

1 **Predicting groundwater arsenic contamination in Southeast Asia**
2 **from surface parameters**

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13 2nd revision submitted to *Nature Geoscience*
14 (Manuscript NGS-2008-01-00049A)

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16 June 16, 2008

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18 **Arsenic contamination of groundwater resources threatens the health of**
19 **millions of people worldwide, particularly in the densely populated river deltas**
20 **of Southeast Asia. Although many arsenic-affected areas have been identified in**
21 **recent years, a systematic evaluation of vulnerable areas remains to be carried**
22 **out. Here we present maps pinpointing areas at risk of groundwater arsenic**
23 **concentrations exceeding 10 µgL⁻¹. These maps were produced by combining**
24 **geological and surface soil parameters in a logistic regression model, calibrated**
25 **with 1756 aggregated and geo-referenced groundwater data points from the**
26 **Bengal, Red River and Mekong deltas. We show that Holocene deltaic and**
27 **organic-rich surface sediments are key indicators for arsenic risk areas and that**
28 **the combination of surface parameters is a successful approach to predict**
29 **groundwater arsenic contamination. Predictions are in good agreement with the**
30 **known spatial distribution of arsenic contamination and further indicate**
31 **elevated risks in Sumatra and Myanmar where no groundwater studies exist.**

32

33 More than 100 million people worldwide ingest excessive amounts of arsenic (As)
34 through drinking water contaminated from natural geogenic sources. Many Asian
35 countries, in particular, are known to be affected by high groundwater As
36 concentrations as a result of chemically reducing aquifer conditions: Bangladesh¹⁻¹⁰,
37 India^{3,11,12}, China^{13,14}, Nepal¹⁵, Cambodia¹⁶⁻¹⁸ and Vietnam^{17,19,20}. However, since As
38 analysis is expensive and time-consuming, groundwater resources of many regions
39 still remain to be tested. Therefore, maps pinpointing areas vulnerable to As
40 contamination can guide households at risk of arsenic contamination, as well as
41 scientists and policy-makers to initiate early mitigation measures and protect the
42 populations from chronic As poisoning.

43

44 Though the exact chemical conditions and reactions leading to As mobilization are still
45 under debate, it is generally accepted that microbial and/or chemical reductive
46 dissolution of As-bearing iron minerals in the aquifer sediments^{1,4,21} is the main cause
47 for the release of As. Reducing conditions are often associated with the presence of
48 natural (bio)degradable organic carbon embedded in sediments^{9,11,22-25}. Other
49 identified key characteristics of contaminated areas are rapidly buried, young
50 (Holocene) sediments and low hydraulic gradients in flat and low-lying areas^{8,18,26,27}.
51 Ideally an As prediction model for groundwater should be based on parameters that
52 indicate the key characteristics mentioned above in three dimensions. However, in the
53 absence of a 3-dimensional spatially continuous database of aquifer conditions to
54 depth, globally and regionally available (two dimensional) surface parameters can be
55 used as indicators for As-enrichment in underlying aquifers^{28,29}.

56

57 In the past, several geostatistical interpolation methods (e.g. kriging) have been used
58 to predict elevated As in groundwater on a regional scale³⁰⁻³². However, for predictions

59 of areas where no groundwater quality data exist, interpolation methods are not
60 applicable³² and models based on logistic regression are more appropriate^{33,34}. In an
61 expert-based statistical model to delineate areas at risk of groundwater As
62 contamination on a coarse global scale, we found that geological information was of
63 crucial importance²⁹. Here we focus on an in-depth assessment of depositional
64 environments in Southeast Asia. We use a logistic regression approach based on
65 relationships between sedimentary information, soil maps and measured groundwater
66 As data of Bangladesh, Cambodia and Vietnam, to assess the relative importance of
67 the different surface proxies in these countries. We apply these relationships to set up
68 prediction maps of As contamination in Southeast Asia including Indonesia (Sumatra)
69 and other countries where groundwater quality data is scarce (Myanmar and Thailand).
70 Furthermore, we verify the predicted risk in South Sumatra, where the groundwater
71 has not previously been tested for the presence of As.

72

73 **ARSENIC PREDICTION MODEL**

74 Our model is based on three assumptions. First, sedimentary depositional
75 environments are characterized by a unique combination of chemical, physical, and
76 biological properties³⁵ and can serve as indicators (proxies) for chemical and physical
77 conditions of the aquifers beneath the surface. Second, soil properties are proxies for
78 present and past drainage conditions and they are also indicators of recent
79 depositional environments. Third, soil textures, for example clay and silt are proxies
80 for the chemical maturity of the sediments, where clay is more mature than silt. An
81 important factor in the development of soil textures is topography³⁶, which allows the
82 delineation of areas where the model is applicable.

83

84 GIS-datasets were established from digital elevation data, countrywide geological
85 maps and global soil data (FAO), which were converted to a raster format using
86 ArcGIS (ver. 9.2). An overview of GIS data used in this study is provided in Table S1
87 (see Supplementary Information). Because each geological map applied a different
88 classification terminology, we created an uniformly classified geological map for all
89 regions (Figure 1). Although Bangladesh does geographically not belong to Southeast
90 Asia, it was included in the model because of the large number of available data
91 points⁵. Statistical relations between As concentrations and 30 parameters related to
92 soil properties, geology, climate, and hydrology (Table S2) were initially evaluated by
93 stepwise regression. The six parameters exhibiting a significance >95% and two
94 additional soil parameters were employed in the final model (see Methods and Table
95 1). Since young geological deposits and As groundwater contamination are rarely
96 observed in areas with steeper slopes, and groundwater As concentration data are
97 only available for regions with a flat topography (slope $<0.1^\circ \approx 0.17\%$), areas with
98 slopes $>0.1^\circ$ were excluded from the model (see Supplementary Figure S1).

99
100 The prediction required three steps: i) Aggregation and binary-coding of measured As
101 concentrations to reduce spatial heterogeneities (dependent variables), ii) Logistic
102 regression to obtain weighting coefficients of independent variables (see Methods
103 section and corresponding results in Table 1), and, iii) Calculation of the probability of
104 As contamination based on the threshold value of $10 \mu\text{gL}^{-1}$. The spatial datasets
105 considered as independent variables for the model are topographic data to delineate
106 the model area, sedimentary depositional environments as a proxy for aquifer
107 conditions, and soil variables as a proxy for drainage and chemical maturity of
108 sediments (see the Methods section).

109

110 **SURFACE PARAMETERS CONTRIBUTING TO THE MODEL**

111 The weighting factors (λ) and significance of the eight independent variables retained
112 in the final model are listed in Table 1. In general, variables describing sedimentary
113 depositional environments have a larger contribution to the model than soil variables,
114 presumably because geological variables come closest to describing As
115 contamination in the aquifer itself. The presence of young deltaic deposits ($\lambda=1.65$) is
116 a particularly significant indicator for As-contaminated aquifers. In Southeast Asia,
117 delta initiation and progradation occurred simultaneously with the Holocene Climate
118 Optimum³⁷. Therefore, delta progradation resulted in the burial of organic matter at a
119 high rate. The presence of relatively fresh organic carbon provides favourable
120 conditions to establish reducing environments, which may lead to As enrichment in
121 groundwater. Logistic regression confirms that organic-rich deposits ($\lambda=1.11$) play an
122 important part in the model. Recent alluvial deposits ($\lambda=0.55$) are also indicative for
123 elevated As concentrations in groundwaters (Table 1). Of the soil parameters, medium
124 textured soils seem to be indicative of As-bearing aquifers evolved from rapid
125 accumulation of young (Holocene) sediments. In contrast, the negative weighting
126 coefficient ($\lambda= -0.95$) for floodplain deposits implies that these fine-grained deposits
127 could overlie aquifers low in dissolved As (compare Figures 1 and 3). Fine-grained
128 deposits (high clay content) thereby point to low-energy depositional environments
129 with condensed sediments where Holocene aquifers are rare and where groundwater
130 is likely drawn from older aquifers.

131
132 Pre-Holocene deposits, other Holocene deposits and tidal deposits (Figure 1) were
133 found to be statistically insignificant ($p >0.05$) and they were excluded from the model
134 in the first stepwise regression (see Table S2 and Table 1). Tidal sediments are

135 generally associated with aquifers abundant in sulfate that may be microbially reduced
136 to sulfide and re-precipitate As^{38,39}. They might serve as proxies for low-risk areas, but
137 our As data in such aquifers were possibly too few to see such a relation.

138

139 **INFERRING DEPOSITIONAL ENVIRONMENTS AT DEPTH FROM THE SURFACE**

140 As mentioned in the introduction, our model is based on two- dimensional data (i.e.
141 surface maps). Nevertheless, geological information (sedimentary depositional
142 environments) inherently contains a three-dimensional component. The recent
143 sedimentary history of major Southeast Asian basins is characterized by delta
144 initiation, which occurred on a global scale at about 8500-6500 years BP and was
145 principally controlled by the deceleration of sea-level rise⁴⁰. Delta progradation
146 resulted in the unconformable deposition of thick late Pleistocene-Holocene sediments
147 on Pleistocene and older sediments, e.g. as incised-valley-fill deposits^{8,18}. As-
148 contaminated aquifers are mainly present in these Holocene aquifers, whereas deeper
149 Pliocene-Pleistocene aquifers are, to a large extent, free of As^{7,8,26}. The boundary
150 between Pleistocene and Holocene sediments is not located at a constant depth⁸ and
151 this can lead to misclassifications, since our model inherently assumes that the
152 underlying aquifer belongs to the same sedimentary depositional environment as on
153 the surface.

154

155 Two situations exist where the environment at the surface does not reflect the geology
156 at depth, i) when the As measurements used in the model were obtained from
157 tubewells tapping deep (Pleistocene) aquifers because shallow (Holocene) aquifers
158 are not present, or shallow groundwater is too saline for consumption (e.g. in coastal
159 regions), and ii) when Holocene sediments were deposited at sedimentation rates too
160 small to form a usable aquifer. In both situations Holocene depositional environments

161 are present at the surface and high-risk areas are indicated, although measured As
162 concentrations are low (false positive cases). Even though the probability maps would
163 be improved by including the thickness of Holocene sediments, the absence of
164 country-wide three-dimensional geological data rules this out. Furthermore, the
165 complexity of aquifer heterogeneity at a local scale makes it inevitable that
166 misclassifications occur.

167

168 **PROBABILITY MAPS**

169 Predicted areas at risk of As contamination agree well with known spatial
170 contamination patterns, a finding which is supported by the results of the model
171 classification indicating the performance of model prediction (Figure 2) and the
172 Hosmer-Lemeshow goodness-of-fit statistics (Table S3). An absolute average
173 deviation of 7.3% is found between expected and modelled probabilities of As being
174 ≤ 10 or $>10 \mu\text{gL}^{-1}$. The model is further characterised by the receiver operating
175 characteristics curve area of ~ 0.7 (Figure S2), which is a good result considering that
176 neither depths of analyzed groundwater wells, nor aquifer hydrological data form part
177 of the model.

178

179 The probability maps for As concentrations exceeding $10 \mu\text{gL}^{-1}$ are presented in
180 Figure 3 and supplementary Figure S3 (probability map of whole Southeast Asia). The
181 highest probabilities (0.7-0.8) for As contamination ($>10 \mu\text{gL}^{-1}$) are found in the south-
182 central part of Bangladesh, with a value of 0.5-0.6 in the north-eastern Sylhet basin
183 (Figure 3a). The probability of finding contaminated wells in the Red River delta
184 reaches a value of 0.7 (Figure 3e). The sedimentary depositional environments
185 present along the Mekong river differ from the deltaic environments of Bangladesh
186 and the Red River delta in that organic-rich deposits are found at close distance of the

187 modern Mekong and Bassac river courses, surrounded by extensive floodplain areas
188 free of As¹⁸ (compare Figures 1 and 3). The probability map for the Mekong delta
189 shows values of up to 0.6 at close distance to the modern Mekong and Bassac river
190 courses and adjacent swampy marshes (Figure 3c). In addition, the large floodplain of
191 Lake Tonle Sap with organic-rich sediments is a risk area with probabilities ranging
192 between 0.4-0.6.

193
194 To compare our predicted low-risk (probability ≤ 0.4) and high-risk areas (probability
195 > 0.4) (Figures 3b and 3d) with measured As contamination, a misclassification
196 analysis was performed based on the 1756 aggregated and binary-coded (≤ 10 or > 10
197 μgL^{-1}) As measurement data for the three deltas. We report 71% (1023 aggregated
198 points), 59% (107 a.p.), and 75% (100 a.p.) of correctly classified cases for
199 Bangladesh, Red River and Mekong deltas respectively (average 70%). In our study,
200 the Red River and Mekong deltas have 6 and 9% false negative cases. In Bangladesh
201 false negative cases (13%) were found specifically in wells lying at a close distance to
202 rivers. The number of false negatives is outnumbered by false positives in all the three
203 deltas with 17, 35 and 16% for Bangladesh, the Red River and Mekong deltas,
204 respectively. Errors in the prediction can arise from the commonly reported well-to-well
205 variability where wells with low As levels are often present at close distance from wells
206 high in arsenic^{5,6,18,19}, as well as from uncertainties in measured As (estimated at 25%)
207 leading to misclassifications of concentrations being close to the threshold of $10 \mu\text{gL}^{-1}$.
208 However, we interpret these misclassifications to be mainly an effect of modelling
209 three-dimensional processes based on two-dimensional data.

210

211 Apart from the three deltas discussed above, our Southeast Asia probability map
212 (supplementary Figure S3) also highlights risk areas that are largely unknown or
213 unreported, particularly in Sumatra, Myanmar and Thailand (Figure 3f).

214

215 **VERIFICATION OF PREDICTIONS FOR SUMATRA**

216 According to the modelled probability, an area of about 100,000 km² at Sumatra's east
217 coast is prone to high risk of As contamination (probability >0.4) (see Figure 4a and
218 supplementary Figure S3). To validate the Sumatra prediction map, 97 groundwater
219 samples were collected in 2007 in the Province of South Sumatra in the vicinity of
220 Palembang (see Methods section). This area was chosen because it is at the border
221 of a low- and high-risk area and not previously studied. Since there is no large
222 variation in geological and topographical features along Sumatra's east coast, the
223 study area is representative of the whole high risk area. Figure 4b shows As
224 concentrations ($\leq 10 \mu\text{gL}^{-1}$ and $> 10 \mu\text{gL}^{-1}$) measured in the groundwater, imposed on
225 the binary probability map. The classification results for Sumatra (63% correctly
226 classified, 36% false positive and 1% false negative) are comparable to those of the
227 Bangladesh, Red River, and Mekong deltas. In total, 94% of the 50 tubewells located
228 in the low-risk area have As levels below $10 \mu\text{gL}^{-1}$. Of the 12 contaminated wells (As
229 $> 10 \mu\text{gL}^{-1}$) 75% are positioned in the high-risk area. However, in the high-risk area the
230 contaminated wells are clearly outnumbered by uncontaminated groundwater
231 measurements (9 contaminated wells vs. 38 uncontaminated wells), for reasons
232 explained below.

233

234 On average, both high and low risk areas in Sumatra are characterized by high DOC,
235 NH_4^+ , HCO_3^- , PO_4^{3-} , and Fe(II) and low SO_4^{2-} concentrations (Supplementary Table
236 S5). At least two-thirds of the sampled groundwaters are reducing in nature, especially

237 those located in the high-risk area. Since the chemical conditions of the aquifers would
238 permit the reductive dissolution of As, other explanations for the overall low As
239 concentrations must be considered.

240
241 To gain a greater insight into the characteristics of the aquifer, the local geology was
242 examined laterally and in depth. The studied area is geologically young (Tertiary-
243 Quaternary) (Figure 4c). The sediment contains deposits from peat swamp forests that
244 developed during the Holocene era (past 5,000 to 10,000 years) and unconformably
245 overlie older sediments⁴¹. The outline of the high-risk As-contamination zone mainly
246 follows the outline of these deposits. The peat deposits are usually found ranging from
247 4-8 meters although depths of up to 24 metres have been reported⁴². However, the
248 depths of sampled tubewells in Sumatra average 46 meters, which implies that most
249 of them tap groundwaters from aquifers below the Holocene peat deposits. This
250 shows that the prediction map is a useful tool for the identification of areas at risk of
251 As contamination, but that understanding the local geology as a function of depth is of
252 vital importance for specific areas.

253

254 **ARSENIC CONTAMINATION IN THAILAND AND MYANMAR**

255 The probability map in Figure 3f shows that the Chao Phraya basin in central Thailand
256 and the Irrawaddy delta in Myanmar have a risk of elevated As being present in
257 groundwater, whereas the Sittang basin (Myanmar) has a lower risk, and the Salween
258 basin (Myanmar) has virtually no risk at all. The problem of As contamination in the
259 Irrawaddy delta is partly known⁴³ although its spatial extent has not been investigated
260 to date, which is particularly worrying considering the size of the area at risk. In the
261 Chao Phraya basin in central Thailand, a groundwater survey was undertaken in 2001
262 using As field test kits to test wells with minimum depths of 80 m⁴⁴. The range of

263 measured As concentrations in the 37 tested wells was $<1 \mu\text{gL}^{-1}$ to $100 \mu\text{gL}^{-1}$ with an
264 average of $11 \mu\text{gL}^{-1}$. These results correlate with our maps that predict a low to
265 moderate As contamination. Indeed, North-South geological profiles across the
266 basin⁴⁵ indicate maximal depths of 20 m for the pre-Holocene incisions implying that
267 sampled tubewells draw water from Pleistocene or older aquifers.

268

269 In contrast to the slow sedimentation rates of the Sittang and Chao Phraya deltas, the
270 Irrawaddy delta received massive amounts of sediments during the Cenozoic era⁴⁶.
271 The Irrawaddy River⁴⁷ still has a 10 times larger sediment load than the Chao Phraya
272 River⁴⁵. In 2002 the Departments of Medical Research and Health (Lower Myanmar),
273 financially supported by UNICEF, conducted a groundwater sampling campaign in the
274 Irrawaddy division⁴³. In total, 99 groundwater samples (90 shallow tubewells and 9
275 dug wells) were collected in 25 villages, and As was quantified by atomic absorption
276 spectroscopy. It was reported that 67% of the sampled wells had As levels $>50 \mu\text{gL}^{-1}$.
277 These results show that the risk of As contamination in the Irrawaddy delta, as
278 indicated by the probability map, should be taken seriously and that there is an urgent
279 need to test shallow tubewells in other townships or districts in the high risk area.

280

281 **ASSESSING ARSENIC ELSEWHERE**

282 In the study presented here, we identified regions in Southeast Asia, based on As
283 prediction maps, where tubewells should be tested for elevated As concentrations
284 ($>10 \mu\text{gL}^{-1}$). Although the use of such maps in other scientific investigations (e.g.
285 climate research) is a common practice, the prediction of As (and other groundwater
286 conditions) is still an emerging technique in the field of natural groundwater
287 contaminants. Our As model differs from earlier models in its ability to predict
288 contamination in areas of unknown groundwater quality and on a sub-continental

289 scale. The strength of the prediction lies rather in the combination of surface
290 parameters than in the individual parameters. As an example, the Sittang basin and
291 the vicinity of the lower Mekong river branches are both characterized by alluvial
292 deposits, but only the latter has a modelled high-risk because of the contribution of soil
293 properties. A limitation to be considered is that shallow, As-bearing groundwater can
294 only be expected where Holocene aquifers are present. Where this is not the case,
295 high risk areas may indicate the presence of reducing aquifers, but As concentrations
296 in groundwater could be lower than predicted.

297

298 Our approach provides a blueprint for further modelling and mapping of As-tainted
299 aquifers in and outside of Southeast Asia. The probability maps can further be
300 improved when data with higher spatial resolution or in three dimensions becomes
301 available, although we emphasize that it will not be possible to account for the local
302 heterogeneities of aquifers. The presented prediction maps are a valuable and
303 resource-saving tool that can serve both scientists and policy-makers to initiate early
304 mitigation measures in order to protect the people from As-related health problems as
305 well as to efficiently guide water resources management.

306

307 **METHODS**

308 Geological maps of Bangladesh, Cambodia, Thailand and Vietnam were available in
309 digital format (Supplementary Table S1). Maps of Myanmar and Sumatra were
310 digitized for this study. Geological maps of Malaysia and Laos were not available. The
311 sedimentary depositional environments employed in the model as independent
312 variables are deltaic deposits, organic-rich sediments (e.g. sediments deposited in
313 marshy environments), floodplains, alluvial deposits, tidal deposits, other Holocene
314 sediments and pre-Holocene sediments. Soil variables are percentages of silt, clay

315 and sand in both the topsoil (0-30 cm) and subsoil (30-100 cm), and coarse, medium
316 and fine soil textures.

317
318 We compiled >4600 data points of groundwater As-concentrations from Bangladesh
319 (median well depth 35 m), the Mekong delta (Cambodia and Southern Vietnam, m.w.d.
320 39 m) and the Red River delta (Northern Vietnam, m.w.d. 30 m). The data originates
321 from BGS and DPHE (Bangladesh, n=3534)⁵, from Buschmann et al. (Mekong delta,
322 n=352)^{18,20}, and from Berg et al. (Red River delta, n=720)^{17,25,48} and was used without
323 a restriction in well depths. To test the model, 97 tubewell were randomly sampled for
324 this study in Sumatra at about 5-10 km intervals at an average sampling density of 1
325 sample per 54 km². This study area is positioned at a latitude of 2°872S to 3°911S
326 and a longitude of 103°949E to 104°993E (sampling area 85 km by 65 km).

327 Procedures of sampling and analysis were carried out as described in Buschmann et
328 al.¹⁸. Concentrations of As and additional parameters measured in Sumatra
329 groundwater samples are provided in Table S5 (Supplementary Information).

330
331 Arsenic concentrations are point measurements within vertical depths of the wells,
332 while the other variables have coarser spatial resolution being generally greater than
333 30 arc seconds. Point data of measured As-concentrations were therefore aggregated
334 using the geometric mean to a resolution of one point per pixel with a size of 5 arc
335 minutes (~9.3 km at the equator), which is the pixel size of the global soil data (FAO).
336 Aggregation resulted in a decreased dataset of 1756 pixel-based data points
337 (Bangladesh 1443, Red River Delta 180, and Mekong delta 133 points). The
338 aggregated point-data were binary-coded using the WHO guideline value for As in
339 drinking water (10 µgL⁻¹) as a threshold. We acknowledge that several countries apply
340 a guideline of 50 µgL⁻¹, but adopting this threshold would result in significantly less

341 data points (992) for the calibration of the model. The binary variable of whether As
342 concentrations exceed the WHO threshold (1) or not (0) was used as the dependent
343 variable in this study.

344

345 Logistic regression was used to determine the weighting of the independent variables.
346 This is a common statistical method used in environmental research and allows for the
347 concurrent use of continuous and categorical variables^{33,34}. The parameter P denotes
348 the probability of As concentrations exceeding the WHO threshold. Logistic regression
349 models log(odds), which is defined as the ratio of the probability that an event occurs
350 to the probability that it fails to occur $\log(P/(1-P))$, as a linear combination of
351 independent variables⁴⁹.

$$352 \log(odds) = C + \sum_{i=1}^n \lambda_i X_i \quad (1)$$

353 where C is the intercept of regression, X_i are independent variables, and λ_i are the
354 weighting coefficients that were obtained using the maximum likelihood procedure⁴⁹.

355 Exponential values of coefficients, Wald statistics, and p-values (see Table 1) indicate
356 the importance of each variable. Statistically insignificant independent variables were
357 excluded from the model during each of the subsequent regression steps (Table S2).

358 The threshold for maintaining a variable in the model was determined by the 95%
359 significance level ($p < 0.05$). The silt contents in the subsoil and medium textured soils
360 were kept in the model because of their good spatial match with known contaminated
361 areas, which is supported by the presence of silty sands at the surface of regions
362 exhibiting contaminated aquifers in West Bengal⁵⁰, and by reported elevated
363 groundwater As concentrations in aquifers capped with fine surface material (silt and
364 clay) in Bangladesh^{10,27}.

365

366 According to the calculated odds, the probability (P) of having an As concentration
367 above 10 µgL⁻¹ was calculated as follows:

$$368 \quad P = \frac{\exp(C + \sum_{i=1}^n \lambda_i X_i)}{1 + \exp(C + \sum_{i=1}^n \lambda_i X_i)} \quad (2)$$

369 In addition, we tested how successful the model predicted the number of
370 contaminated cases for probability intervals of 0.1 (Figure 2 and Supplementary
371 Information).

372

373 Misclassification occurs when either a point with As concentration >10 µgL⁻¹ falls in an
374 uncontaminated area (false negative) or an As concentration <10 µgL⁻¹ falls in a
375 contaminated area (false positive). Based on the model classification results (Figure 2
376 and Table S4), a probability threshold of 0.4 resulted in a minimum misclassification
377 rate. The binary risk maps was hence categorized into low-risk areas (probability <0.4)
378 and high risk areas (probability >0.4).

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381 REFERENCES

- 382 1. Nickson, R. *et al.* Arsenic poisoning of Bangladesh groundwater. *Nature* **395**, 338
383 (1998).
- 384 2. Smith, A. H., Lingas, E. O. & Rahman, M. Contamination of drinking water by
385 arsenic in Bangladesh: a public health emergency. *Bull. World Health Org.* **78**,
386 1093–1102 (2000).
- 387 3. Chowdhury, U. K. *et al.* Groundwater arsenic contamination in Bangladesh and
388 West Bengal, India. *Environ. Health Perspect.* **108**, 393-397 (2000).
- 389 4. McArthur, J. M., Ravenscroft, P., Safiulla, S. & Thirlwall, M. F. Arsenic in
390 groundwater: Testing pollution mechanisms for sedimentary aquifers in
391 Bangladesh. *Water Resour. Res.* **37**, 109-117 (2001).

- 392 5. BGS and DPHE. *Arsenic contamination of groundwater in Bangladesh*. Eds.
393 Kinniburgh, D. G. & Smedley, P. L. (British Geological Survey, Keyworth, U.K.,
394 2001). www.bgs.ac.uk/arsenic/bangladesh.
- 395 6. van Geen, A. *et al.* Spatial variability of arsenic in 6000 tube wells in a 25 km²
396 area of Bangladesh. *Water Resour. Res.* **39**(2003).
- 397 7. Ahmed, K. M. *et al.* Arsenic enrichment in groundwater of the alluvial aquifers in
398 Bangladesh: an overview. *Appl. Geochem.* **19**, 181-200 (2004).
- 399 8. Ravenscroft, P., Burgess, W. G., Ahmed, K. M., Burren, M. & Perrin, J. Arsenic in
400 groundwater of the Bengal Basin, Bangladesh: Distribution, field relations, and
401 hydrogeological setting. *Hydrogeol. J.* **13**, 727-751 (2005).
- 402 9. Meharg, A. A. *et al.* Codeposition of organic carbon and arsenic in Bengal Delta
403 aquifers. *Environ. Sci. Technol.* **40**, 4928-4935 (2006).
- 404 10. van Geen, A. *et al.* Flushing history as a hydrogeological control on the regional
405 distribution of arsenic in shallow groundwater of the Bengal Basin. *Environ. Sci.*
406 *Technol.* **42**, 2283-2288 (2008).
- 407 11. McArthur, J. M. *et al.* Natural organic matter in sedimentary basins and its
408 relation to arsenic in anoxic ground water: the example of West Bengal and its
409 worldwide implications. *Appl. Geochem.* **19**, 1255-1293 (2004).
- 410 12. Ahamed, S. *et al.* Arsenic groundwater contamination and its health effects in the
411 state of Uttar Pradesh (UP) in upper and middle Ganga plain, India: A severe
412 danger. *Sci. Total Environ.* **370**, 310-322 (2006).
- 413 13. Smedley, P. L., Zhang, M., Zhang, G. & Luo, Z. Mobilisation of arsenic and other
414 trace elements in fluvio-lacustrine aquifers of the Huhhot Basin, Inner Mongolia.
415 *Appl. Geochem.* **18**, 1453-1477 (2003).
- 416 14. Yu, G. Q., Sun, D. J. & Zheng, Y. Health effects of exposure to natural arsenic in
417 groundwater and coal in China: An overview of occurrence. *Environ. Health*
418 *Perspect.* **115**, 636-642 (2007).
- 419 15. Shrestha, R. R. *et al.* Groundwater arsenic contamination, its health impact and
420 mitigation program in Nepal. *Environ. Sci. Health, Part A* **38**, 185-200 (2003).
- 421 16. Polya, D. A. *et al.* Arsenic hazard in shallow Cambodian groundwaters. *Mineral.*
422 *Mag.* **69**, 807-823 (2005).
- 423 17. Berg, M. *et al.* Magnitude of arsenic pollution in the Mekong and Red River
424 Deltas - Cambodia and Vietnam. *Sci. Total Environ.* **372**, 413-425 (2007).

- 425 18. Buschmann, J., Berg, M., Stengel, C. & Sampson, M. L. Arsenic and Manganese
426 Contamination of Drinking Water Resources in Cambodia: Coincidence of Risk
427 Areas with Low Relief Topography. *Environ. Sci. Technol.* **41**, 2146–2152 (2007).
- 428 19. Berg, M. *et al.* Arsenic contamination of groundwater and drinking water in
429 Vietnam: A human health threat. *Environ. Sci. Technol.* **35**, 2621-2626 (2001).
- 430 20. Buschmann, J. *et al.* Contamination of drinking water resources in the Mekong
431 delta floodplains: Arsenic and other trace metals pose serious health risks to
432 population. *Environ. Int.* **34**, doi:10.1016/j.envint.2007.12.025 (2008).
- 433 21. Ford, R. G., Fendorf, S. & Wilkin, R. T. Introduction: Controls on arsenic transport
434 in near-surface aquatic systems. *Chem. Geol.* **228**, 1-5 (2006).
- 435 22. Harvey, C. F. *et al.* Arsenic mobility and groundwater extraction in Bangladesh.
436 *Science* **298**, 1602-1606 (2002).
- 437 23. Islam, F. S. *et al.* Role of metal-reducing bacteria in arsenic release from Bengal
438 delta sediments. *Nature* **430**, 68-71 (2004).
- 439 24. Rowland, H. A. L. *et al.* The control of organic matter on microbially mediated
440 iron reduction and arsenic release in shallow alluvial aquifers, Cambodia.
441 *Geobiology* **5**, 281-292 (2007).
- 442 25. Berg, M. *et al.* Hydrological and sedimentary controls leading to arsenic
443 contamination of groundwater in the Hanoi area, Vietnam: The impact of iron-
444 arsenic ratios, peat, river bank deposits, and excessive groundwater abstraction.
445 *Chem. Geol.* **249**, 91-112 (2008).
- 446 26. Smedley, P. L. & Kinniburgh, D. G. A review of the source, behaviour and
447 distribution of arsenic in natural waters. *Appl. Geochem.* **17**, 517-568 (2002).
- 448 27. Stute, M. *et al.* Hydrological control of As concentrations in Bangladesh
449 groundwater. *Water Resour. Res.* **43**(2007).
- 450 28. Twarakavi, N. K. C. & Kaluarachchi, J. J. Arsenic in the shallow ground waters of
451 conterminous United States: Assessment, health risks, and costs for MCL
452 compliance. *J. Am. Water Resour. Assoc.* **42**, 275-294 (2006).
- 453 29. Amini, M. *et al.* Statistical modeling of global geogenic arsenic contamination in
454 groundwater. *Environ. Sci. Technol.* **42**, 3669-3675 (2008).
- 455 30. Goovaerts, P. *et al.* Geostatistical modeling of the spatial variability of arsenic in
456 groundwater of southeast Michigan. *Water Resour. Res.* **41**(2005).

- 457 31. Lee, J. J., Jang, C. S., Wang, S. W. & Liu, C. W. Evaluation of potential health
458 risk of arsenic-affected groundwater using indicator kriging and dose response
459 model. *Sci. Total Environ.* **384**, 151-162 (2007).
- 460 32. Hossain, F., Hill, J. & Bagtzoglou, A. C. Geostatistically based management of
461 arsenic contaminated ground water in shallow wells of Bangladesh. *Water*
462 *Resour. Manag.* **21**, 1245-1261 (2007).
- 463 33. Twarakavi, N. K. C. & Kaluarachchi, J. J. Aquifer vulnerability assessment to
464 heavy metals using ordinal logistic regression. *Ground Water* **43**, 200-214 (2005).
- 465 34. Ayotte, J. D. *et al.* Modeling the probability of arsenic in groundwater in New
466 England as a tool for exposure assessment. *Environ. Sci. Technol.* **40**, 3578-
467 3585 (2006).
- 468 35. Reading, H. G. (ed.) *Sedimentary Environments: Processes, Facies and*
469 *Stratigraphy*, third edition (Blackwell Science, Oxford, 1996).
- 470 36. Tan, K. H. *Environmental Soil Science*, second edition (M. Dekker, New York,
471 2000).
- 472 37. Hori, K. *et al.* Delta initiation and Holocene sea-level change: example from the
473 Song Hong (Red River) delta, Vietnam. *Sediment. Geol.* **164**, 237-249 (2004).
- 474 38. Kirk, M. F. *et al.* Bacterial sulfate reduction limits natural arsenic contamination in
475 groundwater. *Geology* **32**, 953-956 (2004).
- 476 39. Lowers, H. A. *et al.* Arsenic incorporation into authigenic pyrite, bengal basin
477 sediment, Bangladesh. *Geochim. Cosmochim. Acta* **71**, 2699-2717 (2007).
- 478 40. Stanley, D. J. & Warne, A. G. Worldwide Initiation of Holocene Marine Deltas by
479 Deceleration of Sea-Level Rise. *Science* **265**, 228-231 (1994).
- 480 41. Wosten, J. H. M. *et al.* Interrelationships between hydrology and ecology in fire
481 degraded tropical peat swamp forests. *Int. J. Water Resour. Dev.* **22**, 157-174
482 (2006).
- 483 42. Giesen, W. *Causes of Peatswamp Forest Degradation in Berbak National Park*
484 *and Recommendations for Restoration, Water for Food & Ecosystems*
485 *Programme.* (Arcadis Euroconsult, Arnhem, Holland, 2004).
- 486 43. Tun, K. M. A. *Report on the Assessment of Arsenic Content in Groundwater and*
487 *the Prevalence of Arsenicosis in Thabaung and Kyonpyaw Townships,*
488 *Ayeyarwaddy Division.* (Department of Medical Research, Yangon, Myanmar,
489 2002).

- 490 44. Kohnhorst, A. Arsenic in groundwater in selected countries in south and
491 southeast Asia: A review. *J. Trop. Med. Parasitol.* **28**, 73-82 (2005).
- 492 45. Tanabe, S. *et al.* Stratigraphy and Holocene evolution of the mud-dominated
493 Chao Phraya delta, Thailand. *Quat. Sci. Rev.* **22**, 789-807 (2003).
- 494 46. Metivier, F., Gaudemer, Y., Tapponnier, P. & Klein, M. Mass accumulation rates
495 in Asia during the Cenozoic. *Geophys. J. Int.* **137**, 280-318 (1999).
- 496 47. Robinson, R. A. J. *et al.* The Irrawaddy River sediment flux to the Indian Ocean:
497 The original nineteenth-century data revisited. *J. Geol.* **115**, 629-640 (2007).
- 498 48. Berg, M. *et al.* Arsenic removal from groundwater by household sand filters:
499 Comparative field study, model calculations, and health benefits. *Environ. Sci.*
500 *Technol.* **40**, 5567-5573 (2006).
- 501 49. Kleinbaum, D. G. & Klein, M. *Logistic Regression: A Self-Learning Text*, second
502 edition (Springer-Verlag, New York, 2002).
- 503 50. Pal, T., Mukherjee, P. K., Sengupta, S., Bhattacharyya, A. K. & Shome, S.
504 Arsenic pollution in groundwater of West Bengal, India - An insight into the
505 problem by subsurface sediment analysis. *Gondwana Res.* **5**, 501-512 (2002).
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510

511 **ACKNOWLEDGEMENTS**

512 We thank Thomas Rosenberg, Rudi Febriamansyah, M.A. Hayatuddin, and Erwin

513 Nofyan for support during groundwater sampling in Sumatra; Caroline Stengel,

514 Thomas Rüttimann, Madeleine Langmeier and Richard Illi for elemental analyses;

515 Karim Abbaspour and Bas den Brok for helpful discussions; Rich Wildman and Helen

516 Rowland for proofreading, and the anonymous reviewers for constructive comments.

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518

519 **Supplementary Information** accompanies this paper

520

521 **Competing financial interests statement**

522 The authors declare that they have no competing financial interests.

523 **Figure captions**

524 **Figure 1. Uniformly classified geological map of Southeast Asia.** It indicates
525 seven different sedimentary depositional environments in the mapped countries of
526 Bangladesh, Myanmar, Thailand, Cambodia, Vietnam, and Sumatra (Indonesia).

527

528 **Figure 2. Model classification results.** The graph shows the sensitivity (true
529 positives) and specificity (true negatives) of the model for different probability cutoff
530 values. The full classification table is provided in the Supplementary Information
531 (Table S4). A probability threshold of 0.4 was applied to delineate low- and high-risk
532 areas in the binary risk maps shown in Figures 3b, 3d, and 4b (see Methods).

533

534 **Figure 3. Modelled probability of As concentrations exceeding $10 \mu\text{g L}^{-1}$ under**
535 **reducing aquifer conditions. a,** Continuous probability map of Bangladesh. **b,** Binary
536 map of Bangladesh (probability threshold 0.4) indicating high- and low-risk areas
537 overlain by aggregated As concentrations. Areas where groundwater is mainly drawn
538 from Pleistocene aquifers are sketched in brown. **c,** Continuous probability map of the
539 Mekong delta (Cambodia and Vietnam). **d,** Binary risk map of the Mekong delta
540 overlain by aggregated As concentrations. **e,** Continuous probability map of the Red
541 River delta (Vietnam). **f,** Continuous probability map of the Irrawaddy delta (Myanmar)
542 and Chao Phraya basin (Thailand).

543

544 **Figure 4. Maps of the model verification study area in Southeast Sumatra. a,**
545 Map of whole Sumatra depicting the probability of groundwater As concentrations
546 exceeding $10 \mu\text{g L}^{-1}$. The corresponding colour code is given in Figure 3. **b,** Binary risk
547 map (probability cutoff 0.4) and As concentrations measured for this study in the
548 vicinity of Palembang (South Sumatra). Swampy areas (high-risk area) are scarcely

549 populated and the number of sampled tubewells in this area is therefore limited. **c**,

550 Geological map (source: see Supplementary Table S1).

551

552 **Table 1. Results of logistic regression analysis.** Weighting coefficients of the
553 independent variables (λ) were used to calculate probabilities of As contamination.
554 Wald values (%) indicate the relative importance of the variables, and p-values the
555 absolute significance, where a value <0.05 indicates a significance of at least 95%.
556 Variables that were not statistically significant (based on a 95% level) were excluded
557 from the model (Table S2), with exception of medium textured soils and silt in subsoil
558 (see Methods). Excluded variables are: tidal deposits, other Holocene deposits, pre-
559 Holocene deposits (see Figure 1), coarse textured soils, soil sand and clay contents,
560 and climate.
561

| Variables | | λ | Wald | p-value |
|---------------------------------------|-----------------------|-----------|-------|---------|
| Sedimentary depositional environments | Deltaic deposits | 1.65 | 71.55 | <0.001 |
| | Organic-rich deposits | 1.11 | 72.20 | <0.001 |
| | Alluvial deposits | 0.55 | 12.51 | <0.001 |
| | Floodplain deposits | - 0.95 | 2.32 | 0.010 |
| Soil variables | Medium textured soils | 0.60 | 7.80 | 0.128 |
| | Fine textured soils | 0.24 | 1.00 | 0.005 |
| | Silt in subsoil | 0.10 | 6.62 | 0.317 |
| | Silt in topsoil | - 0.09 | 6.28 | 0.012 |
| - | Regression constant | - 1.54 | 20.67 | <0.001 |

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