Electronic Supplementary Information

Enhanced functional DNA biosensor for distance-based read-by-eye quantification of various analytes based on starch-hydrolysis-adjusted wettability change in paper devices

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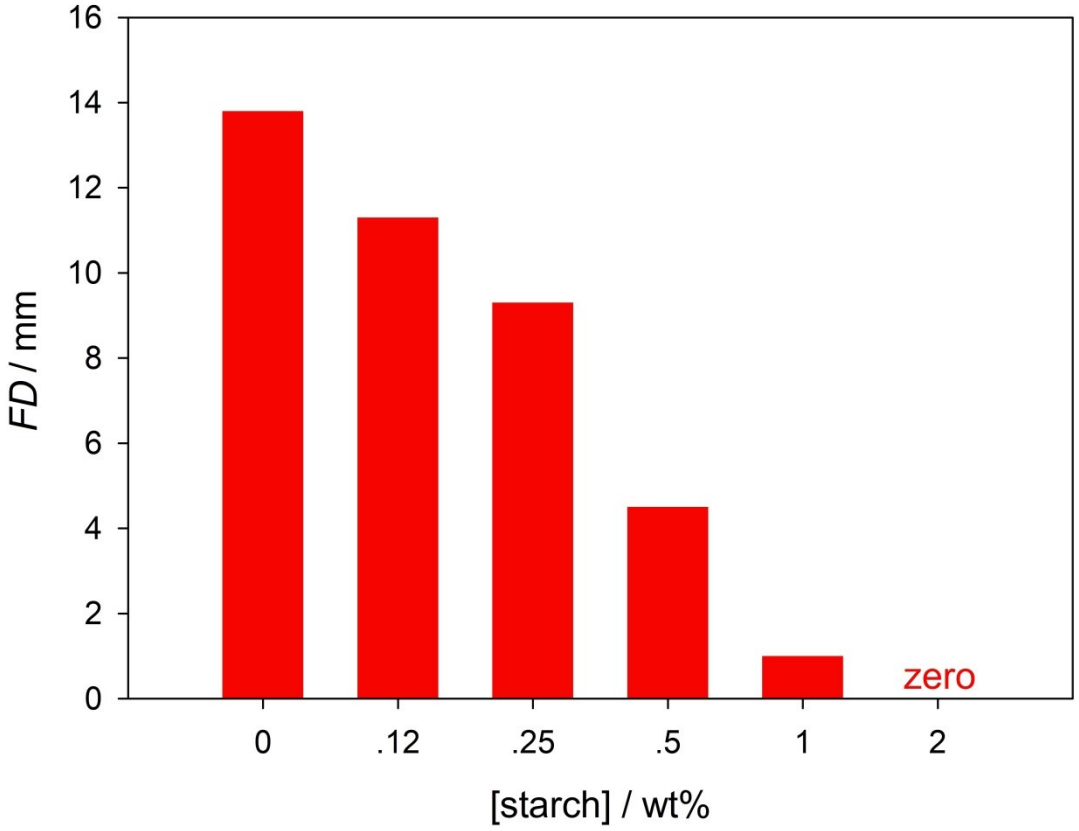

Fig. S1. Investigation of the effect of starch concentration on flowing distance (*FD*) of red ink in paper devices. The results showed that the ink's *FD* values were inversely yet linearly associated with the starch levels in a proper range, but too high a starch level (e.g., 2 wt%) could totally block the paper's porous microstructures resulting in no ink flowing in the paper body.

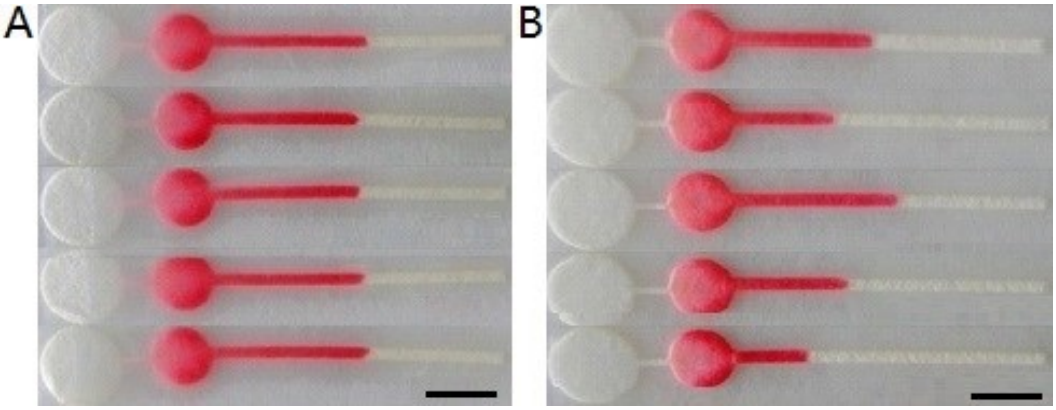

Fig. S2. Comparative study of the flowing of red ink in the paper devices with (A) or without (B) the encapsulation of transparent adhesive tape. The results showed that more stable (repeatable) ink flowing were obtained in the encapsulated paper devices. The scale bar is 3.5 mm.

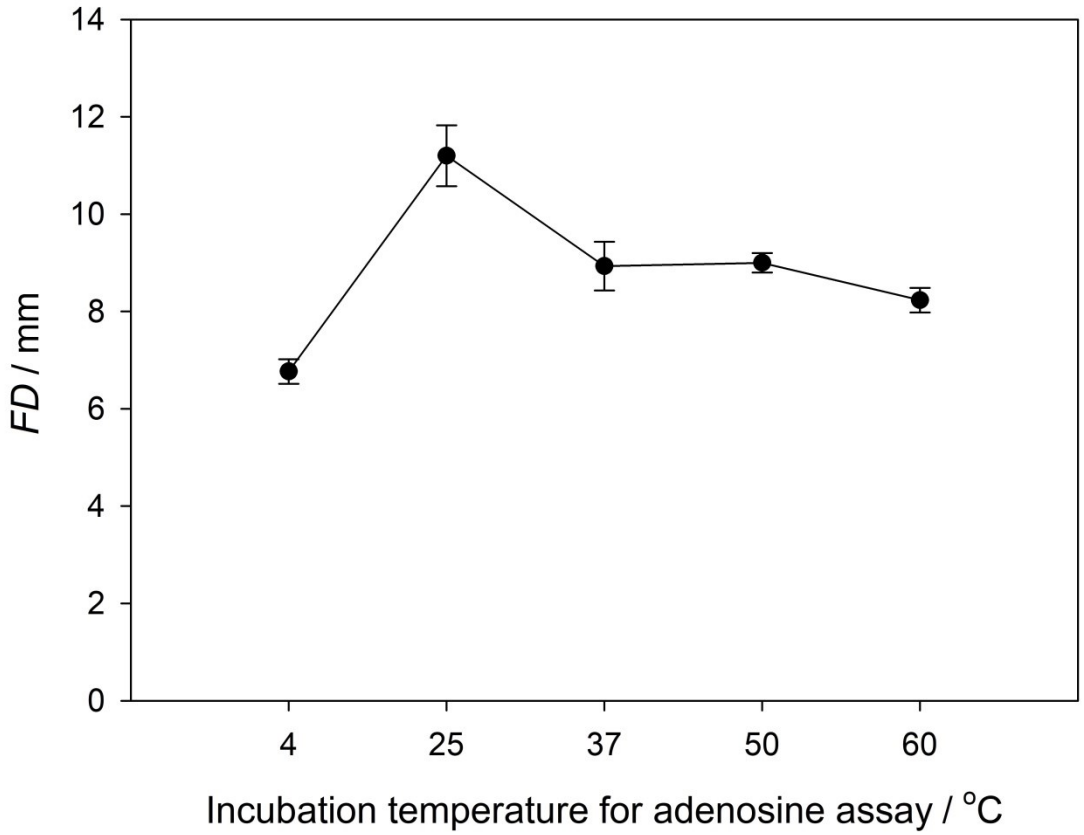

Fig. S3. Optimization of the temperature for incubating aptamer-loaded super-paramagnetic microbeads, adenosine sample (50 μM), and enzyme-loaded SiO_2 microbeads. The *FD* results showed that 25 °C should be chosen as the optimal incubation temperature to produce the highest *FD* signal.

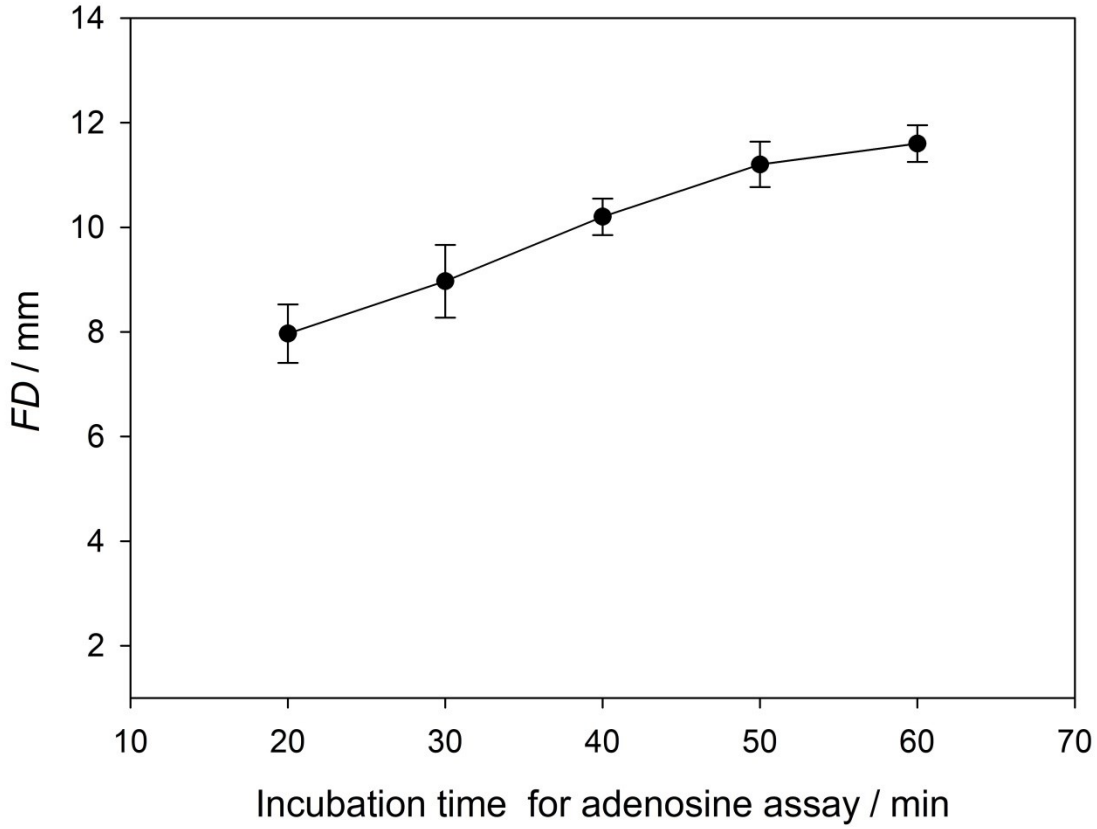

Fig. S4. Optimization of the time for incubating aptamer-loaded super-paramagnetic microbeads, adenosine sample (50 μM), and enzyme-loaded SiO_2 microbeads. The *FD* results showed that 60 min should be chosen as the optimal incubation temperature because no obvious increase in the *FD* signal was observed as such a period of incubation time was applied.

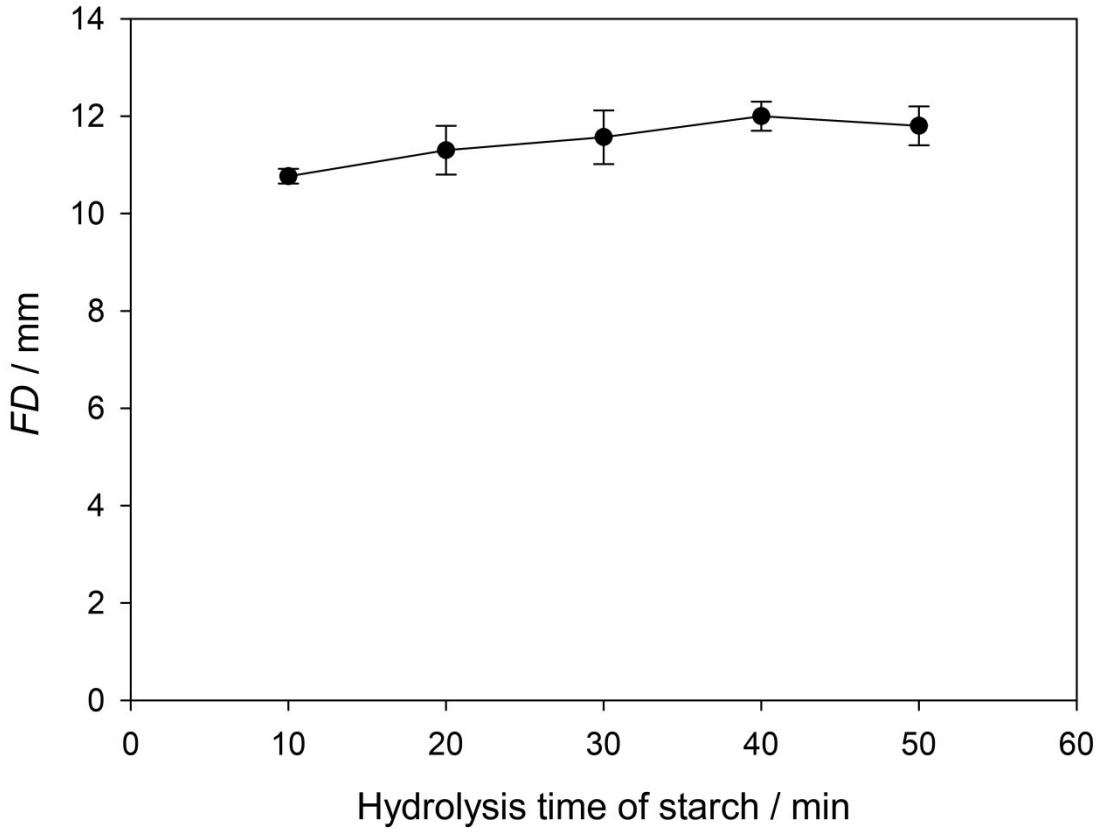

Fig. S5. Optimization of the time for hydrolyzing starch in the assay of 50 μM adenosine sample. The *FD* results showed that 40 min should be chosen as the optimal hydrolysis time because no obvious increase in the *FD* signal was observed as longer periods of hydrolysis time were applied.

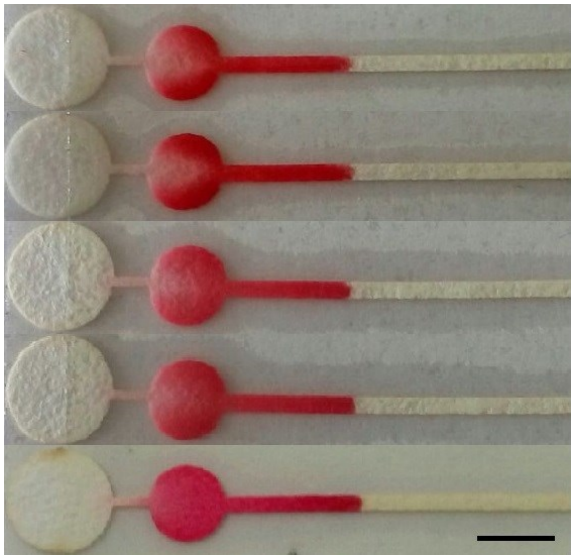

Fig. S6. Assay results obtained from five 1.7 μM adenosine samples (in buffer) under the same experimental conditions using the developed adenosine assay method. The relative standard deviation of the resultant *FD* values was calculated to be ~3.1% (n = 5). The scale bar is 3.5 mm.

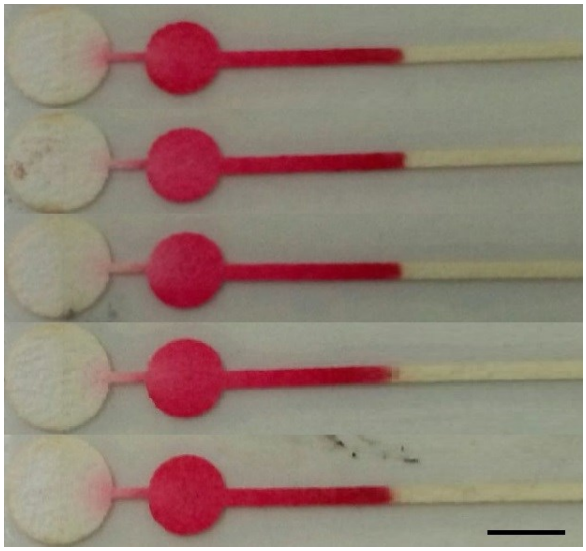

Fig. S7. Assay results obtained from five 15 μM adenosine samples (in buffer) under the same experimental conditions using the developed adenosine assay method. The relative standard deviation of the resultant *FD* values was calculated to be ~4.4% (n = 5). The scale bar is 3.5 mm.

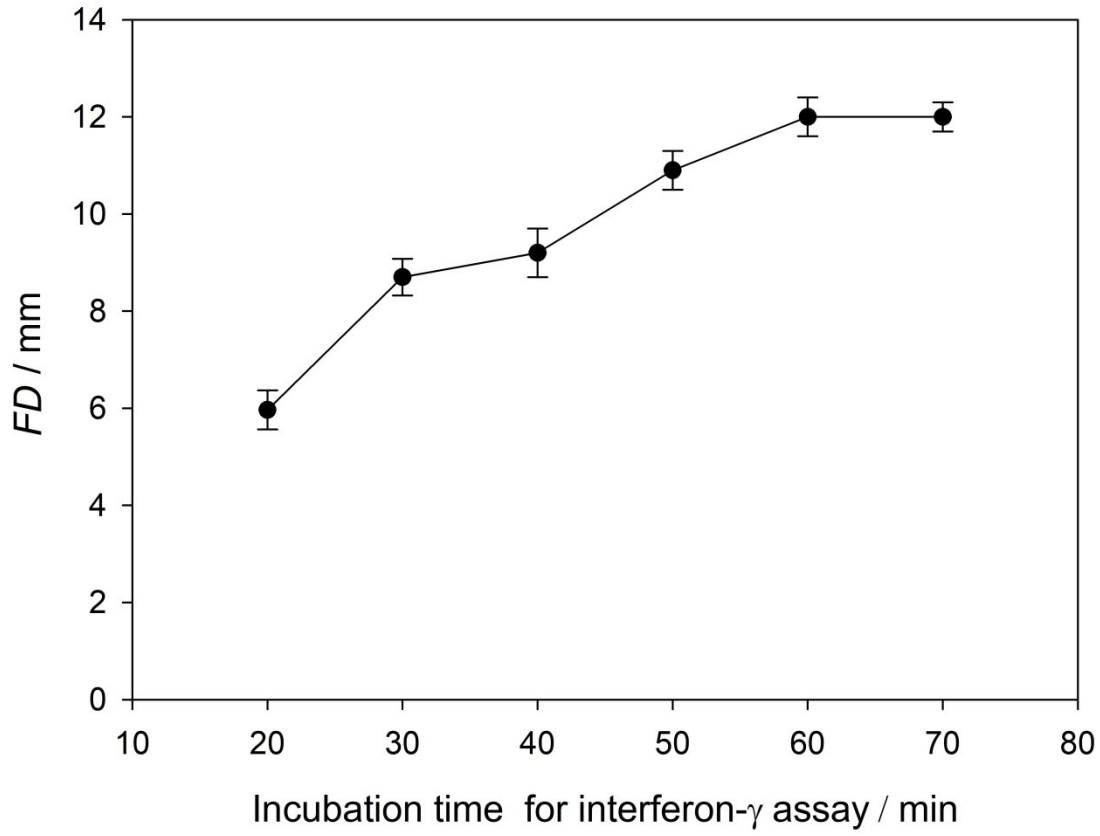

Fig. S8. Optimization of the time for incubating aptamer-loaded super-paramagnetic microbeads, interferon-γ sample (35 nM), and enzyme-loaded SiO_2 microbeads. The *FD* results showed that 60 min should be chosen as the optimal incubation temperature because no obvious increase in the *FD* signal was observed as longer periods of incubation time were applied.

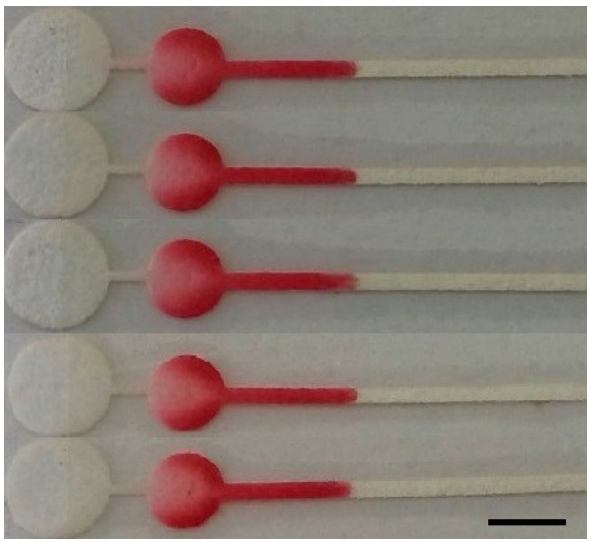

Fig. S9. Assay results obtained from five 0.5 nM interferon-γ samples (in buffer) under the same experimental conditions using the developed adenosine assay method. The relative standard deviation of the resultant *FD* values was calculated to be ~2.7% (n = 5). The scale bar is 3.5 mm.

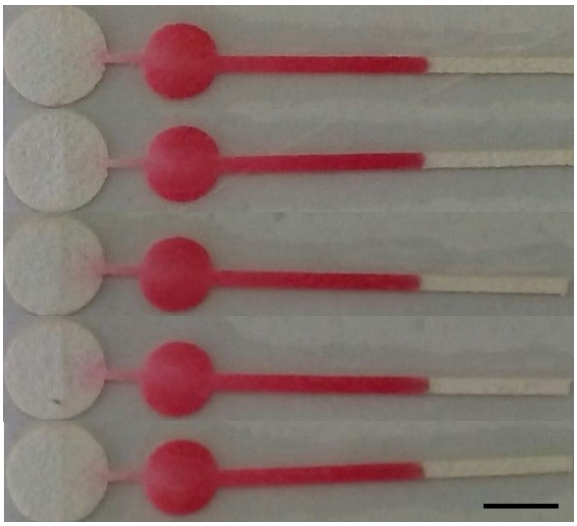

Fig. S10. Assay results obtained from five 32 nM interferon-γ samples (in buffer) under the same experimental conditions using the developed adenosine assay method. The relative standard deviation of the resultant *FD* values was calculated to be ~4.6% (n = 5). The scale bar is 3.5 mm.

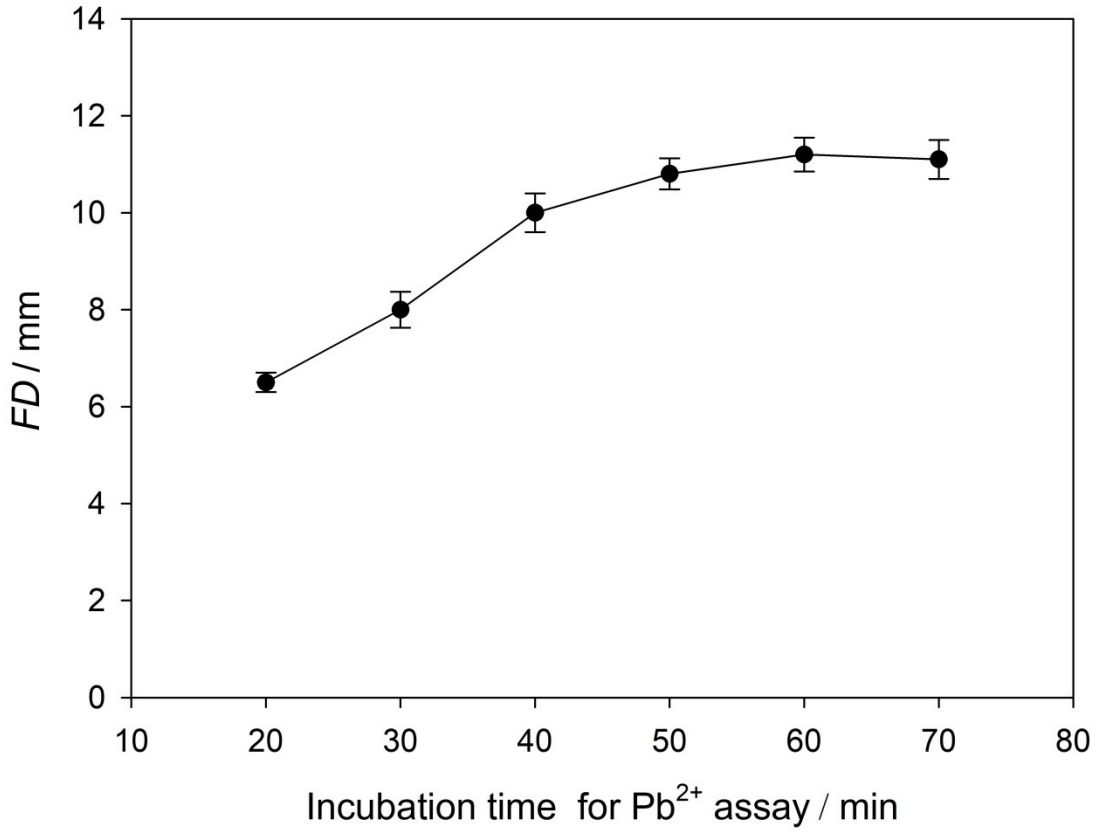

Fig. S11. Optimization of the time for incubating aptamer-loaded super-paramagnetic microbeads, Pb^{2+} sample (50 nM), and enzyme-loaded SiO_2 microbeads. The *FD* results showed that 60 min should be chosen as the optimal incubation temperature because no obvious increase in the *FD* signal was observed as longer periods of incubation time were applied.

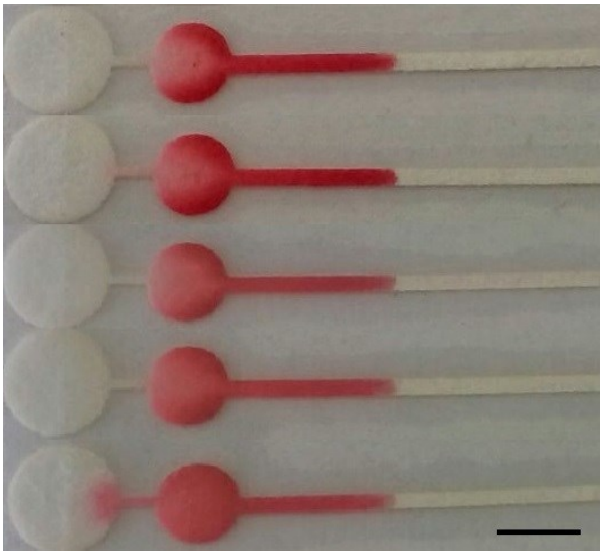

Fig. S12. Assay results obtained from five 3 nM Pb^{2+} samples (in buffer) under the same experimental conditions using the developed adenosine assay method. The relative standard deviation of the resultant *FD* values was calculated to be ~1.9% (*n* = 5).

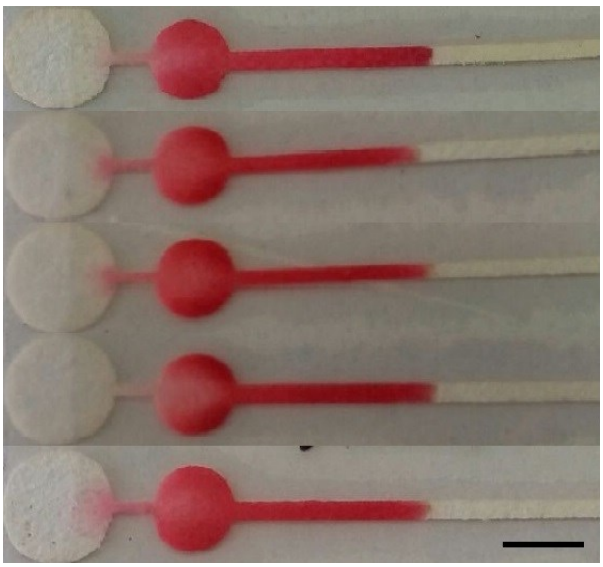

Fig. S13. Assay results obtained from five 12 nM Pb^{2+} samples (in buffer) under the same experimental conditions using the developed adenosine assay method. The relative standard deviation of the resultant *FD* values was calculated to be ~2.7% (n = 5).

Table S1 Performance comparison of the developed adenosine assay method with some recently-reported representative ones

Detection technique	LOD[a]	LCR[b]	Instrument cost[d] (US $)	Ref.
Fluorescent measurement using a Hitachi F-7000 spectrometer	1 μM	1 μM-100 mM	~29,000-43,000	1
Electrochemical measurement using an Autolab PGSTAT302N	10.9 μM	NA[c]	~29,000-43,000	2
Electrochemical measurement using an Autolab PGSTAT302N	3 μM	10-2500 μM	~29,000-43,000	3
Electrochemical measurement for nanopore platform using a CEZ-2400 patchclamp amplifier	0.9 μM	1-100 μM	~24,000-26,000	4
Surface plasmon resonance	1 μM	NA[c]	Typically tens to hundreds of thousands of dollars	5
Surface-enhanced Raman scattering	10 nM	10 nM-10μM	Typically tens to hundreds of thousands of dollars	6
Electromagnetic piezoelectric acoustic platform	0.3 μM	0.5-5 μM	NA[c]	7
Glucose measurement using a personal glucose meter	3.4 μM	0-500 μM	(No analytical instrument was used; a personal glucose meter typically costs tens of dollars)	8
Colorimetric analysis with UV/Vis spectroscopy recorded on a Hitachi U-3010 spectrophotometer	1 μM	0-200 μM	＞1,000	9
Distance-based measurement in paper devices	NA[c]	0-800 μM	(No analytical instrument was used, a ruler typically costs < 1 dollar)	10
	20 μM	0-200 μM		11
	0.16 μM	0.39-25 μM		12
	1.6 μM	1.7-62.5 μM		This work

[a] LOD, limit of detection.
[b] LCR, linear concentration range.
[c] NA, not available in the literature.
[d] The information on the reference price per instrument was obtained from the internet.

Table S2 Recovery of adenosine in human serum samples

Sample	Added (pM)	Found[a] (pM)	Recovery (%)	RSD[b] (%, *n*=6)
1	2.00	1.95	97.74%	7.13
2	5.00	4.67	97.26%	3.05
3	10.00	10.11	101.07%	5.83

[a] The adenosine concentrations in the samples determined using the proposed method.

[b] RSD, relative standard deviations.

Table S3 Performance comparison of the developed interferon-γ assay method with some recently-reported representative ones

Detection technique	LOD[a]	LCR[b]	Instrument cost[c] (US $)	Ref.
Surface plasmon resonance	10 pM	0.01-1 nM	Typically tens to hundreds of thousands of dollars	13
Fluorescent measurement using a plate reader	5 nM	5 nM-100 μM	Typically tens to hundreds of thousands of dollars	14
Electrochemical measurement using an Agilent semiconductor parameter analyzer	83 pM	0-10 μM	Typically thousands to tens of thousands of dollars	15
Electrochemical measurement using a MultiWE32 multichannel potentiostat	3.9 nM	3-1.5 μM	NA[d]	16
Electrochemical measurement using a CHI6273E potentiostat	11.56 pM	22.22-110 pM	~3,600-8,300	17
Electrochemical measurement using an electrochemical workstation	0.3 nM	0.5-300 nM	Typically thousands of dollars	18
Electrochemical measurement using a CHI 842B electrochemical workstation	60 pM	60 pM-10 nM	NA[d]	19
Glucose measurement with a personal glucose meter	3.4 μM	0-500 μM	(No analytical instrument was used; a personal glucose meter typically costs tens of dollars)	8
Distance measurement in paper devices	0.2 nM	0.25-32 nM	(No analytical instrument was used, a ruler typically costs < 1 dollar)	This work

[a] LOD, limit of detection.
[b] LCR, linear concentration range.
[c] The information on the reference price per instrument was obtained from the internet.
[d] NA, not available on the internet.

Table S4 Performance comparison of the developed Pb^{2+} assay method with some recently-reported representative ones

Detection technique	LOD[a]	LCR[b]	Instrument cost[c] (US $)	Ref.
Fluorescent measurement using a Hitachi F-7000 spectrometer	34 nM	0.1-5 μM	~29,000-43,000	20
Fluorescent measurement using a Tecan Spark multimode microplate reader	23.5 nM	100-600 nM	~4,600	21
Fluorescent measurement using a Edinburgh FS5 spectrometer	10 nM	10 nM-1 μM	~70,000-140,000	22
Rayleigh scattering measurement using a Hitachi F-4500 spectrometer	0.5 nM	2 nM-5 μM	＞29,000	23
Evanescent wave all-fiber measurement using a home-made platform	20 nM	0-10 μM	NA[d]	24
Colorimetric analysis with UV/Vis spectroscopy recorded on a PerkinElmer Lambda 25 UV−vis spectrophotometer	59.3 pM	0-1 nM	~9,800-14,000	25
Colorimetric analysis with UV/Vis spectroscopy recorded on a Shimadzu UV2550 spectrophotometer	2 μM	2-250 μM	~7,000-9,800	26
Colorimetric analysis with UV/Vis spectroscopy recorded on a PerkinElmer Lambda 950 spectrophotometer	13 nM	0.03-2 μM	~70,000-127,000	27
Colorimetric analysis with UV/Vis spectroscopy recorded on a Shimadzu UV-1800 spectrophotometer	0.02 nM	0.05-5 nM	~5,000	28
Electrochemical measurement using a CHI 660D electrochemical workstation	3 nM	5 nM-2 μM	＞7,000	29
Electrochemical measurement using a CHI760D electrochemistry system	0.012 nM	0.05-100 nM	＞7,000	30
Distance-based measurement in paper devices	50 mg/L (~0.24 mM)	50-100 mg/L (~0.24-2.4 mM)	(No analytical instrument was used and a ruler typically costs < 1 dollar)	31
	NA[d]	0.1-2000 nM		32
	0.5 nM	0.75-50 nM		This work

[a] LOD, limit of detection.
[b] LCR, linear concentration range.
[c] The information on the reference price per instrument was obtained from the internet.
[d] NA, not available on the internet.

Table S5 Recovery of Pb^{2+} in several real water samples.

Sample	Found[a] (nM)	Added (nM)	Total[b] (nM)	Recovery (%)	RSD[c] (%, *n*=6)
Drinking water					
1	0.00	5.00	5.08	101.6	2.99
2	0.00	10.00	9.98	99.8	1.70
3	0.00	20.00	19.7	98.5	2.09
Tap water					
1	0.00	5.00	5.21	104.2	2.70
2	0.00	10.00	9.71	97.1	3.22
3	0.00	20.00	20.73	103.6	3.80
Pond water					
1	0.00	5.00	5.48	109.6	3.70
2	0.00	10.00	10.07	100.7	4.93
3	0.00	20.00	19.08	95.4	3.40

[a] The original Pb^{2+} concentrations in the samples detected using atomic absorption spectroscopy.

[b] The total Pb^{2+} concentrations in the samples determined using the proposed method.

[c] RSD, relative standard deviations.

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