Supplementary Materials

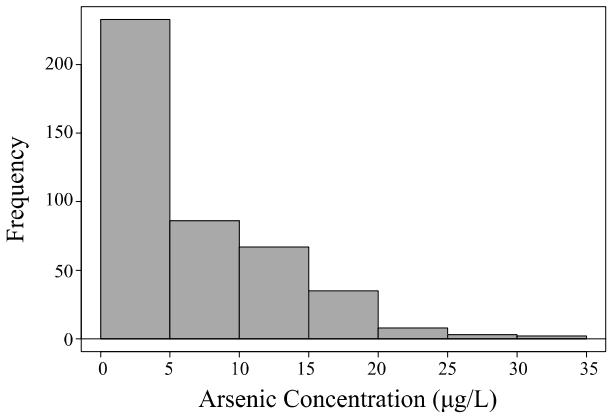

Figure S1. Frequency distribution of groundwater arsenic concentrations in the entire OSE dataset for Uruguay.

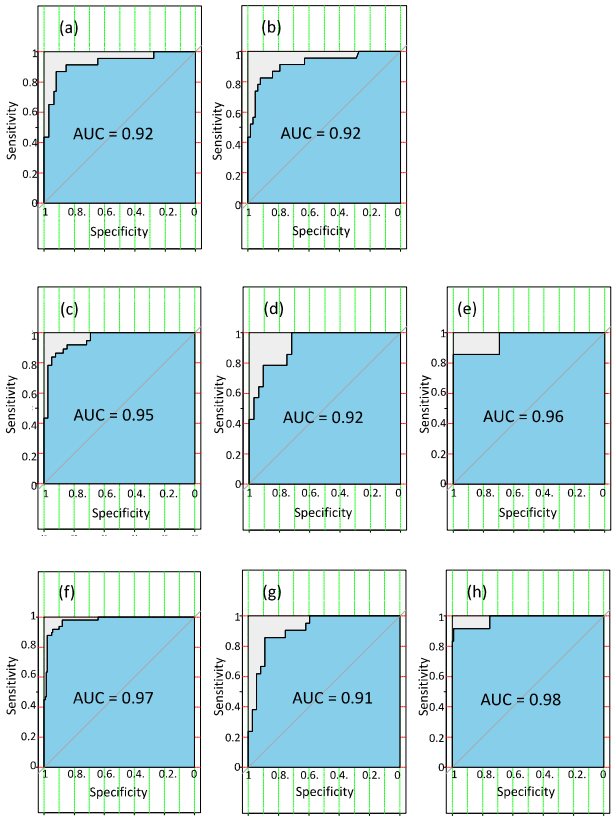

Figure S2. The cross-validation results. ROC curves and AUC values of (a) pure overall (1A-ML-Pure), (b) pure overall with "sedimentary or crystalline" as a predictor (1B-ML-Pure with Geol), (d) hybrid shallow (2A-HML-Shal), (e) hybrid deep (2B-HML-Deep), (g) hybrid sedimentary (3A-HML-Sed), (h) hybrid crystalline (3B-HML-Cry) machine learning models calculated on their testing datasets. The AUC values of (c) hybrid depth (2C-HML-Depth), and (f) hybrid geology (3C-HML-Geol) machine learning models calculated on their combined (i.e. deep and shallow for depth; sedimentary and crystalline for geology) testing datasets.

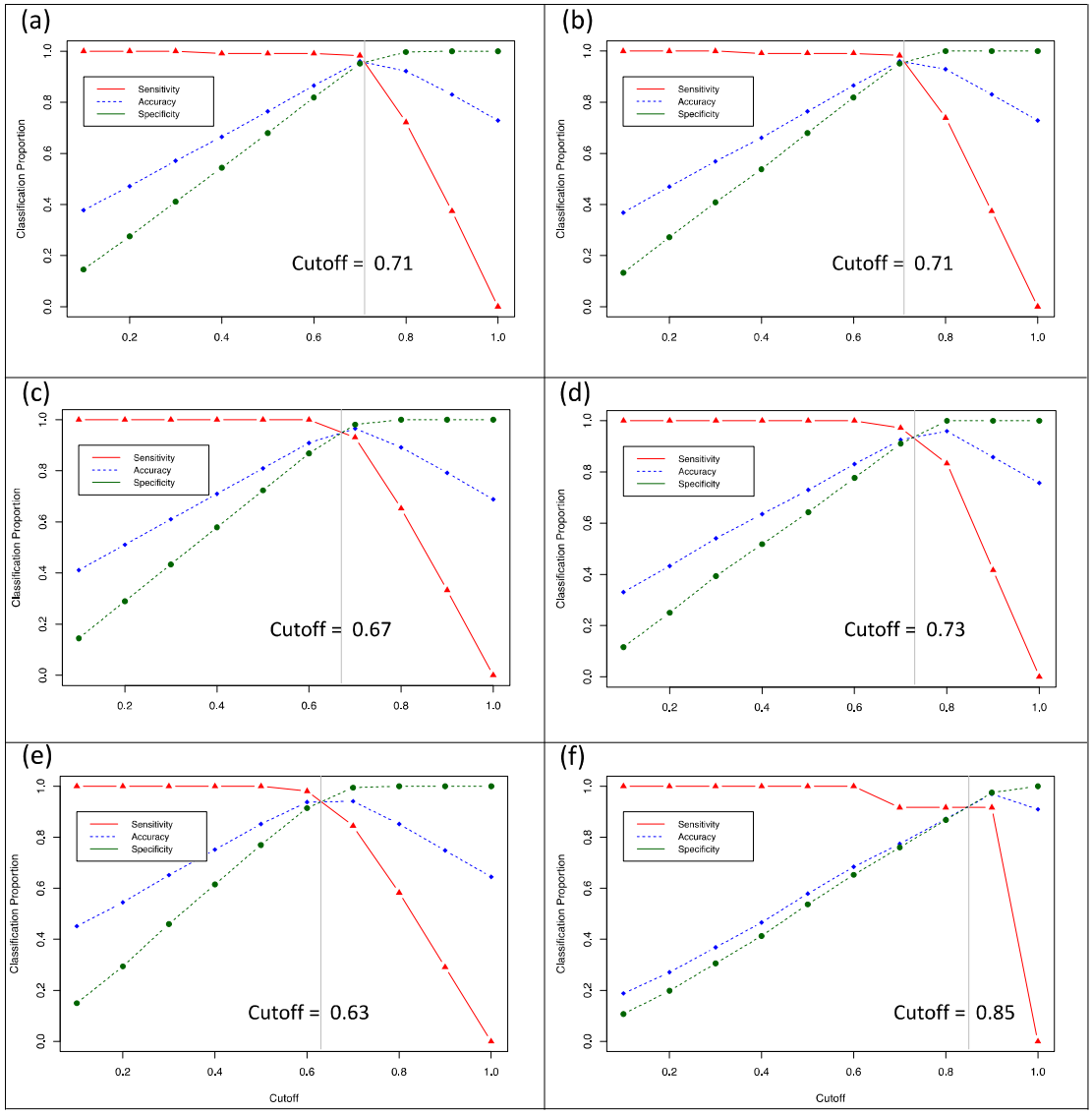

Figure S3. Performance of (a) pure overall (1A-ML-Pure), (b) pure overall with "sedimentary or crystalline" as a predictor (1B-ML-Pure with Geol), (c) hybrid shallow (2A-HML-Shal), (d) hybrid deep (2B-HML-Deep), (e) hybrid sedimentary (3A-HML-Sed), (f) hybrid crystalline (3B-HML-Cry) models using entire modelling dataset.

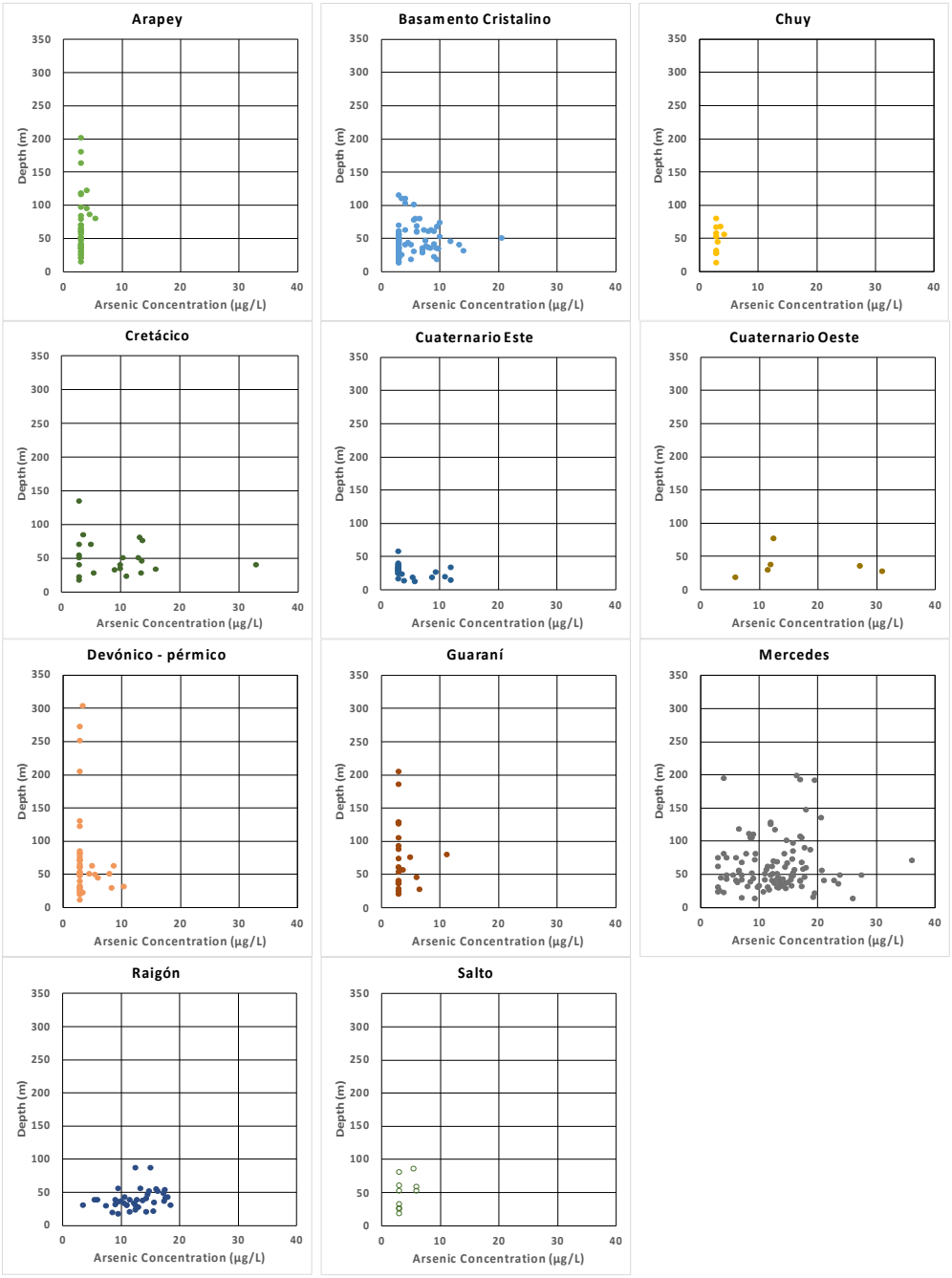

Figure S4. Depth vs. arsenic concentration for the main aquifers in Uruguay. Data from OSE.

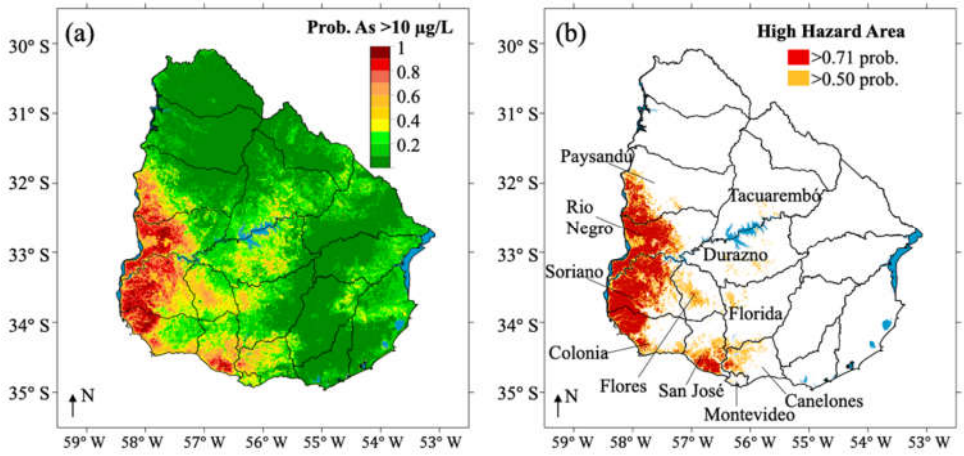

Figure S5. Pure overall model with "sedimentary or crystalline" as a predictor (1B-ML-Pure with Geol) of Uruguay groundwater arsenic concentrations: (a) map of probability of groundwater arsenic concentration exceeding 10 µg/L; (b) map of high groundwater arsenic hazard areas (defined here by a probability exceeding cut-off values of 0.50 or 0.71).

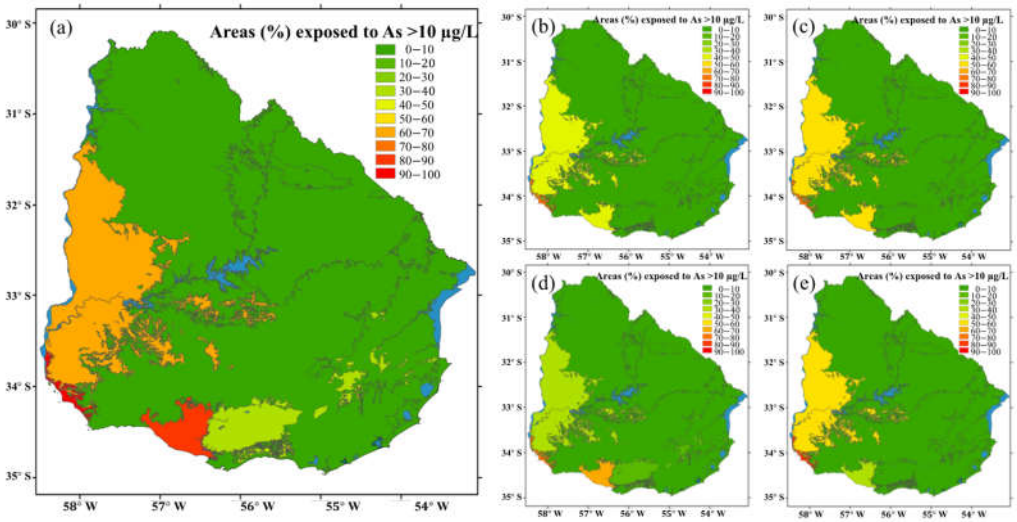

Figure S6. Proportion of areas in each aquifer of Uruguay with high groundwater arsenic hazard: comparison of (a) simple expert selection aquifer model (4-ES-Aqui; observed value) and (b) pure overall (1A-ML-Pure), (c) hybrid geology (sedimentary and crystalline; 3C-HML-Geol), (d) hybrid shallow (2A-HML-Shal), and (e) hybrid deep (2B-HML-Deep) models. Simple expert selection model (observed value) based on the percentage of high arsenic (> 10 µg/L) in the aquifers, and geometric mean of arsenic concentrations was taken within a pixel (1km^2). Modelled high groundwater hazard areas defined as those with a probability of arsenic concentration being than 10 µg/L exceeding a cutoff value of 0.5 and a specific cutoff value where sensitivity is equal to specificity (overall: 0.71; shallow: 0.67; deep: 0.73; sedimentary: 0.63; crystalline: 0.85). The mean of modelled high groundwater hazard areas defined by two cutoffs were used to produce the figure (b), (c), (d), and (e) and been shown in Table S3.

Table S1. Potential predictors used in the machine learning models. Descriptions and data sources are listed. Predictors are grouped into 4 categories: climate, soil, topography, and lithology.

Variable Type	Variables	Descriptions	Data Sources
Climate	Actual evapotranspiration	Average rate of actual evapotranspiration (mm/yr)	[40]
Climate	Aridity	PET/Precipitation	[41]
Climate	Potential evapotranspiration	Average rate of potential evapotranspiration (mm/yr)	[41]
Climate	Precipitation	Average rate of precipitation (mm/yr)	[40]
Climate	Priestley-Taylor alpha coefficient	AET/PET	[40]
Climate	Temperature	Average r temperature (℃)	[41]
Soil	Calcisols	Probability of the occurrence of calcisols	[42]
Soil	Clay	Weight % of clay particles (<0.0002 mm) at 2 m depth	[42]
Soil	Coarse fragments	Volume % of coarse fragments (>2 mm) at 2 m depth	[42]
Soil	Fluvisols	Probability of the occurrence of fluvisols	[42]
Soil	Gleysols	Probability of the occurrence of gleysols	[42]
Soil	Sand	Weight % of sand particles (0.05 - 2 mm) at 2 m depth	[42]
Soil	Silt	Weight % of silt particles (0.0002 - 0.05 mm) at 2 m depth	[42]
Soil	Soil cation exchange capacity	Cation exchange capacity (cmolc/kg) at 2 m depth	[42]
Soil	Soil pH	Soil pH measured in water at 2 m depth	[42]
Soil	Soil organic carbon content	Soil organic carbon content (g/kg) at 2 m depth	[42]
Soil	Soil organic carbon density	Soil organic carbon density (kg/m^3) at 2 m depth	[42]
Soil	Soil organic carbon stock	Soil organic carbon stock (t/ha) at 2 m depth	[42]
Soil	Soil and sedimentary deposit thickness	The thickness of soil and sedimentary deposit (m)	[43]
Soil	Solonchaks	Probability of the occurrence of solonchaks	[42]
Soil	Water wilting point	Volume % of available soil water until wilting point at 2 m depth	[42]
Topography	Elevation	Elevation (m)	[44]
Topography	Slope	Slope (arc)	[45]
Topography	Landform	7 different categories of landform	[45]
Topography	Topographic wetness index	Combination of upslope contributing area and slope	[45]
Lithology	Lithology	15 different categories of lithology	[45]

Table S2. Comparison of number of available predictors at each branch in random forest models from 1 to 26 (the total number of potential predictors). The number shown in grey shadow is the optimum number of predictors at each branch in the model.

Number of available predictors	Out-of-bag (OOB) error				
	Pure Overall Model	Hybrid Shallow Model	Hybrid Deep Model	Hybrid Sedimentary Model	Hybrid Crystalline Model
1	0.2331	0.2536	0.2356	0.2152	0.3644
2	0.2060	0.2316	0.1820	0.1993	0.3652
3	0.2135	0.2236	0.1793	0.2287	0.3641
4	0.2133	0.2247	0.1643	0.2176	0.3672
5	0.2181	0.2404	0.1650	0.2243	0.3642
6	0.2220	0.2340	0.1600	0.2301	0.3675
7	0.2101	0.2374	0.1586	0.2297	0.3663
8	0.2027	0.2325	0.1525	0.2285	0.3668
9	0.2123	0.2429	0.1627	0.2359	0.3671
10	0.2144	0.2381	0.1668	0.2240	0.3661
11	0.2103	0.2321	0.1631	0.2316	0.3702
12	0.2261	0.2453	0.1616	0.2277	0.3651
13	0.2232	0.2368	0.1634	0.2150	0.3655
14	0.2264	0.2481	0.1562	0.2282	0.3654
15	0.2190	0.2353	0.1472	0.2272	0.3658
16	0.2160	0.2419	0.1656	0.2262	0.3655
17	0.2250	0.2404	0.1619	0.2188	0.3693
18	0.2172	0.2464	0.1623	0.2266	0.3703
19	0.1995	0.2430	0.1623	0.2246	0.3693
20	0.2161	0.2461	0.1630	0.2277	0.3664
21	0.2214	0.2505	0.1643	0.2244	0.3723
22	0.2155	0.2463	0.1612	0.2260	0.3719
23	0.2167	0.2435	0.1618	0.2317	0.3717
24	0.2179	0.2464	0.1669	0.2244	0.3704
25	0.2230	0.2396	0.1560	0.2279	0.3723
26	0.2204	0.2396	0.1651	0.2298	0.3755

Table S3. Proportion of areas of each aquifer of Uruguay with high groundwater arsenic hazard: comparison of (a) simple expert selection model (4-ES-Aqui; observed values) and ((b) pure overall (1A-ML-Pure), (c) hybrid geology (sedimentary and crystalline; 3C-HML-Geol), (d) hybrid shallow (2A-HML-Shal), and (e) hybrid deep (2B-HML-Deep) models. Simple expert selection aquifer model (observed value) based on the percentage of high arsenic (> 10 μg/L) in the aquifers, and geometric mean of arsenic concentrations was taken within individual 1 km^2 pixels. Modelled high groundwater hazard areas defined as those with a probability of arsenic concentration being than 10 μg/L exceeding a cutoff value of 0.5 and a specific cutoff value where sensitivity is equal to specificity (overall: 0.71; shallow: 0.67; deep: 0.73; sedimentary: 0.63; crystalline: 0.85). Mean of modelled high groundwater hazard areas defined by the two cutoffs are shown.

Aquifer	Simple expert selection model (observed value)	Mean of areas exposed to arsenic concentrations greater than 10 μg/L			
		Pure Overall Model (%)	Hybrid Shallow Model (%)	Hybrid Deep Model (%)	Hybrid Sedimentary and Crystalline Models (%)
Arapey	0.00 (n=0)	0.08	0.49	0.33	0.02
Basamento Cristalino	5.93 (n=7)	3.33	6.99	3.51	1.69
Chuy	0.00 (n=0)	0.00	0.00	0.98	0.00
Cretácico	33.33 (n=6)	8.60	16.66	7.74	9.59
Cuaternario Este	4.00 (n=1)	0.04	0.28	0.87	0.00
Cuaternario Oeste	100.00 (n=4)	70.80	79.79	88.34	77.01
Devónico - pérmico	2.22 (n=1)	0.26	0.74	0.19	0.20
Guaraní	0.00 (n=0)	0.02	0.00	0.01	0.07
Mercedes	65.69 (n=67)	47.67	36.77	51.82	50.89
Raigón	85.29 (n=29)	40.82	61.96	37.28	54.94
Salto	0.00 (n=0)	0.00	0.00	0.00	0.00