

Supporting Information for:

Label-free Fluorescent Functional DNA Sensors Using Unmodified DNA: A Vacant Site Approach

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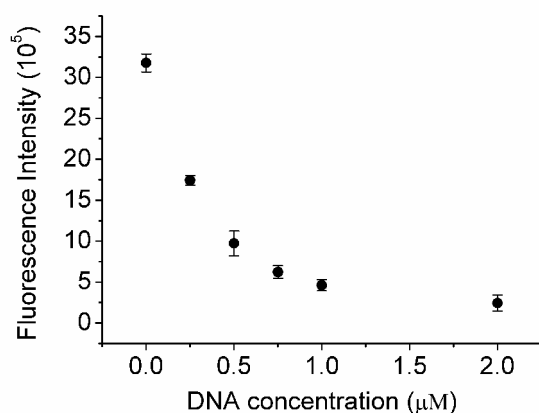


Figure S1. Fluorescence titration of 0.75 μM ATMND with different amounts of 17S_{va}/17E_{va} duplex containing a vacant site in buffer A. The binding constant K_a is calculated to be more than 10^6 M^{-1} according to the 1:1 binding mode, and the fluorescence intensity for each data point was found to be stable at least in one week (within 10% derivation).

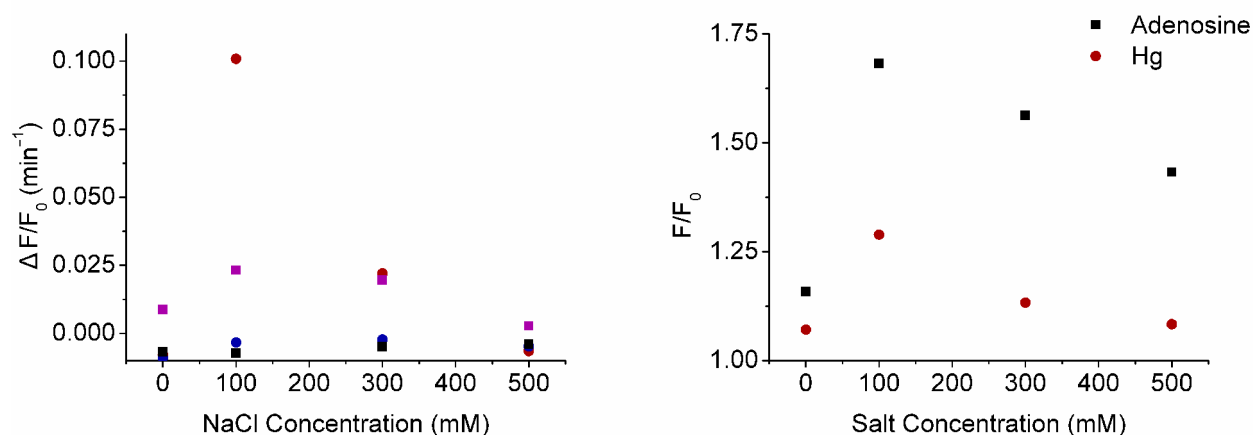


Figure S2. Effect of salt concentration on the performance of DNAzyme (left) and aptamer (right) based sensors in this work. Left: red dots, ATMND/17S_{va}/17E_{va} in the presence of 200 nM Pb²⁺; blue dots, ATMND/17S_{va}/17E_{va} in the absence of Pb²⁺; pink squares: ATMND/39S_{va}/39E_{va} in the presence of 100 nM UO₂²⁺; black squares: ATMND/39S_{va}/39E_{va} in the absence of UO₂²⁺. Right: black squares, ATMND/AdAP_{va}/AdL1_{va} in the presence of 125 μM adenosine; red dots, ATMND/HgAP_{va}/HgL_{va} in the presence of 500 nM Hg²⁺. The salt for Hg²⁺ sensor was NaNO₃, while others were NaCl. The pH was fixed at the values of buffer A~D, respectively.

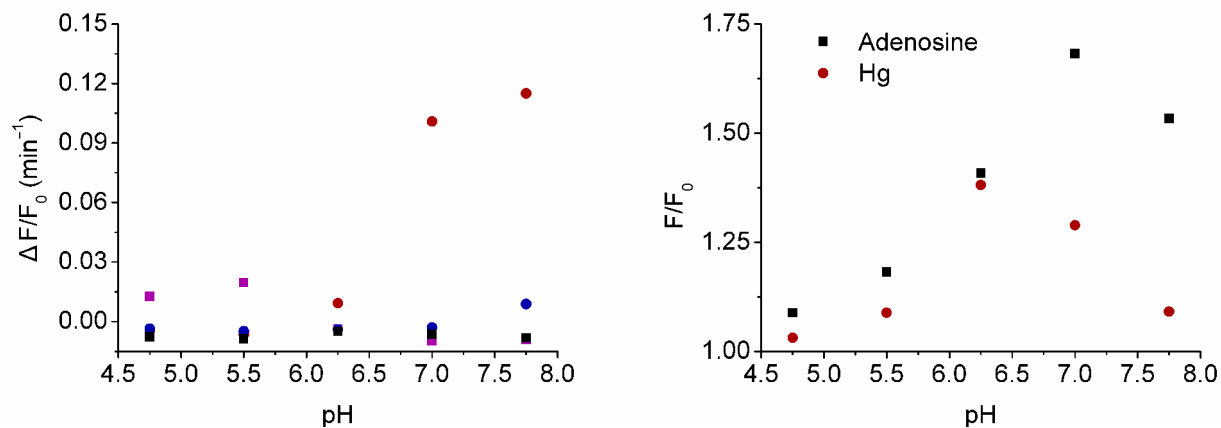


Figure S3. Effect of pH on the performance of DNAzyme (left) and aptamer (right) based sensors in this work. Left: red dots, ATMND/17S_{va}/17E_{va} in the presence of 200 nM Pb²⁺; blue dots, ATMND/17S_{va}/17E_{va} in the absence of Pb²⁺; pink squares: ATMND/39S_{va}/39E_{va} in the presence of 100 nM UO₂²⁺; black squares: ATMND/39S_{va}/39E_{va} in the absence of UO₂²⁺. Right: black squares, ATMND/AdAP_{va}/AdL1_{va} in the presence of 125 μM adenosine; red dots, ATMND/HgAP_{va}/HgL_{va} in the presence of 500 nM Hg²⁺. The pH of 4.75 and 5.5 were adjusted by MES while 7.0 and 7.75 were by HEPES. The pH of 6.25 was checked using both MES and HEPES and the results were similar. Salt concentration was fixed at 100 mM.

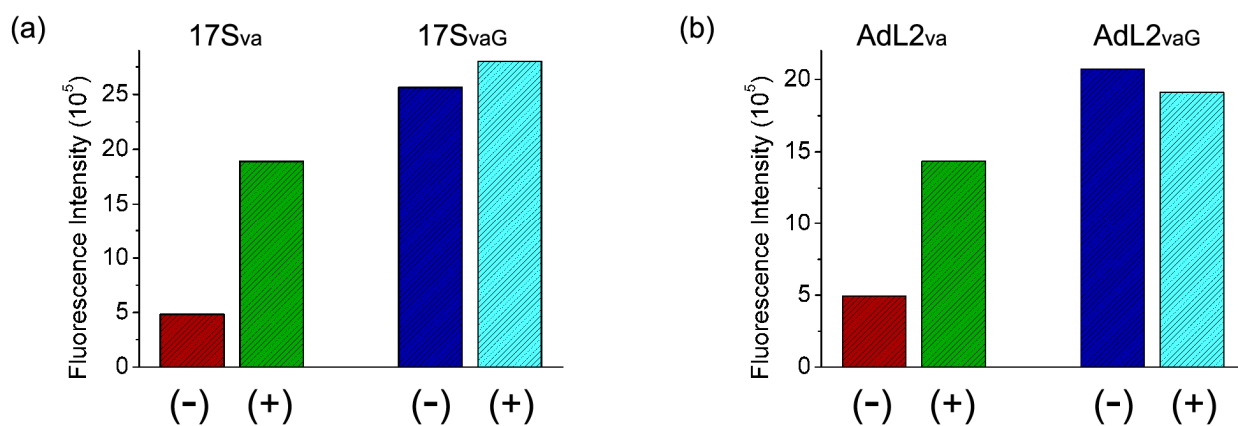


Figure S4. Effect of vacant site on the response of (a) ATMND/17S_{va}(17S_{va}G)/17E_{va} to Pb²⁺, and (b) ATMND/AdAP_{va}/AdL2_{va}(AdL2_{va}G) to adenosine. (-) and (+): in the absence and presence of (a) 1 μM Pb²⁺ or (b) 200 μM adenosine, respectively.

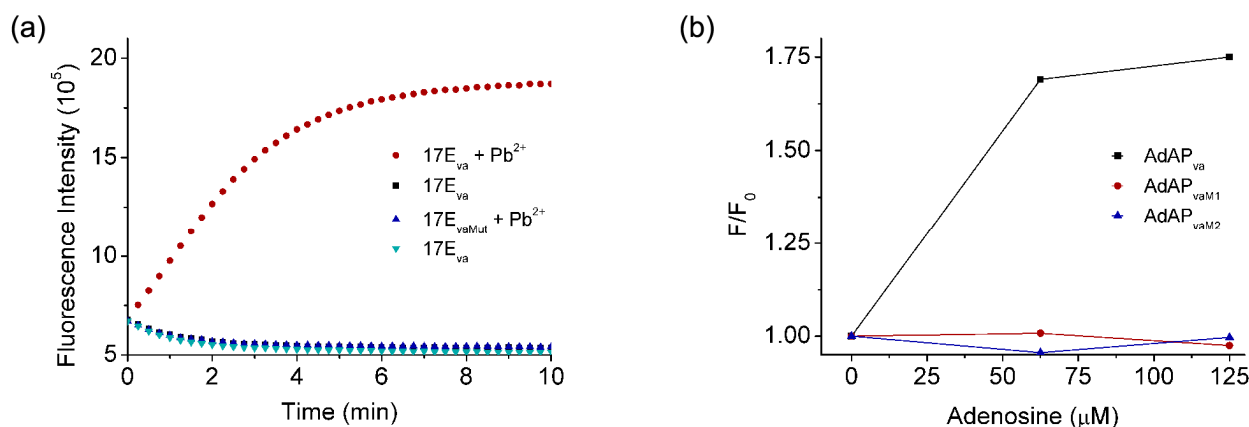


Figure S5. (a) Response of 17E_{va} and its inactive mutant 17E_{vaMut} to Pb²⁺ in the presence of 17S_{va} and ATMND. (b) Response of AdAP_{va} and its inactive mutants AdAP_{vaM1} and AdAP_{vaM2} to adenosine in the presence of AdL1_{va} and ATMND.

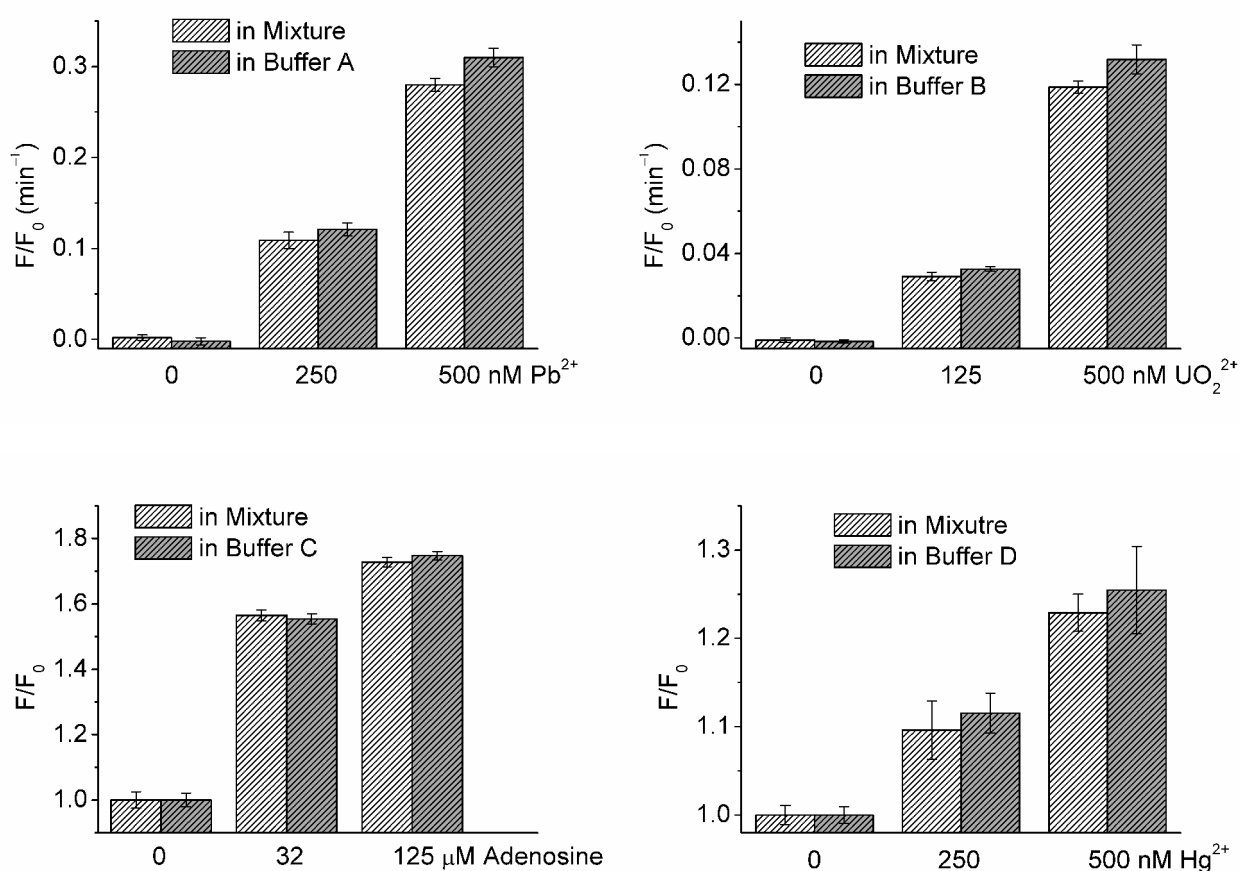


Figure S6. Comparison of the sensor responses in buffer and mixture to evaluate the selectivity and immunity to matrix effect. For the four sensors, 1 μM Cd²⁺, Fe²⁺, Ni²⁺, Co²⁺, Ca²⁺ and Mg²⁺ were added to the buffer as mixture. In addition, 500 nM UO₂²⁺ and Hg²⁺ were added to the mixture for Pb²⁺ sensor; 500 nM Pb²⁺ and Hg²⁺ were added to the mixture for UO₂²⁺ sensor; 500 nM Pb²⁺ and UO₂²⁺; 500 nM Pb²⁺, UO₂²⁺, Hg²⁺ and 500 μM cytidine, uridine were added to the mixture for adenosine sensor.

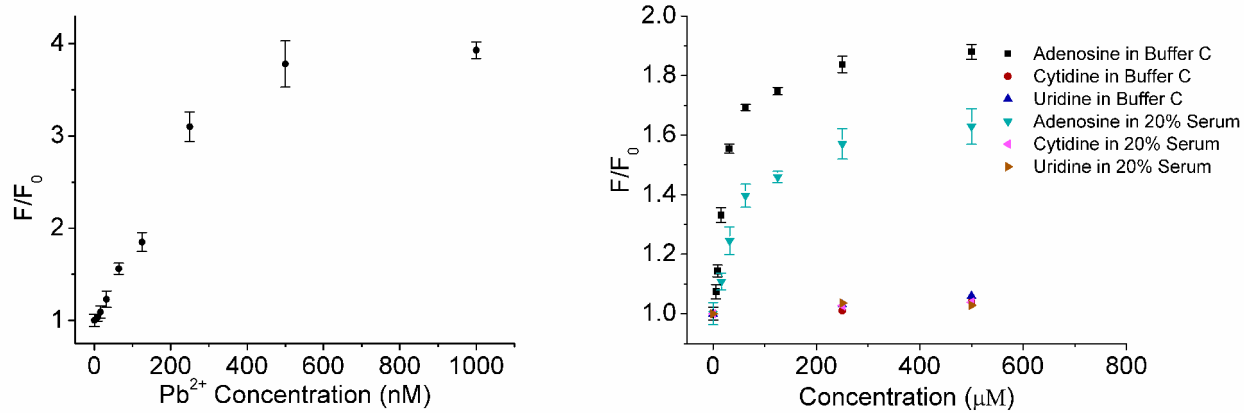


Figure S7. Calibration curves for quantification of Pb^{2+} (left) and adenosine (right, light blue) in drinking water and human serum, respectively.