed by FI—HG—AAS	no. of samples below 50 μg/L	no. of samples above 50 µg/L	false detection (%)
<3	665	267	398	59.85
3-<10	508	417	91	17.91
10-<20	259	181	78	30.12
20-<30	210	121	89	42.38
30-<40	168	51	117	69.64
40-<50	110	21	89	80.91
below 50	1920	1058	862	44.90
50-<60	95	15	80	15.79
60-<70	87	20	67	22.99
70-<80	73	7	66	9.59
80-<90	73	7	66	9.59
90-<100	67	5	62	7.46
100-<200	358	23	335	6.42
200-<300	97	3	94	3.09
300-<400	51	3	48	5.88
400-<500	21	1	20	4.76
500-<600	24	1	23	4.17

indicated that 74.5% of the total hand tube wells in Bangladesh and West Bengal together contain arsenic $<70 \ \mu g/L$. If the total number of hand tube wells in Bangladesh are between 6 and 11 million (*14*) and that of 9 arsenic affected districts of West Bengal are 1.5–2.0 million (*34*), then according to Table 6, 74.5% of hand tube wells are in critical level according to field kit assessment.

Significance of the Results

The weak correlation's between the kits and laboratory methods (Table 2 with Figure 6a,b and Table 3 with Figure 8a–d) indicates the random behavior of the kits. Though negative values are the indication of inverse correlation but at this level it does not bear any significance. The random behavior of the kits is also reflected in Table 6, where performance of kit was appreciably improved above 70 μ g/L. But it cannot be recommended to use the kits above 70 μ g/L if we examine Tables 2 and 3. Considering the behavior of the kits, it may be confidently mentioned that the kit is not at all quantitative and at best qualitative.

The most important limitation of using these field kits is visual identification of the color in the lower range. The identification of color or judgment in the lower range also varies from man to man. The persons with developed expertise through practice may not be engaged later on for estimation. As a result, the accuracy of the results suffers when the estimation is done by new hands as is generally the case. Several attempts have been made to develop different field kits by different manufacturers to quantify arsenic concentration even up to 10 μ g/L levels. However none of the field kits manufacturer reported their field kit efficiency in different arsenic concentration level after comparing with large number of field samples measured by highly sensitive instrumental techniques. Quantification as needed in the lower range was not checked.

Cost is an important consideration but requires comparison with the even higher cost of falsely labeling a well as unsafe. The average cost of each field kit is around U.S. \$40 for 100 determinations. The salary and maintenance expenses of the field analysts increases the cost of kit testing to U.S. \$2 plus organizational and training expenses and data analyses. Our experience of around 2 00 000 water and biological samples analyzed for arsenic by FI–HG–AAS has established (*35*) that a totally reliable assay can be completed in 30–40 s at a cost even lower than that of the field kit. The amount of chemicals to be used for testing kits is not also

515

516

517

518

519

520

521

522 523

524

525

526

527

528

529

530

531

532

533

534

535

536

537

538

539

540

541

542

543

544

545

546

547

548

549

550

551

552

508

509



FIGURE 8. (a) Outliers in the correlation between AAS and Merck (doubling method) kit values at the range of $50.1-100 \mu g/L$ of arsenic (some points are overlapping each other). (b) Outliers in the correlation between AAS and AAN kit values at the range of $50.1-100 \mu g/L$ of arsenic (some points are overlapping each other). (c) Outliers in the correlation between AAS and Merck (doubling method) kit values above 100 $\mu g/L$ of arsenic. (d) Outliers in the correlation between AAS and AAN kit values above 100 $\mu g/L$ of arsenic.

568

569

553

negligible. For testing of 100 samples by field kits (average of NIPSOM, GPL and Merck), the amount of concentrated HCl required will be 150 mL and the mixture of Zn, KI and SnCl₂ is about 65 g (Table 1), whereas by FI-HG-AAS, the amount of chemicals required will be about 50 mL of acid and a mixture of 2.0 g of NaBH₄ and NaOH. If 10 million tube wells are to be tested by field kits, this will be a huge amount compared to FI-HG-AAS. The environmental contamination of the field kits is a real concern not evoked by FI-HG-AAS: in a micro-assay waste chemicals are minimal and the laboratory personnel are not exposed to arsine gas. Any field kit used in arsenic affected areas must be more reliable and sensitive than existing methods, otherwise the 'bad' kit data mixed with 'good' instrumental data contaminates the entire data pool for meaningful policy decision. No cost benefit can be attributed to field kits that waste water resources and fail to eliminate real hazards.

570 Acknowledgments

571 We thank to BRAC, Bangladesh for permission to use their 572 comparative study of field kits and continuous hydride generation AAS data for statistical analysis. Thanks to UNICEF, Calcutta for sending their field kit samples to us for validation.

573

574

575

576

577

578

579

580

581

582

583

584

585

586

587

588

589

590

591

592

593

594

Literature Cited

- Chowdhury, U. K.; Biswas, B. K.; Roy Chowdhury, T.; Samanta, G.; Mandal, B. K.; Basu, G. K.; Chanda, C. R.; Lodh, D.; Saha, K. C.; Mukherjee, S. C.; Roy, S.; Kabir, S.; Quamruzzaman, Q.; Chakraborti, D. *Environ. Health Perspect.* **2000**, *108* (5), 393– 397.
- (2) Smith, A. H.; Lingas, E. O.; Rahman, M. Bull. World Health Org. 2000, 78(9), 1093–1103.
- (3) Ahsan, H.; Perrin, M.; Rahman, A.; Parvez, F.; Stute, M.; Zheng, Y.; Milton, A. H.; Brandt-Rauf, P.; Geen, A. V.; Graziano. J. J. Occup. Environ. Med. 2000, 42(12), 1195–1201.
- (4) Tondel, M.; Rahman, M.; Magnuson, A.; Chowdhury, I. A.; Faruquee, M. H.; Ahmad, S. A. *Environ. Health Perspect.* 1999, 107(9), 727–729.
- (5) Bagla, P.; Kaiser, J. Science 1996, 274, 174-175.
- (6) Rahman, M. M.; Chowdhury, U.K.; Mukherjee, S. C.; Mondal, B. K.; Paul, K.; Lodh, D.; Biswas, B. K.; Chanda, C. R.; Basu, G. K.; Saha, K. C.; Roy, S.; Das, R.; Palit, S. K.; Quamruzzaman, Q.; Chakraborti. D. *J. Toxicol. Clin. Toxicol.* **2001**, *39*(7), 683–700.

(7) Chakraborti, D.; Rahman, M. M.; Chowdhury, U.K.; Paul, K.; Sengupta, M. K.; Lodh, D.; Chanda, C. R.; Saha, K. C.; Mukherjee, S. C. Talanta 2002, 58(1), 3-22.

595

596

597

598 599

600

601

602

603

604

605

606

607

608

609

610

611

612

613

614

615 616

617

618

619

620

621

622

623

624

625

626

627

628

629

630

631

632

633

634

635

636

637

638

639 640

- (8) Chowdhury, U. K.; Rahman, M. M.; Mondal, B. K.; Paul, K.; Lodh, D.; Biswas, B. K.; Chanda, C. R.; Basu, G. K.; Saha, K. C.; Mukherjee, S. C.; Roy, S.; Das, R.; Kaies, I.; Barua, A. K.; Palit, S. K.; Quamruzzaman, Q.; Chakraborti, D. Environ. Sci. 2001, 8(5), 393-415.
 - Roy Chowdhury, T.; Basu, G. K.; Mandal, B. K.; Biswas, B. K.; (9)Samanta, G.; Chowdhury, U.K.; Chanda, C. R.; Lodh, D.; Roy, S. L.; Saha, K. C.; Roy, S.; Kabir, S.; Quamruzzaman, Q.; Chakraborti, D. Nature 1999, 401, 545-546.
 - Mandal, B. K.; Roy Chowdhury, T.; Samanta, G.; Mukherjee, D. P.; Chanda, C. R.; Saha, K. C.; Chakraborti, D. *Sci. Total Environ.* (10)1998, 218, 185-201.
- (11) Economic and Social Commission for Asia and the Pacific; Geology and Health: Solving the Arsenic Crisis in the Asia Pacific Region. ESCAP-UNICEF-WHO Expert Group Meeting, Bangkok, May 2-4, 2001.
 - (12) Tandukar, N. Abstracts; International Conference on Arsenic in the Asia-Pacific Region: Managing Arsenic for our Future, Adelaide, Australia, November 20-23, 2001, p103-105.
- Berg, M.; Tran, H. C.; Nguyen, T. C.; Pham, M. V.; Schertenleib, (13)R.; Giger, W. Environ. Sci. Technol. 2001, 35(13), 2621-2626.
- (14) Arsenic Contamination of Groundwater in Bangladesh; BGS Technical Report WC/00/19; British Geological Survey: Keyworth, UK, 2001.
- International Conference on Arsenic Pollution of Groundwater (15)in Bangladesh: Causes. Effects and Remedies. LGED Auditorium, Dhaka, Bangladesh, February 8-12, 1998.
- (16) Pearce, F. Death and The Devil's Water. New Scientist, London September 16, 1995, 14-15.
- (17)Chowdhury, A. Q. Abstracts; International workshop on Arsenic Mitigation, Local Government Division, Ministry of Local Government, Rural development and Cooperatives, Government of Bangladesh: Dhaka, Bangladesh, January 14-16, 2002, p 50-53.
- (18) DPHE-UNICEF Monthly Progress Report, Dhaka, Bangladesh, December 2001
- (19) Das, R.; Rahman, M.; Quamruzzaman, Q.; Yusuf, J.; Mostafa, M. G.; Mohonta, R.; Basit, S. M.; Mannan, M. A. Dhaka Community Hospital Bulletin, Dhaka, Bangladesh, July 2002.
- (20)Half Yearly Report, NGO Forum for Drinking Water Supply & Sanitation, Dhaka, Bangladesh, January 2002.
- Fact Sheet 12 on Arsenic: A Disaster Forum Publication, Dhaka, Bangladesh (e-mail: df@bangla.net).

- (22) Combating a Deadly Menace: Early Experiences with a Community-Based Arsenic Mitigation Project in Bangladesh, June 1999-June 2000, BRAC Research Monograph, Series No. 16, August 2000.
- (23) Joint Plan of Action: Arsenic Contamination of Drinking Water: Government of West Bengal and UNICEF, Nodal Department: Public Health Engineering Department, 1999.
- Arsenic exposure and Health effects; Abernathy, C. O., Calderon, (24)R. L., Chappell, W. R., Eds.; Chapman & Hall: London, UK, 1997.
- (25) Hussam, A.; Alauddin, M.; Khan, A. H.; Rasul, S. B.; Munir, A. K. M. Environ. Sci. Technol. 1999, 33(20), 3686-3688.
- (26) Chakraborti, D.; Valentova, M.; Sucha, L. Anal. Chem. 1982, H-17. 31-41.
- Chatterjee, A.; Das, D.; Mandal, B. K.; Chowdhury, T. R.; Samanta, (27)G.; Chakraborti, D. Analyst 1995, 120(3), 643-650.
- Samanta, G.; Roy Chowdhury, T.; Mandal, B. K.; Biswas, B. K.; (28)Chowdhury, U.K.; Basu, G. K.; Chanda, C. R.; Lodh, D.; Chakraortri, D. Microchemical J. 1999, 62, 174-191.
- Das, D., Chatterjee, A., Mandal, B. K., Samanta, G., Chanda, B.; (29)Chakraborti, D. Analyst 1995, 120(3), 917-924.
- (30)Shresetha, R. R. Environment & Public Health Organization (ENPHO), Kathmandu, Nepal,
- (31) Samanta, G.; Chakraborti, D. Frenius J. Anal. Chem 1997, 3577 (7), 827 - 832.
- (32) Roy Chowdhury, T. Groundwater Arsenic Contamination in West Bengal, India: Characterisation and Chemical Analysis of Borehole Sediment Samples, Rotoevaporated Arsenic Rich Water Residues and Present Status of Three Arsenic Affected Blocks of 24-Parganas (North), Doctoral Dissertation, Jadavpur University, Calcutta, India, February 1999.
- Personal Communication, Alauddin, M. Chemistry Department, (33)Wagner College, City University of New York, USA
- Groundwater Arsenic Contamination Status of Kalvani Mu-(34)nicipal Area, Nadia. A report by School of Environmental Studies, Jadavpur University, Calcutta-700 032, India, June 2002.
- Comparative Study of Field Testing Kits with FI-HG-AAS. A report by School of Environmental Studies, Jadavpur University, Calcutta-700 032, India, September 1999.

Received for review February 12, 2002. Revised manuscript received September 23, 2002. Accepted September 25, 2002. 681

ES020591O

682

641

642

643

644

645

646

647

648

649

650

651

652

653

654

655

656

657

658

659

660

661

662

663

664

665

666

667

668

669

670

671

672

673

674

675

676

677

678

679