Supplementary information for

Discrimination of nucleoside phosphates using principal component analysis of spectral changes in a single europium complex

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Contents

1. Materials and instrumental details

NMR spectra were recorded with a Bruker AVIIIHD 400 NanoBay, Bruker AVII 500, Bruker AVII 500 with ^{13}C detect He CryoProbe™, Bruker AVIIIHD 500, or Bruker AVIIIHD 600 with broadband Prodigy CryoProbe™.

Mass spectrometry used a Waters LCT Premier or an Agilent 6120 mass spectrometer equipped with Agilent 1260 LC pump and CTC autosampler, and high resolution and accurate mass spectra were performed by staff at the Chemistry Research Laboratory, University of Oxford, using a Bruker μTOF or Waters Micromass® GCT.

CHN elemental analysis was performed Mrs Birgitta Kegel at the University of Copenhagen, Denmark.

UV-vis absorption spectra used a PerkinElmer Lambda 750S. Spectra were measured in solution using pure water as the baseline.

Luminescence data was measured using a Horiba FluoroLog-3 and processed using FluorEssence™, OriginLab Origin®, a custom-written Microsoft Excel macro, and DynaFit®.[1]

Reagents were laboratory grade and obtained from standard commercial sources including Sigma-Aldrich, Fisher Scientific, Alfa Aesar, Fluorochem, and CheMatech. Water was deionised and microfiltered with a Milli-Q Millipore machine.

Principal component analysis was performed using Quasar 1.9.1 (Orange), and graphs were plotted in OriginLab Origin®.

2. Experimental methods

2.1 Titration experiments

Stock solutions (2.6×10^{-4} M) of the europium complex were made up in aqueous HEPES buffer solution (0.1 M) and the pH adjusted to 7.4 with a Hanna benchtop pH meter (±0.01 pH units) with microlitres of concentrated $HCl_{(aq)}$ or $NaOH_{(aq)}$. Stock solutions of AMP (1.6×10^{-2} M), ADP (1.6×10^{-2} M), and ATP (1.7×10^{-2} M) were made using the europium complex stock solution as the solvent, to ensure that the europium complex concentration was kept constant so that the luminescence intensity was not altered by dilution. All solids for the solutions were weighed out on a balance (±0.01 mg).

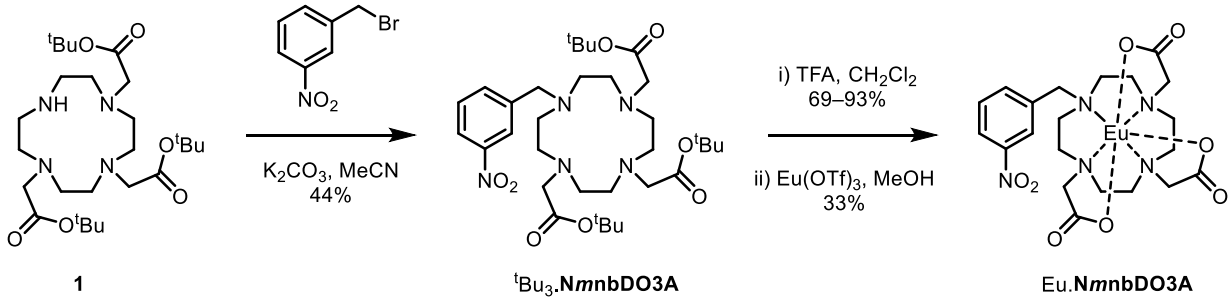
1.00 mL of the europium complex stock solution was added to a 1.5 mL cuvette with a volume-displacement micropipette. During the titration, successive emission spectra were recorded as 2 to 130 μL aliquots of the nucleoside phosphate stock solution were added using a volume-displacement micropipette. Titrations were carried out at room temperature on the same day. All solutions were allowed to equilibrate to room temperature before use.

2.2 Principal component analysis

Raw spectra were processed using Quasar 1.9.1 (Orange). Full spectra were first subjected to linear baseline correction, followed by Gaussian smoothing (SD = 1), and finally area

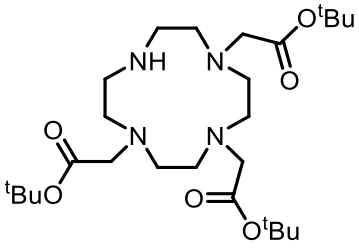
normalisation. Processed spectral data was then subjected to principal component analysis (PCA).

3. Synthetic details



Scheme S1: Synthesis of complex Eu.**N*m*nbDO3A**.

1,4,7,10-Tetraazacyclododecane-1,4,7-triacetate *tert*-butyl triester · HBr (1)



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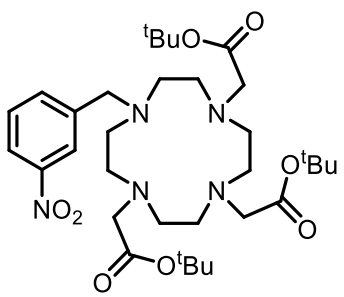
Prepared according to the procedure described by Dadabhoy *et al.*[2]

Yield: 8.76 g, 42%. ^{1}H NMR ($CDCl_3$, 400 MHz) δ (ppm) 10.02 (1H, br. s, NH), 3.36 (4H, s, NCH_2CO), 3.28 (2H, s, NCH_2CO), 3.09 (4H, br. m, ring NCH_2), 2.89 (12H, br. m, ring NCH_2), 1.44 (27H, s, 3 × ^{t}Bu). ^{13}C NMR ($CDCl_3$, 101 MHz) δ (ppm) 170.61, 169.72, 81.96, 81.80, 58.35, 51.49, 51.37, 49.33, 48.92, 47.65, 28.36 (1 × ^{t}Bu), 28.32 (2 × ^{t}Bu). ES^+ MS (*m/z*) 515 ($[M + H]^+$), 537 ($[M + Na]^+$). ES^- MS (*m/z*) 515 (M^-).

α-(DO3A *tert*-butyl triester)-3-methylnitrobenzene (^{t}Bu$_3$.N*m*nbDO3A)



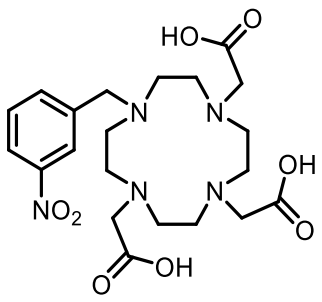
^{t}Bu$_3$.N*m*nbDO3A

3-Nitrobenzylbromide (0.42 g, 1.94 mmol), compound **1** · HBr (1.00 g, 1.68 mmol), and potassium carbonate (1.30 g, 9.41 mmol) were combined in anhydrous acetonitrile (50 mL)

and the mixture stirred at 55°C for 3.5 hours. The white suspension was filtered to remove inorganic salts and the resulting yellow solution evaporated under reduced pressure. The light yellow solid was dissolved in dichloromethane (30 mL) and washed with brine (3 × 30 mL). The organic layer was dried over anhydrous sodium sulphate, filtered, and the filtrate concentrated to dryness under reduced pressure. The crude product was purified by silica gel column chromatography in 19:1 dichloromethane:methanol, yielding a static very pale yellow solid (482 mg, 44%).

^{1}H NMR ($CDCl_3$, 400 MHz) δ (ppm) 8.71 (1H, t, *J*=2.1 Hz, Ar-H2), 8.13 (1H, ddd, *J*=7.9, 2.1, 1.2 Hz, Ar-H4), 7.76 (1H, dt, *J*=7.9, 1.2 Hz, Ar-H6), 7.53 (1H, t, *J*=7.9 Hz, Ar-H5), 3.98 – 1.98 (24H, br. m, NCH_2), 1.42 (27H, br. s, 3 × ^{t}Bu). ^{13}C NMR ($CDCl_3$, 126 MHz) δ (ppm) 173.45, 172.51, 148.53, 139.60, 137.10, 129.54, 124.71, 122.51, 83.10, 82.55, 58.76, 55.93, 55.59, 27.79. ES^+ MS (*m*/*z*) 650 ($[M + H]^+$), 672 ($[M + Na]^+$), 594 ($[M – {}^{t}Bu + 2H]^+$), 538 ($[M – 2{}^{t}Bu + 3H]^+$), 482 ($[M – 3{}^{t}Bu + 4H]^+$).

α-(H$_3$.DO3A)-3-methylnitrobenzene · 2TFA (H$_3$.N*m*nbDO3A)

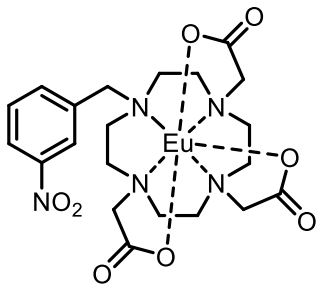


H$_3$.N***m*nbDO3A**

^{t}Bu$_3$.N***m*nbDO3A** (100 mg, 0.15 mmol) was dissolved in dichloromethane (2 mL) and trifluoroacetic acid (2 mL) added dropwise to the stirring solution. The reaction mixture was stirred at room temperature for 26 hours, monitored by mass spectrometry. The solvent was removed under reduced pressure. Residual trifluoroacetic acid was co-evaporated with methanol three times. The crude product was dissolved in the minimum amount of methanol and precipitated with excess diethyl ether, and the diethyl ether removed by decantation. The methanol-diethyl ether trituration was performed three times. The resulting white powder was dried under reduced pressure (75 mg, 69%).

^{1}H NMR (MeOD, 400 MHz) δ (ppm) 8.37 (1H, s, Ar-H2), 8.23 (1H, d, *J*=8.2 Hz, Ar-H4), 8.07 (1H, d, *J*=7.8 Hz, Ar-H6), 7.60 (1H, t, *J*=7.8 Hz, Ar-H5), 4.23 (2H, br. s, $ArCH_2$), 3.55 (22H, br. m, CH_2 cyclen ring, CH_2CO). ES^+ MS (*m*/*z*) 482 ($[M + H]^+$), 504 ($[M + Na]^+$). ESI^+ HRMS (*m*/*z*) calculated: 482.22454, measured: 482.22448 $[C_{21}H_{32}N_5O_8]^+$ ($[M + H]^+$). Elemental calculated: C, 42.32; H, 4.69; N, 9.87; measured: C, 40.51; H, 4.97; N, 9.84; $[C_{25}H_{33}F_6N_5O_{12}]$ (M · 2TFA).

***α*-(Eu.DO3A)-3-methylnitrobenzene · 3NaOTf (Eu.N*m*nbDO3A)**



Eu.**N*m*nbDO3A**

H_3.**N*m*nbDO3A** · 2TFA (45 mg, 0.063 mmol) and europium triflate (59 mg, 0.098 mmol) were combined in methanol (5 mL) and stirred at 60°C for 21 hours, then adjusted to pH 5 by dropwise addition of aqueous sodium hydroxide (1.0 M) and further reacted at 60°C for 5 days. The methanol was removed under reduced pressure, the resulting suspension dissolved in water and adjusted to pH 10 by dropwise addition of aqueous sodium hydroxide (1.0 M). The resulting suspension was centrifuged and the supernatant filtered through an Acrodisc® syringe filter (0.2 μm), neutralised to pH 7 by dropwise addition of aqueous hydrochloric acid (1.0 M), and the water removed under reduced pressure followed by desiccation. The salt content of the crude product was reduced by dialysis in a Float-A-Lyzer® G2. The water was removed under reduced pressure and the product dried in a desiccator (24 mg, 33%).

^{1}H NMR (D_2O, 400 MHz) δ (ppm) 37.43, 27.82, 27.77, 17.49, 12.84, −2.31, −5.15, −10.04, −10.06, −11.94, −15.31, −16.66, −18.67, −19.32, −20.63, −24.10, −24.42, −25.33. Only major resolved peaks outside the +10 to 0 ppm range are reported. ES^+ MS (*m*/*z*) 632 ($[M + H]^+$), 1261 ($[2M + H]^+$). ESI^+ HRMS (*m*/*z*) calculated: 632.1223, measured: 632.1210 $[C_{21}H_{29}EuN_5O_8]^+$ ($[M + H]^+$). UV-vis (H_2O) λ (I/I_{max}) 204 (1.00), 268 (0.48) nm.

4. Supplementary figures

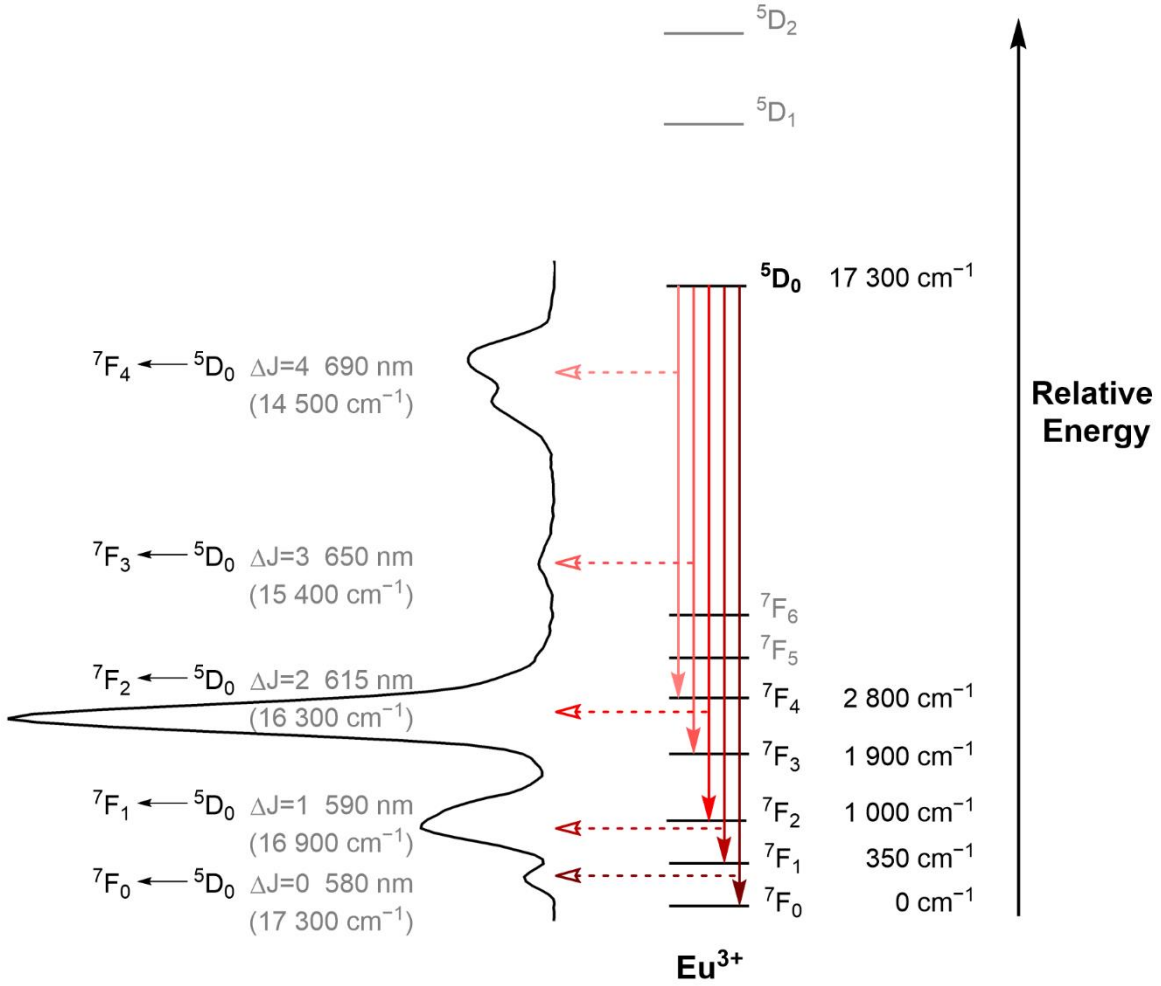


Figure S1: Partial energy level diagram outlining relevant transitions for Eu^{3+} complexes.

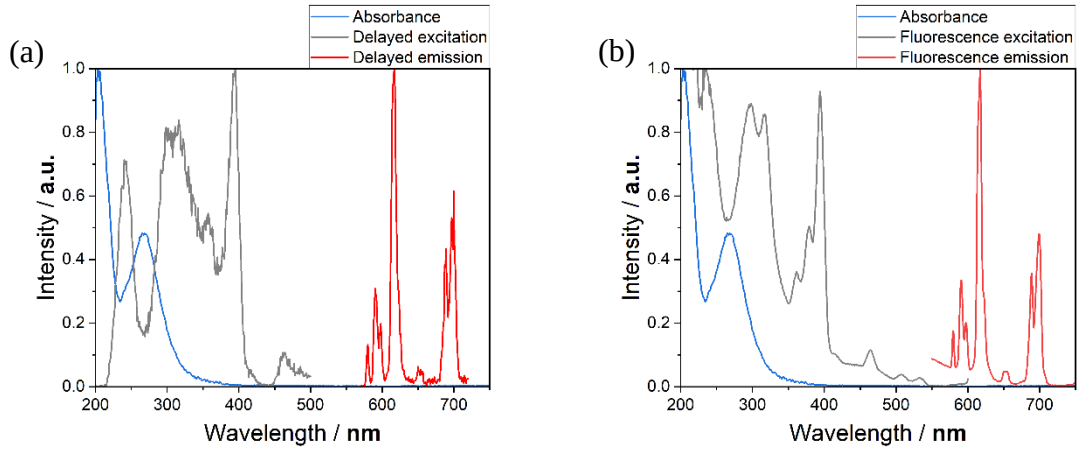


Figure S2: UV-vis absorption (blue traces), (a) time-gated luminescence (λ_{ex} = 394 nm, λ_{em} = 616 nm, delay time 0.10 ms, sample window 0.2 ms), (b) steady state fluorescence (λ_{ex} = 394 nm, λ_{em} = 616 nm) excitation (grey traces) and emission (red traces).

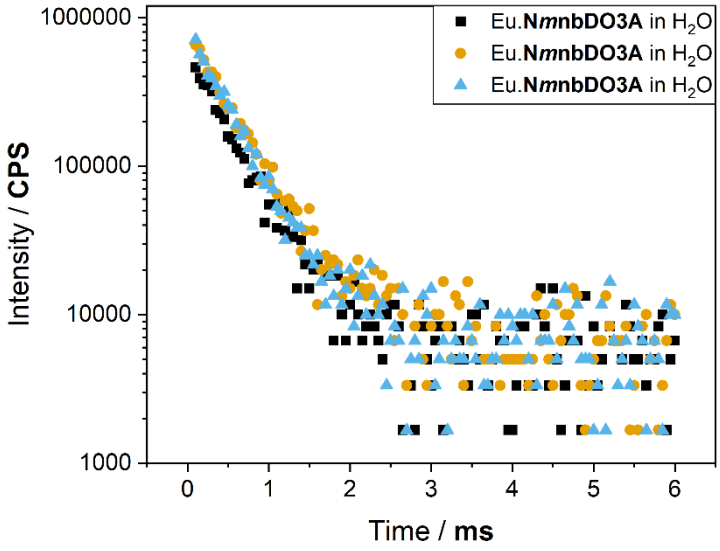


Figure S3: Eu.**N*m*nbDO3A** luminescence lifetime in H_2O. τ = 0.41 ms.

λ_{ex} = 394 or 230 nm, s.w.$_{ex}$ = 14 nm, λ_{em} = 616 nm, s.w.$_{em}$ = 14 or 10 or 5 nm, sample window 0.2 ms, time per flash 41.00 ms, flash count 10, 3 averaged scans.

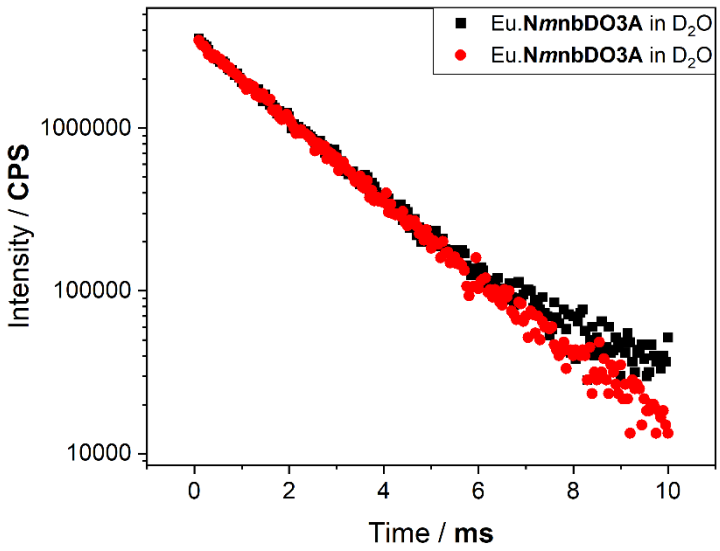


Figure S4: Eu.**N*m*nbDO3A** luminescence lifetime in D_2O. τ = 1.70 ms.

λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, λ_{em} = 616 nm, s.w.$_{em}$ = 14 nm, sample window 0.2 ms, time per flash 41.00 ms, flash count 10, 3 averaged scans.

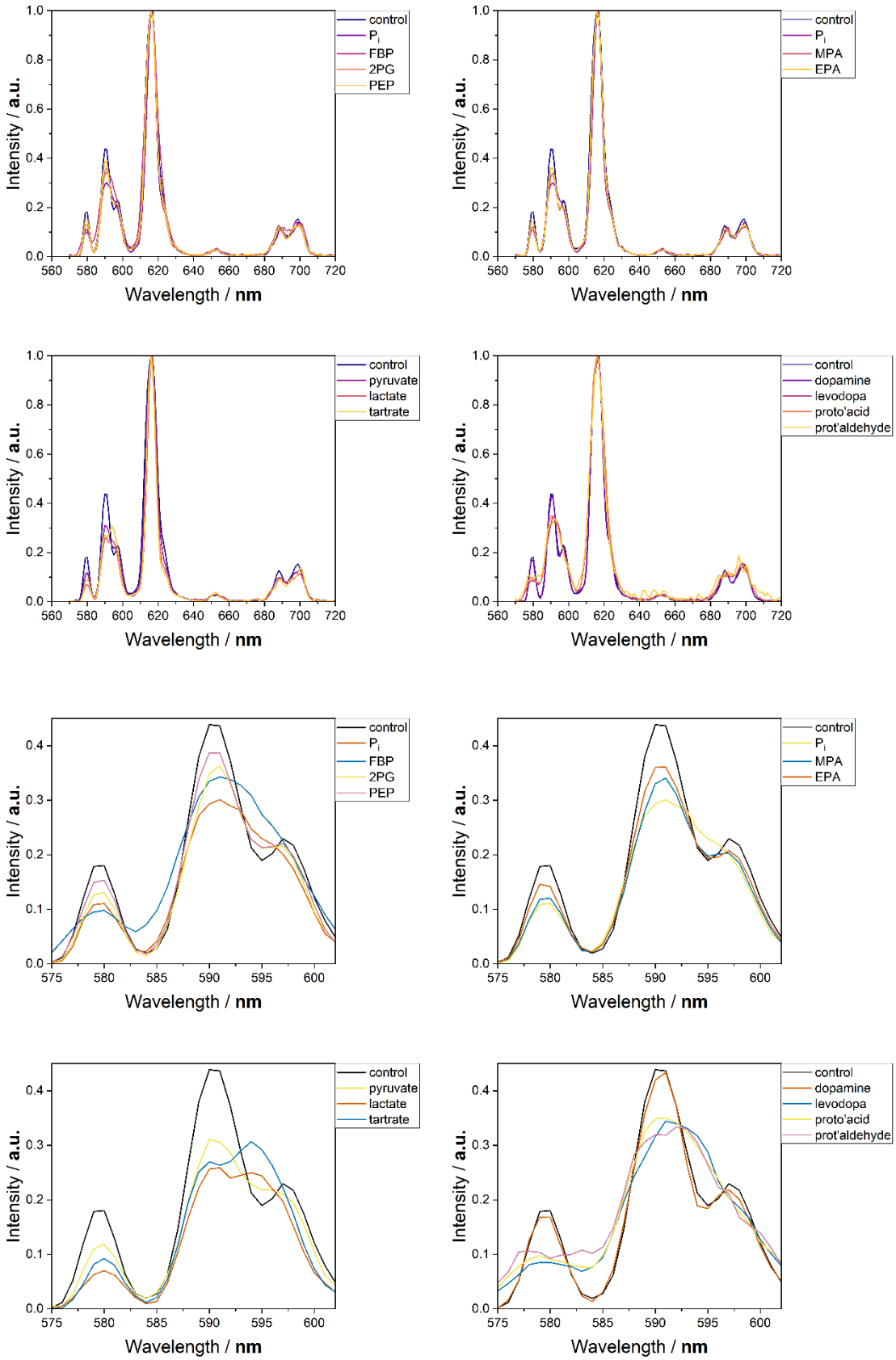


Figure S5: Average spectra for each condition for Eu.**N*m*nbDO3A** with other anionic guests. Top: full emission spectra, bottom: ΔJ=0 peak and fine structure in ΔJ=1 peak. P_i = inorganic phosphate, FBP = *D*-fructose 1,6-bisphosphate, 2PG = *D*(+)2-phosphoglycerate, PEP = phospho(enol)pyruvate, MPA = methylphosphonate, EPA = ethylphosphonate, proto'acid = 3,4-dihydroxybenzoic acid, prot'aldehyde = 3,4-dihydroxybenzaldehyde.

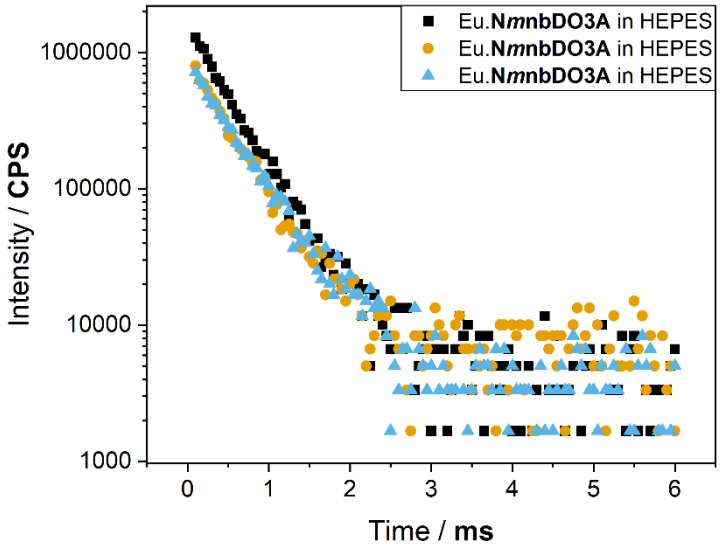


Figure S6: Eu.**N*m*nbDO3A** luminescence lifetime in HEPES solution at pH 7.4. τ = 0.41 ms. [Eu.**N*m*nbDO3A**] = 0.26 mM, λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, λ_{em} = 616 nm, s.w.$_{em}$ = 5 or 4 nm, sample window 0.2 ms, time per flash 41.00 ms, flash count 10, 3 averaged scans.

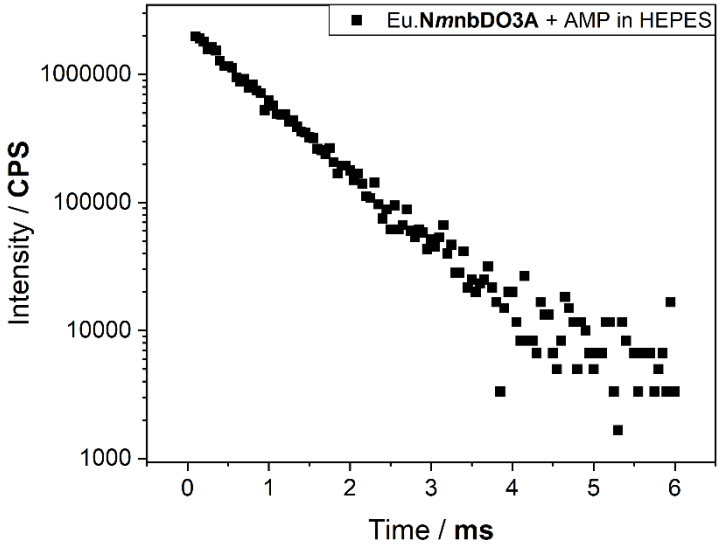


Figure S7: Eu.**N*m*nbDO3A** + AMP luminescence lifetime in HEPES solution at pH 7.4. τ = 0.74 ms.

[Eu.**N*m*nbDO3A**] = 0.26 mM, [AMP] = 4.8 mM (18 equivalents), λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, λ_{em} = 616 nm, s.w.$_{em}$ = 5 nm, sample window 0.2 ms, time per flash 41.00 ms, flash count 10, 3 averaged scans.

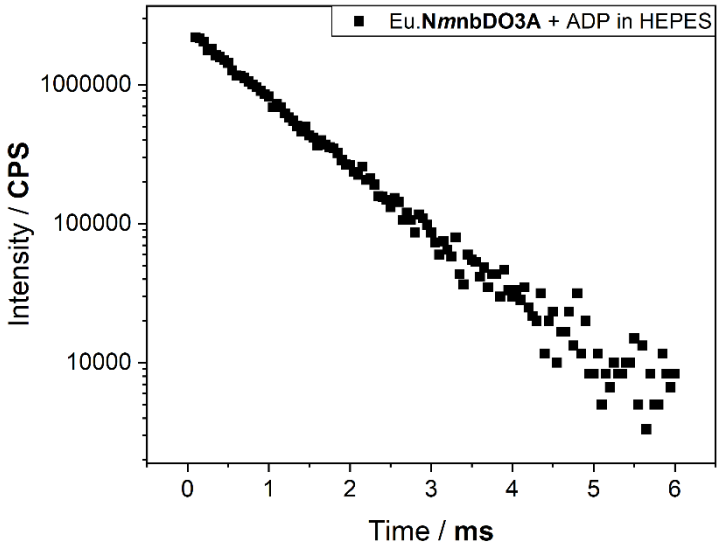


Figure S8: Eu.N*m*nbDO3A + ADP luminescence lifetime in HEPES solution at pH 7.4. τ = 0.87 ms.

[Eu.N*m*nbDO3A] = 0.26 mM, [ADP] = 4.9 mM (19 equivalents), λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, λ_{em} = 616 nm, s.w.$_{em}$ = 5 nm, sample window 0.2 ms, time per flash 41.00 ms, flash count 10, 3 averaged scans.

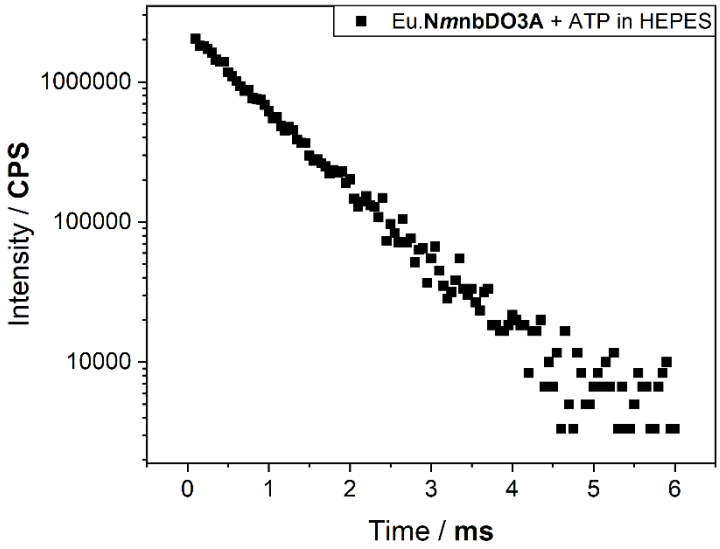


Figure S9: Eu.N*m*nbDO3A + ATP luminescence lifetime in HEPES solution at pH 7.4. τ = 0.76 ms.

[Eu.N*m*nbDO3A] = 0.26 mM, [ATP] = 5.1 mM (20 equivalents), λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, λ_{em} = 616 nm, s.w.$_{em}$ = 5 nm, sample window 0.2 ms, time per flash 41.00 ms, flash count 10, 3 averaged scans.

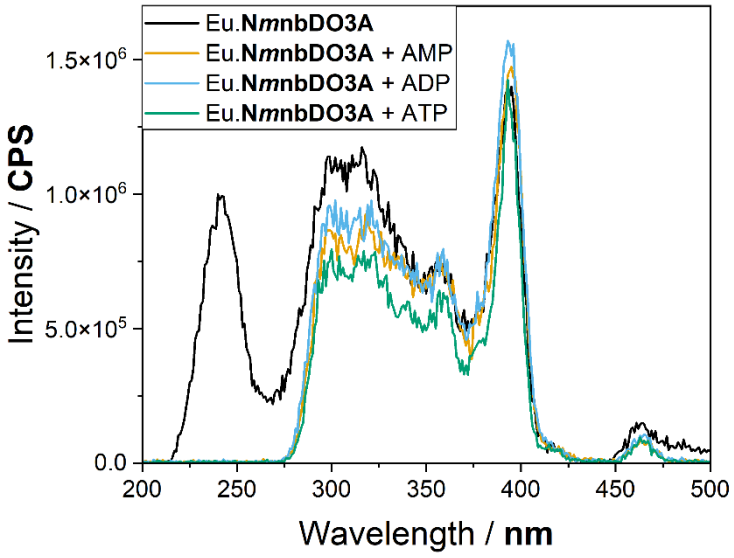


Figure S10: Change in excitation spectrum upon addition of 19 equivalents of adenosine guest to a 0.26 mM solution of Eu.**N*m*nbDO3A** in 0.1 M HEPES buffer at pH 7.4. λ_{em} = 616 nm, s.w.$_{ex}$ = 14 nm (Eu.**N*m*nbDO3A** alone, with AMP, with ADP) or 12 nm (with ATP), s.w.$_{em}$ = 4 nm, flash delay: 0.10 ms, sample window: 0.20 ms, time per flash: 41.00 ms, flash count: 10, integration time 100.00 ms, 3 averaged scans.

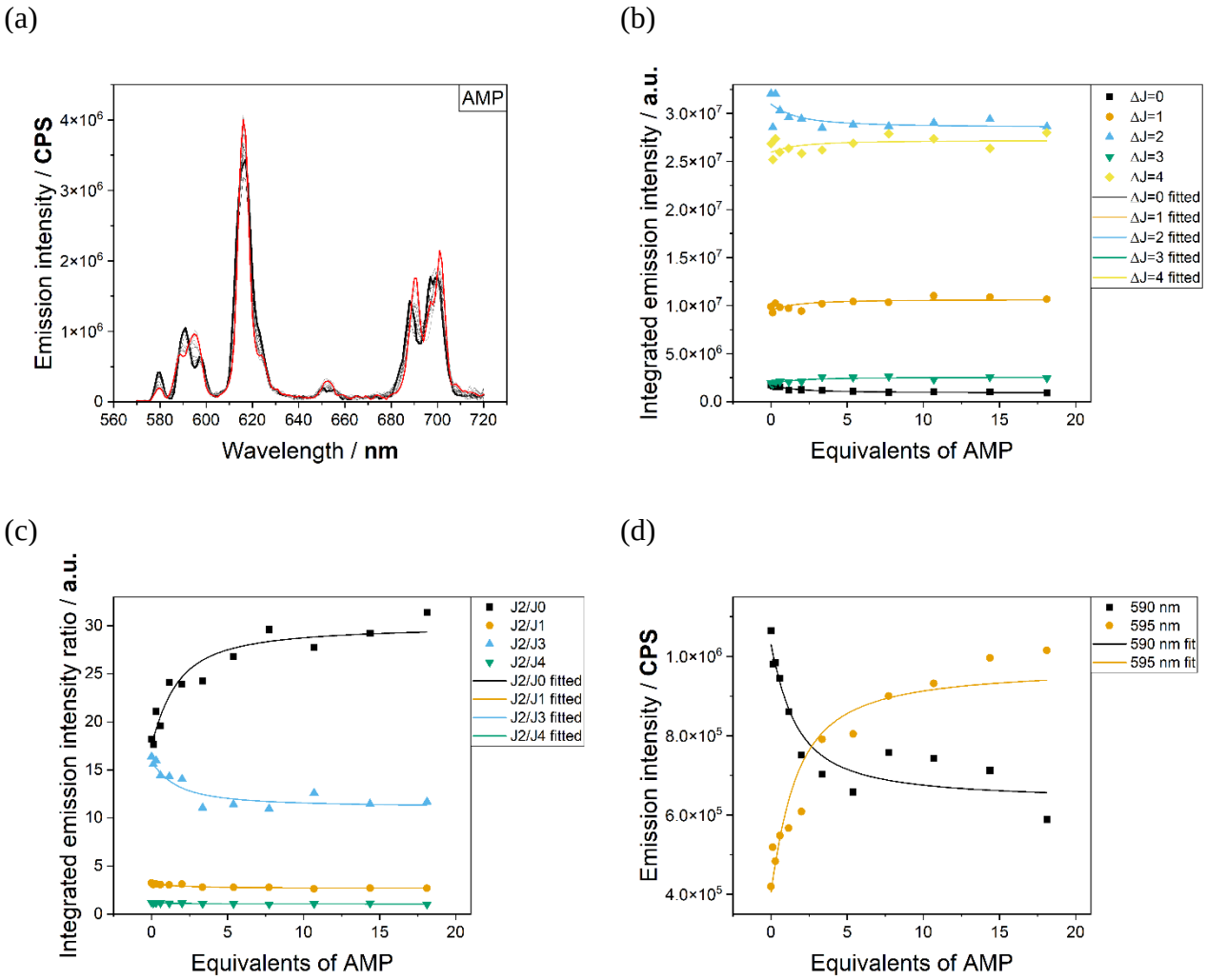


Figure S11: Titration data for Eu.**N*m*nbDO3A** + AMP.

(a) Overlaid luminescence emission spectra for titrations of Eu.**N*m*nbDO3A** with AMP (black trace at start, to grey, to red trace at end) in HEPES at pH 7.4, with data correction files applied for detector sensitivity at longer wavelengths. λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, s.w.$_{em}$ = 3 nm, flash delay: 0.10 ms, sample window: 0.20 ms, time per flash: 41.00 ms, flash count: 10, integration time 100.0 ms, 5 averaged scans, with 5 points Savitzky-Golay smoothing applied.

(b) Binding isotherms from integration of europium emission peaks.

(c) Binding isotherms from ratios of integration of hypersensitive ΔJ=2 peak relative to other peaks.

(d) Binding isotherms from intensities at wavelengths relevant to the splitting of the ΔJ=1 peak.

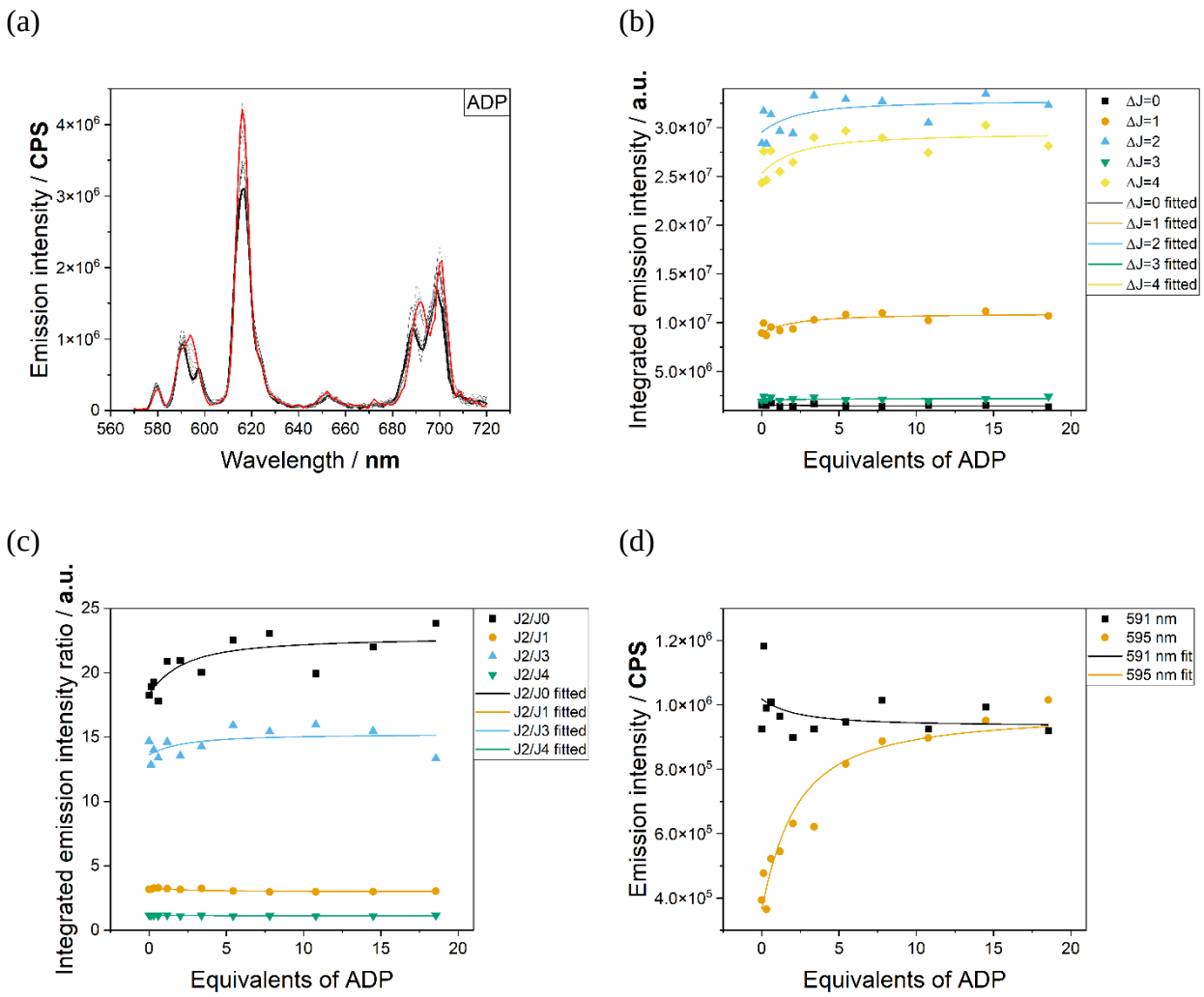


Figure S12: Titration data for Eu.N*m*nbDO3A + ADP.

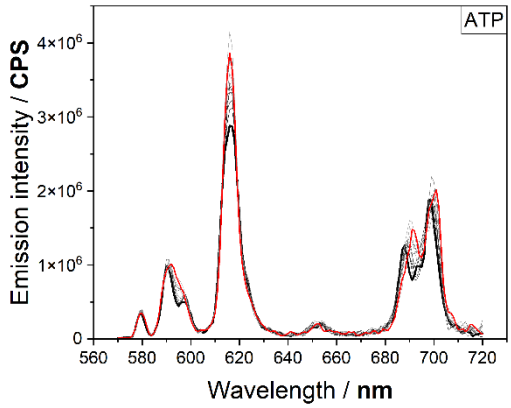
(a) Overlaid luminescence emission spectra for titrations of Eu.N*m*nbDO3A with ADP (black trace at start, to grey, to red trace at end) in HEPES at pH 7.4, with data correction files applied for detector sensitivity at longer wavelengths. λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, s.w.$_{em}$ = 3 nm, flash delay: 0.10 ms, sample window: 0.20 ms, time per flash: 41.00 ms, flash count: 10, integration time 100.0 ms, 5 averaged scans, with 5 points Savitzky-Golay smoothing applied.

(b) Binding isotherms from integration of europium emission peaks.

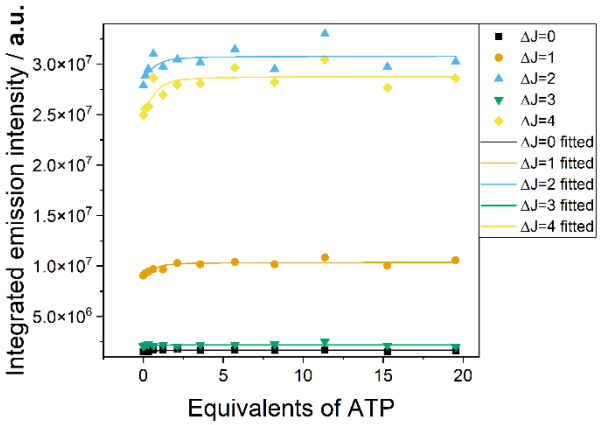
(c) Binding isotherms from ratios of integration of hypersensitive ΔJ=2 peak relative to other peaks.

(d) Binding isotherms from intensities at wavelengths relevant to the splitting of the ΔJ=1 peak.

(a)



(b)



(c)

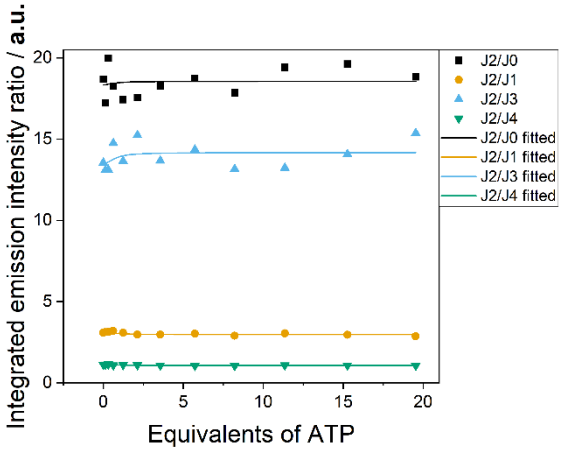


Figure S13: Titration data for Eu.**N*m*nbDO3A** + ATP.

(a) Overlaid luminescence emission spectra for titrations of Eu.**N*m*nbDO3A** with ATP (black trace at start, to grey, to red trace at end) in HEPES at pH 7.4, with data correction files applied for detector sensitivity at longer wavelengths. λ_{ex} = 394 nm, s.w.$_{ex}$ = 14 nm, s.w.$_{em}$ = 3 nm, flash delay: 0.10 ms, sample window: 0.20 ms, time per flash: 41.00 ms, flash count: 10, integration time 100.0 ms, 5 averaged scans, with 5 points Savitzky-Golay smoothing applied.

(b) Binding isotherms from integration of europium emission peaks.

(c) Binding isotherms from ratios of integration of hypersensitive ΔJ=2 peak relative to other peaks.

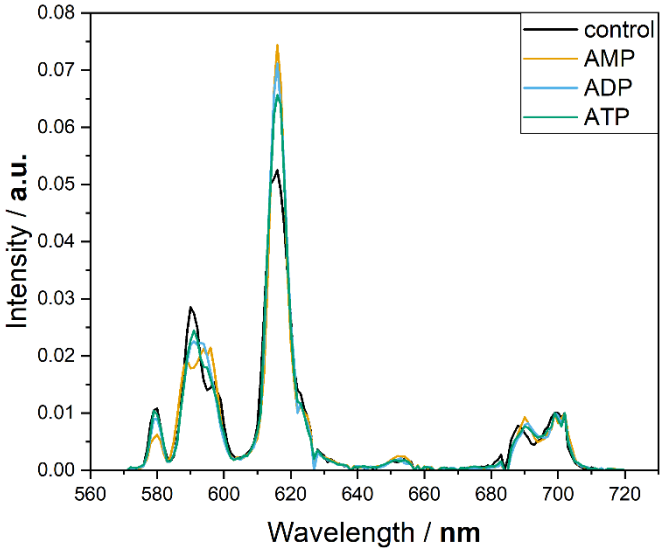


Figure S14: Average spectra for each condition.

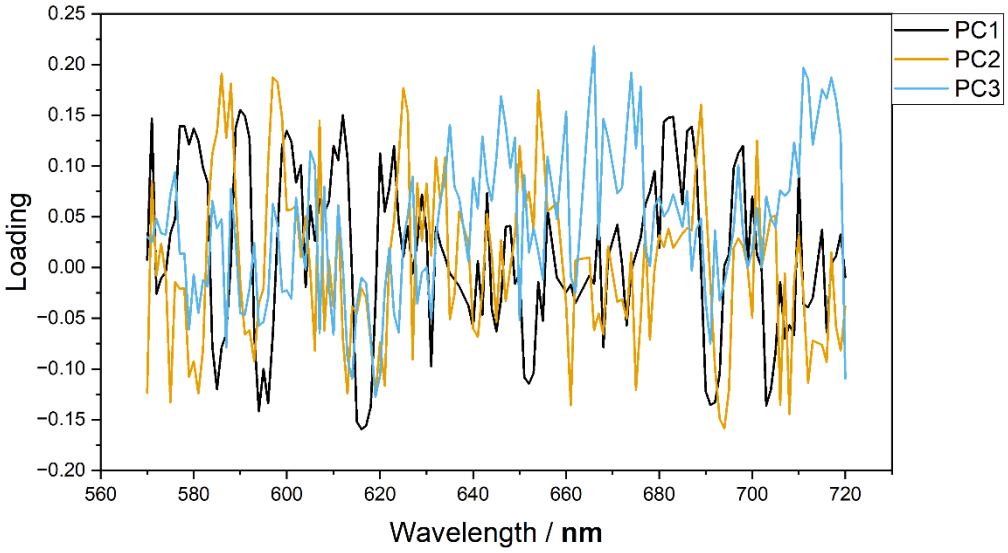


Figure S15: PCA scores plot against wavelengths.

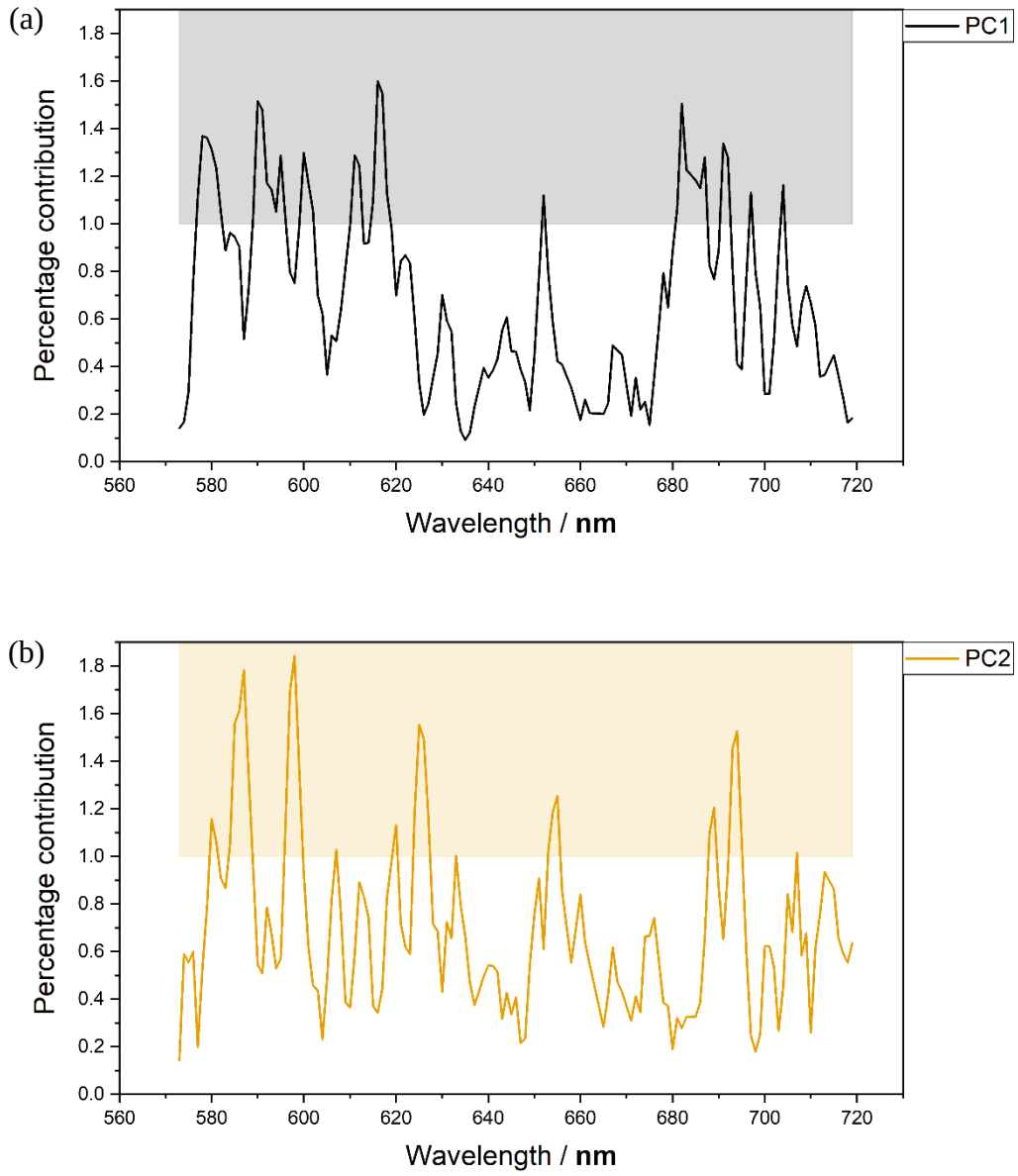
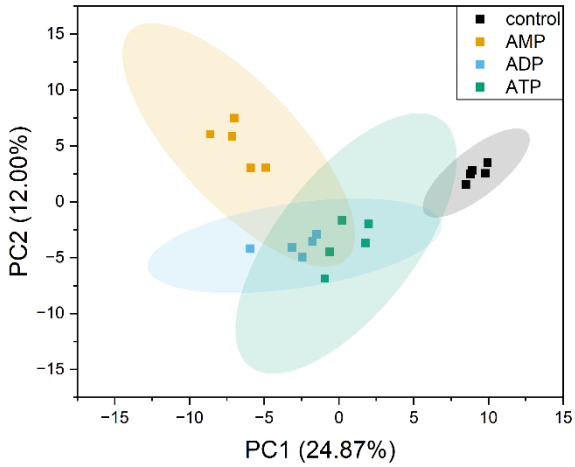
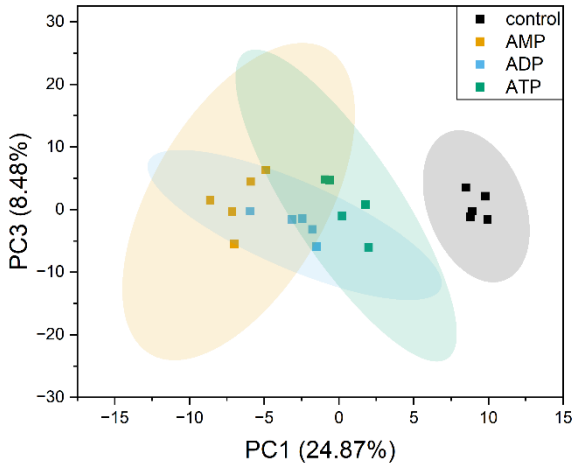


Figure S16: Percentage contribution of each wavelength to (a) principal component 1 (PC1) and (b) principal component 2 (PC2). Three-point smoothing applied, absolute values of loadings taken. Shaded area signifies wavelengths that contribute to more than 1% of the respective PC.

(a)



(b)



(c)

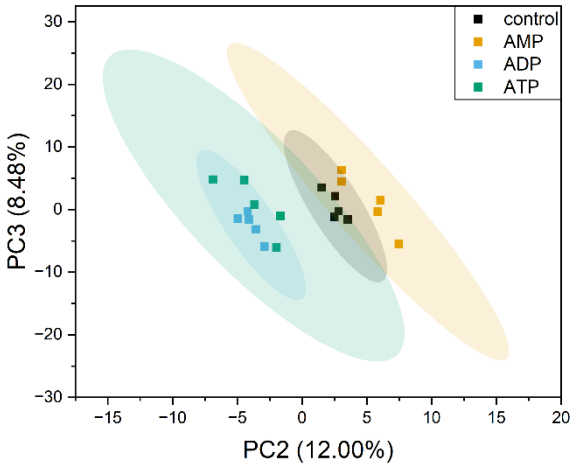


Figure S17: Plot of (a) PC2 vs PC1, (b) PC3 vs PC1, and (c) PC3 vs PC2 for the principal component analysis of baseline-corrected and area normalised spectra of Eu.**N*m*nbDO3A** (control) and Eu.**N*m*nbDO3A** treated with AMP, ADP, and ATP (5–20 equivalents). 0.26 mM Eu.**N*m*nbDO3A** in 0.1 M HEPES buffer, λ_{ex} = 394 nm. Ellipses represent 95% confidence intervals.

Table S1: Contribution of components to variance of data.

	% variance	cumulative variance
PC1	24.87	24.87
PC2	12.00	36.88
PC3	8.48	45.35
PC4	5.79	51.14
PC5	5.10	56.24
PC6	4.87	61.11
PC7	4.59	65.71
PC8	4.16	69.86
PC9	3.82	73.68
PC10	3.53	77.20

Table S2: PC loadings for the major peaks in the Eu.**N*m*nbDO3A** spectrum.

Wavelength / nm	Assignment	PC1	PC2	PC3
580	$\Delta J=0$	0.137	−0.0926	−0.00688
590	$\Delta J=1$ (1^{st} max.)	0.155	−0.0150	−0.0450
598	$\Delta J=1$ (2^{nd} max.)	0.0326	0.183	0.0425
616	$\Delta J=2$	−0.159	−0.0204	−0.0101
689	$\Delta J=4$ (1^{st} max.)	−0.00162	0.160	0.0481
698	$\Delta J=4$ (2^{nd} max.)	0.119	0.0211	0.0349

5. References

1 P. Kuzmič, *Anal. Biochem.*, 1996, **237**, 260–273.

2 A. Dadabhoy, S. Faulkner and P. G. Sammes, *J. Chem. Soc., Perkin Trans. 2*, 2002, 348–357.

6. Full titration data

EuNmnbDO3A_AMP_LH230_S1,,,,,,,,,,,
,0,0.11832,0.29491,0.58691,1.16231,2.00456,3.35533,5.38887,7.73186,10.6894,14.37032,18.11259
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692,57000,61000,53000,78000,68000,72000,79000,89000,90000,95000,85000,119000
693,75000,54000,66000,53000,75000,64000,79000,93000,82000,97000,76000,87000
694,73000,75000,55000,84000,84000,89000,105000,72000,93000,81000,77000,82000
695,58000,72000,72000,97000,60000,85000,83000,102000,83000,85000,75000,77000
696,84000,92000,93000,90000,94000,91000,81000,86000,101000,88000,82000,77000
697,96000,115000,89000,118000,100000,99000,98000,98000,102000,104000,100000,100000
698,138000,121000,122000,109000,122000,98000,135000,116000,88000,109000,102000,115000
699,112000,111000,113000,123000,122000,118000,96000,152000,105000,124000,114000,118000
700,96000,123000,116000,122000,119000,127000,98000,130000,115000,122000,133000,117000
701,80000,77000,101000,97000,103000,121000,94000,108000,89000,113000,102000,127000
702,59000,63000,73000,76000,57000,104000,94000,103000,103000,100000,85000,112000
703,41000,51000,41000,61000,63000,39000,64000,67000,74000,77000,68000,71000
704,32000,33000,30000,42000,32000,37000,29000,51000,45000,41000,50000,45000
705,15000,11000,20000,29000,19000,22000,27000,28000,22000,26000,21000,23000
706,19000,10000,10000,12000,16000,12000,13000,14000,22000,26000,26000,25000
707,10000,6000,11000,13000,17000,20000,16000,8000,8000,19000,16000,16000
708,6000,11000,10000,5000,8000,6000,10000,10000,12000,7000,16000,18000
709,15000,9000,6000,10000,12000,8000,8000,8000,6000,8000,8000,13000
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711,5000,1000,8000,6000,6000,5000,3000,9000,7000,7000,13000,10000
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715,2000,6000,6000,4000,6000,2000,9000,6000,11000,4000,9000,8000
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717,0,6000,4000,8000,7000,4000,3000,7000,5000,5000,7000,5000
718,5000,6000,2000,4000,5000,6000,5000,4000,5000,6000,6000,6000
719,3000,2000,2000,5000,3000,7000,4000,8000,4000,5000,5000,5000
720,3000,5000,1000,4000,9000,8000,12000,3000,8000,5000,3000,2000