

Supporting Information

Crystallographic and Spectroscopic Characterization of Americium Complexes Containing the Bis[(phosphino)methyl]pyridine-1-oxide (NOPOPO) Ligand Platform

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Table of Contents

Sample configuration for X-ray analysis	S3
Experimental details and structure of $[\text{Am}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, Am 2:1 , at 80 K.....	S3
Experimental details and structure of $[\text{Nd}(\mathbf{L})_2(\text{NO}_3)][\text{Nd}(\text{NO}_3)_5]$	S5
Luminescence decay curves for Eu 2:1 and Am 2:1	S7
ATR spectra of Am 2:1 , Eu 2:1 , Nd 2:1 and ligand starting material L	S8
X-ray data collection, structure solution, and refinement details for:	
$[\text{Eu}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, Eu 2:1 , (Tables S1-S4).....	S9
$[\text{Nd}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, Nd 2:1 , (Tables S5-S8).....	S25
$[\text{Am}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, Am 2:1 , (Tables S9-S12).....	S42
$[\text{Eu}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$ at 80K, Eu 2:1 , (Tables S13-S16).....	S57
$[\text{Nd}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$ at 80K, Nd 2:1 , (Tables S17-S20).....	S74
$\text{Nd}(\mathbf{L})(\text{NO}_3)_3$, Nd 1:1 , (Tables S21-S24).....	S91
$\text{Am}(\mathbf{L})(\text{NO}_3)_3$, Am 1:1 , (Tables S25-S28).....	S102
$[\text{Eu}(\mathbf{L})_2(\text{H}_2\text{O})_2][3(\text{NO}_3)]$, (Tables S29-S32).....	S114
$[\text{Nd}(\mathbf{L})_2(\text{H}_2\text{O})_2][3(\text{NO}_3)]$, (Tables S33-S36).....	S127
References and Definitions.....	S139

Sample configuration for X-ray analysis

Due to the tendency of the reported crystalline species to desolvate, all crystals were mounted in quartz capillaries (Hampton Research) with a small amount of their mother liquor, and the capillaries were sealed with sealing wax and superglue. In the cases with americium, the capillaries were coated with clear nail polish before loading, and the sealed capillaries were encased in polyester heat-shrink tubing, once they were mounted in their respective pins, to create a completely nondispersible configuration (Figure S1).

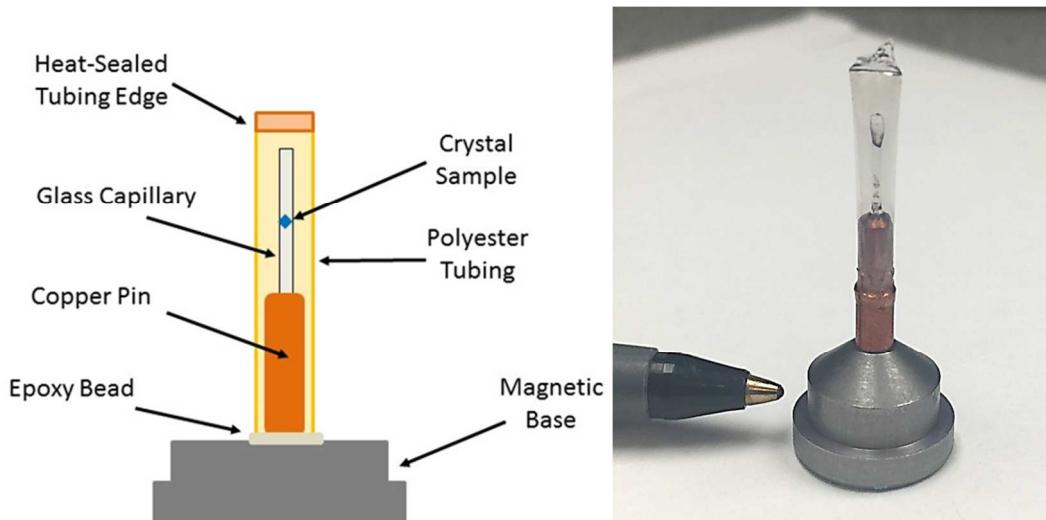


Figure S1. Left) Schematic of the sample configuration for single-crystal X-ray diffraction analysis of air-sensitive, radiological materials. Right) Image of air-sensitive sample mounted in sealed capillary and encapsulated in heat-shrink tubing.

Experimental details and structure of $[\text{Am}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, Am 2:1, at 84 K

A batch of crystals a few months old identified by X-ray crystallography at room temperature as $[\text{Am}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Am 2:1**, and prepared according to the procedure described in the manuscript was recrystallized to produce pale yellow crystals over the course of

several weeks. The recrystallized batch of crystals was analyzed at 84 K using single-crystal X-ray diffraction. After mounting a relatively large crystal plate ($0.632 \times 0.619 \times 0.060$ mm) on the diffractometer, it became evident that a smaller crystal had grown out of the center, Figure S2. The host crystal was large enough that the X-ray beam from our microfocus Ag source could be focused in an area that would prevent perturbation of the data potentially caused by the twin. The resulting structure solution was of high enough quality only to verify connectivity of the expected species. A satisfactory model could not be achieved for the outer-sphere nitrate counter anions. However, a disorder model was included for the inner-sphere nitrate ligand that seems to fit reasonably well with a 60:40 occupancy of a bidentate:monodentate binding mode shown in Figure S3. This lability was not observed for any other species reported here.

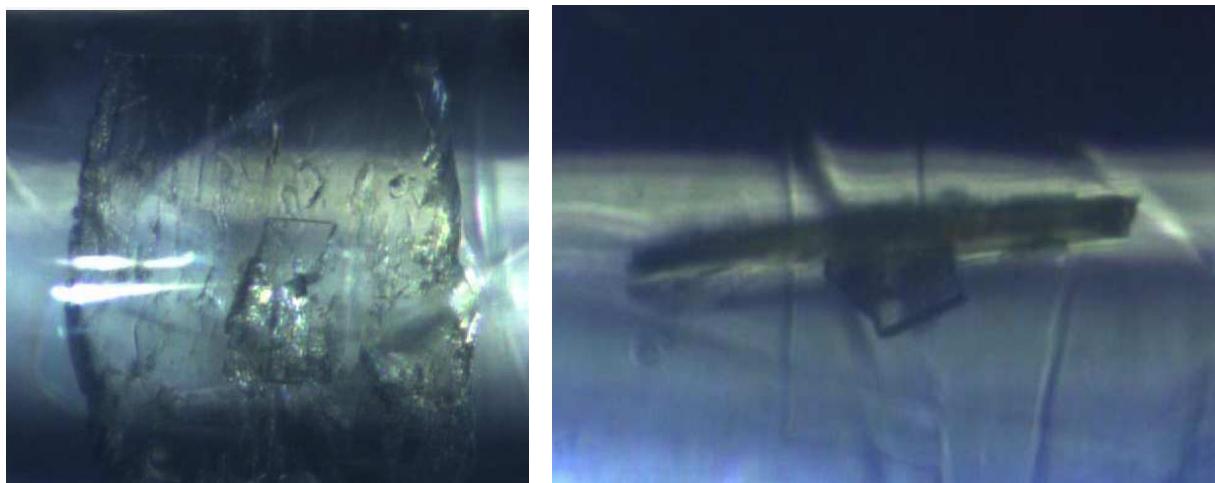


Figure S2. Top view (left) and side view (right) of a large ($0.632 \times 0.619 \times 0.060$ mm) crystal of **Am 2:1** identified to have grown a twin from its center.

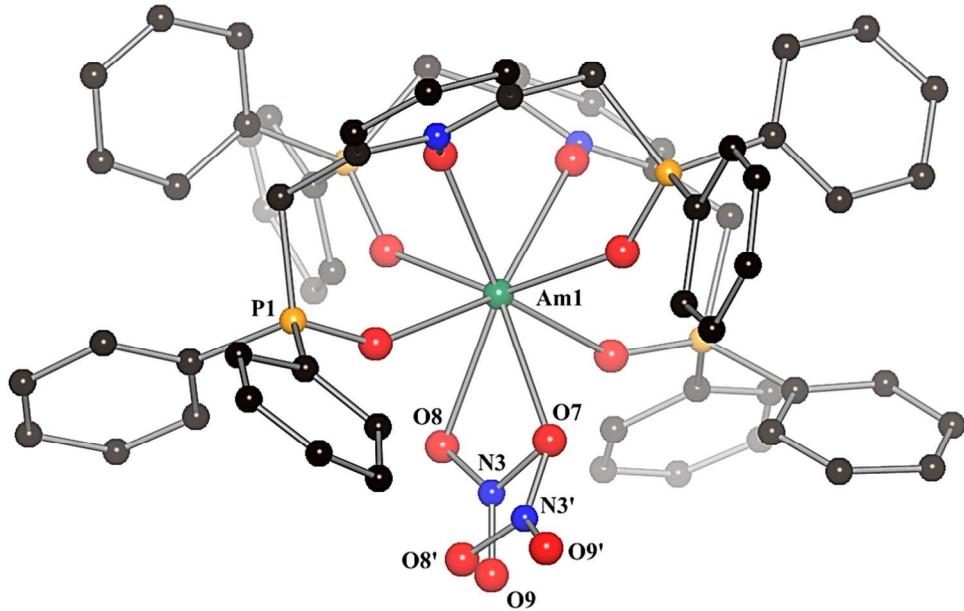


Figure S3. Crystal structure of the cation in $[Am(L)_2(No_3)][2(No_3)]$, **Am 2:1**, showing modeled disorder in the bound nitrate ligand. Black, red, blue, orange and green atoms are carbon, oxygen, nitrogen, phosphorous and americium, respectively. Hydrogen atoms have been omitted for clarity.

Experimental details and structure of $[Nd(L)_2(No_3)][Nd(No_3)_5]$

Following the literature procedure for $Yb(L)(No_3)_3$,⁹ $Nd(No_3)_3 \cdot 6(H_2O)$ (438 mg, 1 mmol) was dissolved in EtOH (25 mL), and a white slurry of 2,6-[$Ph_2P(O)CH_2$]₂C₅H₃-NO, L, (523 mg L, 1 mmol) in a 4:1 EtOH:CHCl₃ mixture (10 mL) was added to form a pale blue precipitate. The suspension was left to stir overnight. Additional EtOH (100 mL), CHCl₃ (20 mL) and water (10 mL) were added until most pale blue solids had dissolved. The remaining reaction suspension was passed through a syringe filter to produce a pale blue solution. Slow evaporation over 3 weeks produced pale blue X-ray quality crystals of Nd(L)(No₃)₃, **Nd 1:1**, and [Nd(L)₂(No₃)][Nd(No₃)₅] (Figure S4).

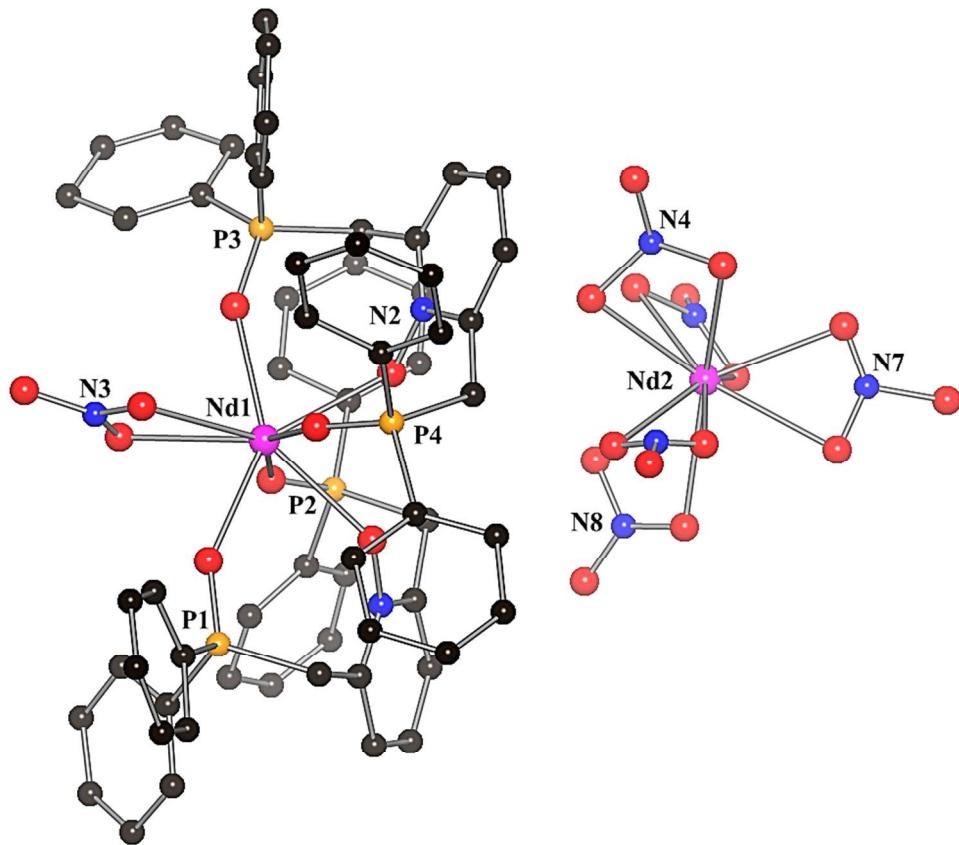


Figure S4. Crystal structure of $[\text{Nd}(\text{L})_2(\text{NO}_3)] [\text{Nd}(\text{NO}_3)_5]$ which cocrystallized with $\text{Nd}(\text{L})(\text{NO}_3)_3$, **Nd 1:1**. Black, red, blue, orange and magenta atoms are carbon, oxygen, nitrogen, phosphorous and neodymium, respectively. Hydrogen atoms and cocrystallized solvent molecules have been omitted for clarity. Disorder in the nitrate ligands is not shown.

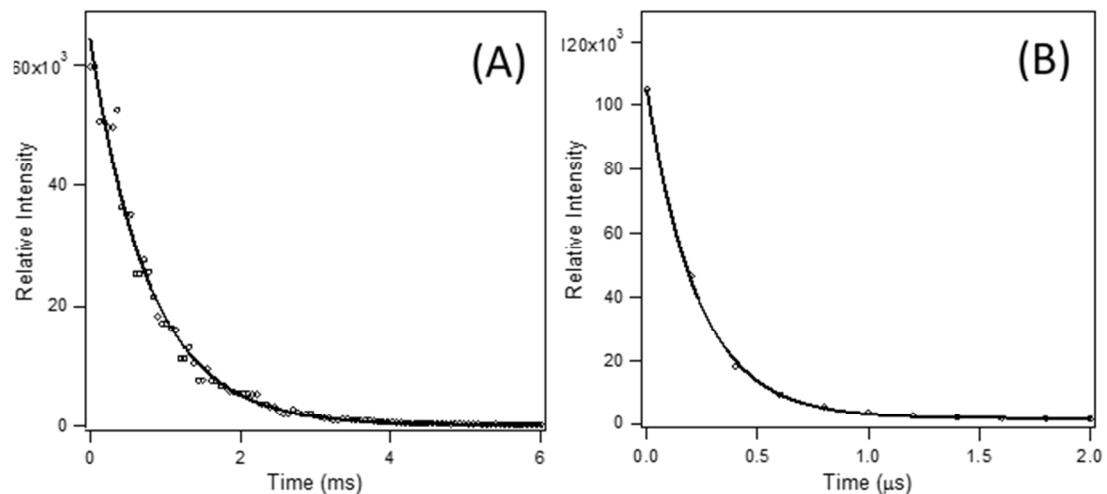


Figure S5. Luminescence decay curves and fitting with a single exponential function for **Eu 2:1** (A) and **Am 2:1** (B) at room temperature. The excitation wavelengths are 250 nm (Eu) and 503 nm (Am), respectively.

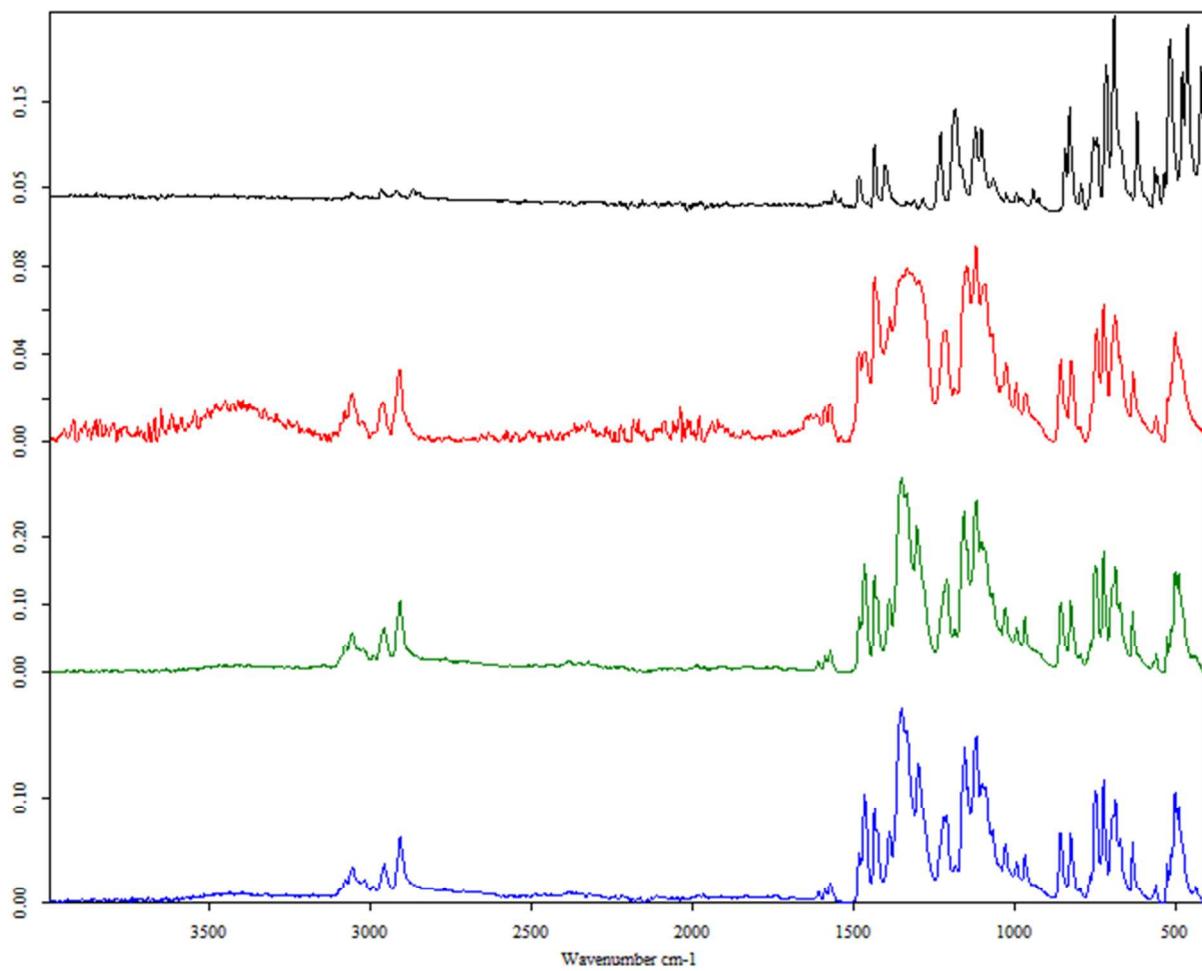


Figure S6. Attenuated total reflectance Fourier transform infrared (ATR) spectra collected on dried crystals of **Am 2:1** (red trace, second from top), **Eu 2:1** (green trace, third from top) and **Nd 2:1** (blue trace, bottom) as well as the ligand starting material 2,6-[Ph₂P(O)CH₂]₂C₅H₃-NO, **L** (black trace, top) as a powder.

X-ray Data Collection, Structure Solution and Refinement for $[\text{Eu}(\text{L})_2(\text{NO}_3)_2][\text{L}_2(\text{NO}_3)_2]$, Eu 2:1.

A colorless crystal of approximate dimensions $0.332 \times 0.245 \times 0.086$ mm was mounted in a quartz capillary tube containing a small amount of its original mother liquor which was sealed using sealing wax to provide a solvent saturated environment. The crystal-containing capillary was then secured with mounting putty in a MiTeGen Reusable BaseTM before being transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at room temperature (40 sec/frame scan time for a sphere of diffraction data) using AgK α radiation. The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The centrosymmetric triclinic space group $P\bar{1}$ was assigned and later determined to be correct.

The structure was solved by direct methods and refined on F^2 by full-matrix least-squares techniques. There was one disordered chloroform molecule of solvation present included using multiple components with partial site-occupancy-factors. Additionally, C(8), C(9), O(9), O(10), O(11), O(13), and O(14) were disordered and included using multiple components with partial site-occupancy-factors. Hydrogen atoms were included using a riding model except in the case of solvent. There were several high residuals present in the final difference-Fourier map. It was not possible to determine the nature of the residuals, although it was probable that another chloroform solvent molecule was present. The SQUEEZE routine in the PLATON⁵ program package was used to account for the electrons in the solvent accessible voids.

Table S1. Crystal data and structure refinement for [Eu(L)₂(NO₃)][2(NO₃)], **Eu 2:1**.

Empirical formula	[C ₆₂ H ₅₄ EuN ₂ O ₆ P ₄ (NO ₃)][2(NO ₃)]•CHCl ₃		
Formula weight	1504.31		
Temperature	293(2) K		
Wavelength	0.56086 Å		
Crystal system	Triclinic		
Space group	P-1		
Unit cell dimensions	a = 13.5311(14) Å	α = 84.116(3)°.	
	b = 15.7968(15) Å	β = 77.263(3)°.	
	c = 19.790(2) Å	γ = 80.558(3)°.	
Volume	4060.4(7) Å ³		
Z	2		
Density (calculated)	1.230 mg/m ³		
Absorption coefficient	0.535 mm ⁻¹		
F(000)	1524		
Crystal size	0.332 x 0.245 x 0.086 mm ³		
Theta range for data collection	2.176 to 20.520°.		
Index ranges	-16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -24 ≤ l ≤ 24		
Reflections collected	106544		
Independent reflections	16570 [R(int) = 0.0862]		
Completeness to theta = 19.665°	99.8 %		
Absorption correction	Numerical		
Max. and min. transmission	0.9792 and 0.7726		
Refinement method	Full-matrix least-squares on F ²		
Data / restraints / parameters	16570 / 0 / 713		
Goodness-of-fit on F ²	1.080		
Final R indices [I>2sigma(I)]	R1 = 0.0674, wR2 = 0.1779		
R indices (all data)	R1 = 0.0913, wR2 = 0.2003		
Largest diff. peak and hole	1.301 and -0.902 e.Å ⁻³		

Table S2. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Eu}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Eu 2:1**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Eu(1)	5152(1)	7230(1)	7360(1)	38(1)
P(1)	2753(1)	8562(1)	8026(1)	44(1)
P(2)	6265(1)	6744(1)	8932(1)	49(1)
P(3)	7411(1)	8124(1)	6518(1)	46(1)
P(4)	4004(1)	7052(1)	5864(1)	42(1)
N(1)	4913(4)	8654(3)	8629(3)	46(1)
N(2)	5212(3)	8730(3)	5963(2)	38(1)
N(3)	5257(6)	5364(4)	7459(3)	65(2)
O(1)	5318(3)	8422(3)	7989(2)	46(1)
O(2)	3451(3)	7736(3)	7848(2)	48(1)
O(3)	5864(4)	6636(3)	8312(2)	57(1)
O(4)	4804(3)	8551(3)	6626(2)	41(1)
O(5)	6772(3)	7413(3)	6711(2)	46(1)
O(6)	4604(4)	6816(3)	6421(2)	50(1)
O(7)	6011(4)	5725(3)	7123(3)	64(1)
O(8)	4473(4)	5849(3)	7751(3)	64(1)
O(9)	5539(10)	4570(8)	7468(6)	67(3)
O(9A)	5051(10)	4616(8)	7511(6)	69(3)
C(1)	3982(5)	9152(5)	8756(4)	54(2)
C(2)	3581(6)	9417(6)	9407(5)	77(2)
C(3)	4115(7)	9197(7)	9928(5)	87(3)
C(4)	5041(7)	8715(6)	9787(4)	73(2)
C(5)	5457(5)	8413(4)	9138(3)	49(1)
C(6)	3451(5)	9403(4)	8164(4)	53(2)
C(7)	1820(5)	8362(5)	8807(4)	61(2)
C(8)	1822(13)	7451(11)	9048(9)	61(4)
C(9)	1067(15)	7260(14)	9637(11)	79(5)
C(8A)	1941(17)	7690(15)	9236(11)	82(6)
C(9A)	1220(20)	7528(19)	9865(15)	113(8)
C(10)	390(11)	7995(9)	9938(7)	123(4)

C(11)	311(9)	8795(8)	9653(6)	98(3)
C(12)	1022(7)	8985(6)	9059(5)	76(2)
C(13)	2056(5)	8991(4)	7363(3)	49(1)
C(14)	2050(7)	9824(5)	7072(5)	76(2)
C(15)	1484(9)	10110(7)	6558(6)	97(3)
C(16)	946(7)	9564(7)	6342(5)	86(3)
C(17)	956(6)	8758(7)	6626(5)	81(2)
C(18)	1508(5)	8456(5)	7131(4)	63(2)
C(19)	6455(5)	7846(5)	8968(4)	56(2)
C(20)	7484(6)	6097(5)	8924(4)	65(2)
C(21)	7821(8)	5497(6)	8434(5)	83(2)
C(22)	8774(11)	4937(10)	8425(8)	129(4)
C(23)	9275(12)	5016(10)	8882(8)	133(5)
C(24)	9023(11)	5632(9)	9367(7)	124(4)
C(25)	8069(8)	6184(7)	9400(5)	89(3)
C(26)	5366(6)	6469(5)	9710(3)	58(2)
C(27)	4502(6)	6159(6)	9646(5)	72(2)
C(28)	3801(8)	5936(7)	10227(6)	95(3)
C(29)	3988(10)	6020(8)	10866(6)	107(4)
C(30)	4847(10)	6297(8)	10949(5)	112(4)
C(31)	5548(8)	6543(7)	10356(4)	86(3)
C(32)	6110(4)	9073(4)	5798(3)	42(1)
C(33)	6509(5)	9297(4)	5112(3)	55(2)
C(34)	6028(6)	9156(5)	4603(4)	61(2)
C(35)	5145(5)	8798(4)	4775(3)	53(2)
C(36)	4723(4)	8589(4)	5462(3)	41(1)
C(37)	6627(5)	9145(4)	6375(4)	50(2)
C(38)	8117(5)	8192(5)	7166(4)	58(2)
C(39)	8129(8)	8951(7)	7458(5)	85(3)
C(40)	8679(10)	8909(8)	8015(6)	109(4)
C(41)	9184(9)	8114(7)	8197(6)	96(3)
C(42)	9177(8)	7411(7)	7930(5)	89(3)
C(43)	8639(6)	7421(6)	7406(4)	70(2)
C(44)	8307(6)	7935(5)	5714(4)	57(2)
C(45)	8261(8)	7279(7)	5364(6)	93(3)
C(46)	8977(12)	7156(10)	4688(8)	137(5)

C(47)	9630(10)	7764(9)	4505(7)	115(4)
C(48)	9688(9)	8379(7)	4823(6)	97(3)
C(50)	3747(5)	8212(4)	5682(3)	44(1)
C(49)	9032(5)	8498(7)	5461(5)	83(3)
C(51)	4723(5)	6591(4)	5086(3)	51(2)
C(52)	5597(6)	6014(6)	5117(5)	73(2)
C(53)	6156(8)	5633(7)	4517(6)	104(4)
C(54)	5845(10)	5801(8)	3914(7)	104(3)
C(55)	4970(11)	6359(7)	3874(5)	105(4)
C(56)	4389(7)	6770(6)	4464(4)	75(2)
C(57)	2782(5)	6681(5)	6103(4)	54(2)
C(58)	2579(6)	6175(5)	6724(4)	70(2)
C(59)	1668(8)	5854(7)	6911(6)	101(3)
C(60)	938(8)	6050(8)	6513(8)	113(4)
C(61)	1137(7)	6533(7)	5913(6)	97(3)
C(62)	2053(6)	6870(6)	5702(5)	73(2)
N(4)	2398(10)	11573(9)	9489(7)	124(4)
O(10)	2940(40)	12030(30)	9210(20)	250(20)
O(11)	2192(18)	11341(15)	8904(12)	127(8)
O(10A)	1978(12)	11133(10)	9117(9)	84(4)
O(11A)	2610(20)	12249(17)	9481(13)	138(8)
O(12)	2370(9)	11214(8)	10040(7)	157(4)
N(5)	12236(7)	8914(6)	4161(5)	87(2)
O(13)	12274(18)	8140(16)	4071(12)	163(8)
O(14)	12560(20)	9120(18)	4544(15)	162(9)
O(13A)	13114(17)	8736(13)	4100(11)	133(6)
O(14A)	11910(20)	9220(18)	4763(15)	175(10)
O(15)	11739(8)	9362(7)	3764(5)	135(3)
C(63)	-2070(17)	4478(15)	6365(12)	96(6)
Cl(1)	-1090(16)	4414(14)	6802(11)	161(6)
Cl(2)	-1761(19)	4914(17)	5535(13)	186(8)
Cl(3)	-2211(11)	3425(9)	6312(7)	113(4)
Cl(1A)	5594(15)	12293(12)	7833(10)	151(6)
Cl(2A)	4914(10)	11034(8)	7050(6)	94(3)
Cl(3A)	5608(13)	10602(10)	8199(8)	116(4)
C(63A)	5570(20)	11365(19)	7534(15)	123(8)

Cl(1B)	4680(40)	11890(30)	8000(20)	340(20)
Cl(2B)	4487(13)	11299(11)	7181(9)	124(5)
Cl(3B)	6090(9)	10429(7)	8097(5)	80(2)
Cl(1C)	-1153(16)	5014(14)	6397(11)	168(7)
Cl(2C)	-2202(18)	4413(15)	5630(12)	170(7)
Cl(3C)	-2037(19)	3631(16)	6841(13)	194(8)

Table S3. Bond lengths [Å] and angles [°] for $[\text{Eu}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Eu 2:1**.

Eu(1)-O(5)	2.330(4)
Eu(1)-O(6)	2.333(4)
Eu(1)-O(2)	2.333(4)
Eu(1)-O(3)	2.345(4)
Eu(1)-O(1)	2.427(4)
Eu(1)-O(4)	2.458(4)
Eu(1)-O(8)	2.489(5)
Eu(1)-O(7)	2.510(5)
Eu(1)-N(3)	2.916(6)
Eu(1)-P(3)	3.5783(16)
P(1)-O(2)	1.504(4)
P(1)-C(13)	1.797(7)
P(1)-C(7)	1.804(7)
P(1)-C(6)	1.824(7)
P(2)-O(3)	1.481(5)
P(2)-C(20)	1.790(7)
P(2)-C(26)	1.803(7)
P(2)-C(19)	1.813(8)
P(3)-O(5)	1.497(4)
P(3)-C(38)	1.779(7)
P(3)-C(44)	1.794(7)
P(3)-C(37)	1.813(7)
P(4)-O(6)	1.491(4)

P(4)-C(51)	1.784(7)
P(4)-C(57)	1.794(7)
P(4)-C(50)	1.822(6)
N(1)-O(1)	1.328(6)
N(1)-C(1)	1.358(8)
N(1)-C(5)	1.363(8)
N(2)-O(4)	1.328(6)
N(2)-C(36)	1.360(7)
N(2)-C(32)	1.372(7)
N(3)-O(9A)	1.246(13)
N(3)-O(9)	1.249(13)
N(3)-O(8)	1.266(9)
N(3)-O(7)	1.274(9)
C(1)-C(2)	1.363(10)
C(1)-C(6)	1.492(10)
C(2)-C(3)	1.371(13)
C(3)-C(4)	1.344(12)
C(4)-C(5)	1.383(10)
C(5)-C(19)	1.484(10)
C(7)-C(8A)	1.30(2)
C(7)-C(12)	1.378(11)
C(7)-C(8)	1.469(19)
C(8)-C(9)	1.41(3)
C(9)-C(10)	1.45(2)
C(8A)-C(9A)	1.43(3)
C(9A)-C(10)	1.22(3)
C(10)-C(11)	1.328(16)
C(11)-C(12)	1.386(14)
C(13)-C(14)	1.380(10)
C(13)-C(18)	1.382(10)
C(14)-C(15)	1.402(12)
C(15)-C(16)	1.369(14)
C(16)-C(17)	1.337(13)
C(17)-C(18)	1.378(11)
C(20)-C(21)	1.378(12)
C(20)-C(25)	1.386(12)

C(21)-C(22)	1.436(17)
C(22)-C(23)	1.270(18)
C(23)-C(24)	1.387(19)
C(24)-C(25)	1.427(16)
C(26)-C(31)	1.372(11)
C(26)-C(27)	1.374(11)
C(27)-C(28)	1.374(12)
C(28)-C(29)	1.368(16)
C(29)-C(30)	1.355(17)
C(30)-C(31)	1.403(12)
C(32)-C(33)	1.379(9)
C(32)-C(37)	1.488(9)
C(33)-C(34)	1.367(10)
C(34)-C(35)	1.366(10)
C(35)-C(36)	1.383(9)
C(36)-C(50)	1.497(8)
C(38)-C(39)	1.388(12)
C(38)-C(43)	1.398(11)
C(39)-C(40)	1.451(15)
C(40)-C(41)	1.381(15)
C(41)-C(42)	1.280(14)
C(42)-C(43)	1.391(13)
C(44)-C(45)	1.322(12)
C(44)-C(49)	1.407(11)
C(45)-C(46)	1.478(18)
C(46)-C(47)	1.378(18)
C(47)-C(48)	1.232(15)
C(48)-C(49)	1.385(13)
C(51)-C(52)	1.378(10)
C(51)-C(56)	1.391(10)
C(52)-C(53)	1.395(13)
C(53)-C(54)	1.339(15)
C(54)-C(55)	1.367(16)
C(55)-C(56)	1.408(13)
C(57)-C(62)	1.374(10)
C(57)-C(58)	1.395(10)

C(58)-C(59)	1.371(12)
C(59)-C(60)	1.373(16)
C(60)-C(61)	1.342(16)
C(61)-C(62)	1.393(13)
N(4)-O(10)	1.13(5)
N(4)-O(11A)	1.15(3)
N(4)-O(12)	1.174(14)
N(4)-O(10A)	1.329(19)
N(4)-O(11)	1.35(2)
N(5)-O(14)	1.06(3)
N(5)-O(13A)	1.16(2)
N(5)-O(15)	1.239(11)
N(5)-O(13)	1.24(2)
N(5)-O(14A)	1.29(3)
C(63)-Cl(2C)	1.52(3)
C(63)-Cl(3C)	1.56(3)
C(63)-Cl(1C)	1.63(3)
C(63)-Cl(2)	1.71(3)
C(63)-Cl(1)	1.72(3)
C(63)-Cl(3)	1.72(3)
Cl(1A)-C(63A)	1.64(3)
Cl(2A)-C(63A)	1.62(3)
Cl(3A)-C(63A)	1.70(3)
C(63A)-Cl(1B)	1.53(5)
C(63A)-Cl(2B)	1.78(3)
C(63A)-Cl(3B)	1.89(3)
Cl(1B)-Cl(2B)	2.04(5)

O(5)-Eu(1)-O(6)	93.77(16)
O(5)-Eu(1)-O(2)	152.78(15)
O(6)-Eu(1)-O(2)	88.87(16)
O(5)-Eu(1)-O(3)	90.54(16)
O(6)-Eu(1)-O(3)	140.80(16)
O(2)-Eu(1)-O(3)	104.37(17)
O(5)-Eu(1)-O(1)	85.38(14)
O(6)-Eu(1)-O(1)	146.13(14)

O(2)-Eu(1)-O(1)	77.64(15)
O(3)-Eu(1)-O(1)	73.04(15)
O(5)-Eu(1)-O(4)	77.10(13)
O(6)-Eu(1)-O(4)	73.70(14)
O(2)-Eu(1)-O(4)	77.69(14)
O(3)-Eu(1)-O(4)	144.73(15)
O(1)-Eu(1)-O(4)	73.12(14)
O(5)-Eu(1)-O(8)	127.42(17)
O(6)-Eu(1)-O(8)	73.56(16)
O(2)-Eu(1)-O(8)	79.26(18)
O(3)-Eu(1)-O(8)	73.02(17)
O(1)-Eu(1)-O(8)	132.16(16)
O(4)-Eu(1)-O(8)	139.97(15)
O(5)-Eu(1)-O(7)	76.10(17)
O(6)-Eu(1)-O(7)	71.98(16)
O(2)-Eu(1)-O(7)	130.01(17)
O(3)-Eu(1)-O(7)	71.36(17)
O(1)-Eu(1)-O(7)	139.38(16)
O(4)-Eu(1)-O(7)	134.35(15)
O(8)-Eu(1)-O(7)	51.35(19)
O(5)-Eu(1)-N(3)	101.9(2)
O(6)-Eu(1)-N(3)	70.64(15)
O(2)-Eu(1)-N(3)	104.5(2)
O(3)-Eu(1)-N(3)	70.35(16)
O(1)-Eu(1)-N(3)	142.67(14)
O(4)-Eu(1)-N(3)	144.19(14)
O(8)-Eu(1)-N(3)	25.56(19)
O(7)-Eu(1)-N(3)	25.79(19)
O(5)-Eu(1)-P(3)	16.45(11)
O(6)-Eu(1)-P(3)	101.99(12)
O(2)-Eu(1)-P(3)	137.43(11)
O(3)-Eu(1)-P(3)	92.82(13)
O(1)-Eu(1)-P(3)	70.57(10)
O(4)-Eu(1)-P(3)	66.61(9)
O(8)-Eu(1)-P(3)	143.31(14)
O(7)-Eu(1)-P(3)	92.24(14)

N(3)-Eu(1)-P(3)	117.94(17)
O(2)-P(1)-C(13)	112.9(3)
O(2)-P(1)-C(7)	108.9(3)
C(13)-P(1)-C(7)	107.0(3)
O(2)-P(1)-C(6)	112.2(3)
C(13)-P(1)-C(6)	106.6(3)
C(7)-P(1)-C(6)	108.9(4)
O(3)-P(2)-C(20)	111.5(4)
O(3)-P(2)-C(26)	110.0(3)
C(20)-P(2)-C(26)	109.8(3)
O(3)-P(2)-C(19)	111.5(3)
C(20)-P(2)-C(19)	106.4(4)
C(26)-P(2)-C(19)	107.5(3)
O(5)-P(3)-C(38)	111.0(3)
O(5)-P(3)-C(44)	110.1(3)
C(38)-P(3)-C(44)	108.0(3)
O(5)-P(3)-C(37)	111.1(3)
C(38)-P(3)-C(37)	110.4(3)
C(44)-P(3)-C(37)	106.1(3)
O(5)-P(3)-Eu(1)	26.14(16)
C(38)-P(3)-Eu(1)	107.3(2)
C(44)-P(3)-Eu(1)	132.9(2)
C(37)-P(3)-Eu(1)	89.6(2)
O(6)-P(4)-C(51)	108.8(3)
O(6)-P(4)-C(57)	111.5(3)
C(51)-P(4)-C(57)	109.8(3)
O(6)-P(4)-C(50)	112.2(3)
C(51)-P(4)-C(50)	108.0(3)
C(57)-P(4)-C(50)	106.4(3)
O(1)-N(1)-C(1)	119.2(5)
O(1)-N(1)-C(5)	119.2(5)
C(1)-N(1)-C(5)	121.6(6)
O(4)-N(2)-C(36)	119.6(5)
O(4)-N(2)-C(32)	119.0(5)
C(36)-N(2)-C(32)	121.3(5)
O(9A)-N(3)-O(8)	108.5(9)

O(9)-N(3)-O(8)	135.0(9)
O(9A)-N(3)-O(7)	133.8(9)
O(9)-N(3)-O(7)	107.5(9)
O(8)-N(3)-O(7)	117.0(5)
O(9A)-N(3)-Eu(1)	164.6(8)
O(9)-N(3)-Eu(1)	165.6(8)
O(8)-N(3)-Eu(1)	58.0(3)
O(7)-N(3)-Eu(1)	59.0(3)
N(1)-O(1)-Eu(1)	132.9(3)
P(1)-O(2)-Eu(1)	140.7(3)
P(2)-O(3)-Eu(1)	150.3(3)
N(2)-O(4)-Eu(1)	128.7(3)
P(3)-O(5)-Eu(1)	137.4(3)
P(4)-O(6)-Eu(1)	148.4(3)
N(3)-O(7)-Eu(1)	95.2(4)
N(3)-O(8)-Eu(1)	96.4(4)
N(1)-C(1)-C(2)	119.3(7)
N(1)-C(1)-C(6)	118.0(6)
C(2)-C(1)-C(6)	122.7(7)
C(1)-C(2)-C(3)	120.6(8)
C(4)-C(3)-C(2)	119.0(8)
C(3)-C(4)-C(5)	121.9(8)
N(1)-C(5)-C(4)	117.6(7)
N(1)-C(5)-C(19)	118.6(5)
C(4)-C(5)-C(19)	123.8(6)
C(1)-C(6)-P(1)	111.6(5)
C(8A)-C(7)-C(12)	114.2(12)
C(12)-C(7)-C(8)	121.7(9)
C(8A)-C(7)-P(1)	123.0(11)
C(12)-C(7)-P(1)	121.8(7)
C(8)-C(7)-P(1)	114.8(8)
C(9)-C(8)-C(7)	116.8(15)
C(8)-C(9)-C(10)	115.6(17)
C(7)-C(8A)-C(9A)	124(2)
C(10)-C(9A)-C(8A)	116(3)
C(9A)-C(10)-C(11)	121(2)

C(11)-C(10)-C(9)	126.0(15)
C(10)-C(11)-C(12)	118.2(12)
C(7)-C(12)-C(11)	119.7(9)
C(14)-C(13)-C(18)	118.8(7)
C(14)-C(13)-P(1)	123.1(6)
C(18)-C(13)-P(1)	118.1(5)
C(13)-C(14)-C(15)	119.6(8)
C(16)-C(15)-C(14)	120.1(9)
C(17)-C(16)-C(15)	120.0(8)
C(16)-C(17)-C(18)	121.3(9)
C(17)-C(18)-C(13)	120.2(8)
C(5)-C(19)-P(2)	110.8(5)
C(21)-C(20)-C(25)	120.7(8)
C(21)-C(20)-P(2)	117.8(7)
C(25)-C(20)-P(2)	121.5(7)
C(20)-C(21)-C(22)	120.0(10)
C(23)-C(22)-C(21)	117.7(15)
C(22)-C(23)-C(24)	126.1(16)
C(23)-C(24)-C(25)	117.3(14)
C(20)-C(25)-C(24)	118.0(10)
C(31)-C(26)-C(27)	120.3(7)
C(31)-C(26)-P(2)	121.1(6)
C(27)-C(26)-P(2)	118.6(6)
C(28)-C(27)-C(26)	120.4(9)
C(29)-C(28)-C(27)	118.7(10)
C(30)-C(29)-C(28)	122.5(9)
C(29)-C(30)-C(31)	118.6(10)
C(26)-C(31)-C(30)	119.5(10)
N(2)-C(32)-C(33)	119.1(6)
N(2)-C(32)-C(37)	117.4(5)
C(33)-C(32)-C(37)	123.4(6)
C(34)-C(33)-C(32)	120.2(7)
C(35)-C(34)-C(33)	120.0(6)
C(34)-C(35)-C(36)	120.4(6)
N(2)-C(36)-C(35)	118.9(6)
N(2)-C(36)-C(50)	118.3(5)

C(35)-C(36)-C(50)	122.8(6)
C(32)-C(37)-P(3)	108.2(5)
C(39)-C(38)-C(43)	119.7(8)
C(39)-C(38)-P(3)	123.4(7)
C(43)-C(38)-P(3)	116.8(6)
C(38)-C(39)-C(40)	118.0(10)
C(41)-C(40)-C(39)	117.1(11)
C(42)-C(41)-C(40)	125.3(12)
C(41)-C(42)-C(43)	119.5(11)
C(42)-C(43)-C(38)	120.5(8)
C(45)-C(44)-C(49)	121.2(9)
C(45)-C(44)-P(3)	119.3(7)
C(49)-C(44)-P(3)	119.5(7)
C(44)-C(45)-C(46)	118.5(11)
C(47)-C(46)-C(45)	113.6(14)
C(48)-C(47)-C(46)	129.1(15)
C(47)-C(48)-C(49)	118.1(12)
C(36)-C(50)-P(4)	111.0(4)
C(48)-C(49)-C(44)	119.5(11)
C(52)-C(51)-C(56)	119.9(7)
C(52)-C(51)-P(4)	118.8(6)
C(56)-C(51)-P(4)	121.3(6)
C(51)-C(52)-C(53)	119.6(9)
C(54)-C(53)-C(52)	121.1(10)
C(53)-C(54)-C(55)	120.3(11)
C(54)-C(55)-C(56)	120.6(10)
C(51)-C(56)-C(55)	118.5(9)
C(62)-C(57)-C(58)	119.5(7)
C(62)-C(57)-P(4)	123.0(6)
C(58)-C(57)-P(4)	117.5(6)
C(59)-C(58)-C(57)	119.2(9)
C(58)-C(59)-C(60)	121.1(10)
C(61)-C(60)-C(59)	119.5(9)
C(60)-C(61)-C(62)	121.2(9)
C(57)-C(62)-C(61)	119.4(9)
O(10)-N(4)-O(12)	128(3)

O(11A)-N(4)-O(12)	110.9(19)
O(11A)-N(4)-O(10A)	138.8(19)
O(12)-N(4)-O(10A)	108.5(15)
O(10)-N(4)-O(11)	95(3)
O(12)-N(4)-O(11)	131.5(18)
O(14)-N(5)-O(15)	128.0(19)
O(13A)-N(5)-O(15)	129.2(14)
O(14)-N(5)-O(13)	122(2)
O(15)-N(5)-O(13)	109.6(15)
O(13A)-N(5)-O(14A)	105.7(18)
O(15)-N(5)-O(14A)	106.5(16)
Cl(2C)-C(63)-Cl(3C)	117(2)
Cl(2C)-C(63)-Cl(1C)	113.2(19)
Cl(3C)-C(63)-Cl(1C)	112.1(19)
Cl(2)-C(63)-Cl(1)	112.3(17)
Cl(2)-C(63)-Cl(3)	107.0(16)
Cl(1)-C(63)-Cl(3)	104.6(15)
Cl(2A)-C(63A)-Cl(1A)	135(2)
Cl(2A)-C(63A)-Cl(3A)	105.2(17)
Cl(1A)-C(63A)-Cl(3A)	107.7(18)
Cl(1B)-C(63A)-Cl(2B)	76(2)
Cl(1B)-C(63A)-Cl(3B)	107(2)
Cl(2B)-C(63A)-Cl(3B)	119.9(17)
C(63A)-Cl(1B)-Cl(2B)	58(2)
C(63A)-Cl(2B)-Cl(1B)	46.5(15)

Table S4. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Eu}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Eu 2:1**. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^*{}^2 U^{11} + \dots + 2 h k a^* b^* U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Eu(1)	33(1)	42(1)	37(1)	1(1)	-8(1)	-2(1)
P(1)	31(1)	51(1)	46(1)	-4(1)	-5(1)	-2(1)

P(2)	46(1)	61(1)	38(1)	8(1)	-13(1)	-4(1)
P(3)	33(1)	61(1)	43(1)	10(1)	-11(1)	-10(1)
P(4)	42(1)	44(1)	44(1)	-4(1)	-15(1)	-5(1)
N(1)	47(3)	50(3)	46(3)	-6(2)	-10(2)	-13(2)
N(2)	37(2)	39(2)	36(2)	5(2)	-7(2)	0(2)
N(3)	116(6)	38(3)	48(3)	6(2)	-39(4)	-7(3)
O(1)	41(2)	57(3)	39(2)	-5(2)	-6(2)	-7(2)
O(2)	33(2)	48(2)	59(3)	-2(2)	-3(2)	-2(2)
O(3)	75(3)	53(3)	47(2)	9(2)	-26(2)	-9(2)
O(4)	35(2)	48(2)	36(2)	3(2)	-4(2)	-2(2)
O(5)	30(2)	52(2)	50(2)	0(2)	-2(2)	-3(2)
O(6)	62(3)	43(2)	52(2)	-2(2)	-29(2)	-3(2)
O(7)	75(3)	51(3)	59(3)	-2(2)	-16(3)	8(2)
O(8)	74(3)	62(3)	61(3)	11(2)	-20(3)	-22(3)
C(1)	51(4)	56(4)	60(4)	-18(3)	-14(3)	-3(3)
C(2)	58(5)	92(6)	82(6)	-43(5)	-12(4)	6(4)
C(3)	80(6)	121(8)	64(5)	-40(5)	-13(4)	-4(5)
C(4)	73(5)	98(6)	54(4)	-20(4)	-19(4)	-12(5)
C(5)	53(4)	60(4)	42(3)	-9(3)	-16(3)	-18(3)
C(6)	43(3)	46(4)	68(4)	-8(3)	-12(3)	5(3)
C(7)	40(3)	90(5)	48(4)	-4(3)	-4(3)	-1(3)
C(13)	33(3)	55(4)	55(4)	-2(3)	-4(3)	2(3)
C(14)	85(6)	64(5)	86(6)	17(4)	-38(5)	-19(4)
C(15)	107(8)	82(6)	106(7)	40(6)	-51(6)	-12(6)
C(16)	57(5)	115(8)	89(6)	23(6)	-37(5)	-12(5)
C(17)	56(5)	103(7)	90(6)	-1(5)	-32(4)	-14(4)
C(18)	47(4)	71(5)	75(5)	-4(4)	-24(3)	-9(3)
C(19)	45(4)	77(5)	49(4)	4(3)	-19(3)	-11(3)
C(20)	50(4)	78(5)	57(4)	16(4)	-12(3)	3(3)
C(26)	56(4)	70(5)	42(3)	9(3)	-10(3)	-4(3)
C(27)	57(5)	82(6)	74(5)	9(4)	-14(4)	-13(4)
C(28)	73(6)	113(8)	94(7)	6(6)	-2(5)	-30(6)
C(29)	102(8)	113(9)	88(8)	4(6)	29(6)	-33(7)
C(30)	139(10)	144(10)	45(5)	17(5)	4(5)	-48(9)
C(31)	85(6)	122(8)	53(4)	9(5)	-9(4)	-34(6)
C(32)	40(3)	36(3)	48(3)	7(2)	-10(2)	-3(2)

C(33)	44(3)	58(4)	54(4)	15(3)	-4(3)	-3(3)
C(34)	64(4)	68(5)	45(4)	15(3)	-10(3)	-3(4)
C(35)	54(4)	61(4)	42(3)	6(3)	-15(3)	-1(3)
C(36)	41(3)	36(3)	45(3)	2(2)	-13(2)	2(2)
C(37)	46(3)	49(4)	58(4)	6(3)	-15(3)	-14(3)
C(38)	45(4)	81(5)	50(4)	12(3)	-11(3)	-20(3)
C(49)	36(4)	125(8)	73(5)	46(5)	-1(3)	-8(4)
C(51)	54(4)	47(4)	53(4)	-14(3)	-11(3)	-4(3)
C(52)	63(5)	76(5)	74(5)	-13(4)	-7(4)	6(4)
C(53)	81(7)	86(7)	129(10)	-32(6)	3(6)	16(5)
C(55)	159(11)	103(8)	58(5)	-16(5)	-30(6)	-22(8)
C(56)	90(6)	72(5)	63(5)	-20(4)	-20(4)	5(4)
C(57)	43(3)	61(4)	61(4)	-7(3)	-11(3)	-6(3)
C(58)	56(4)	73(5)	77(5)	8(4)	-7(4)	-9(4)
C(59)	74(6)	100(8)	116(8)	25(6)	0(6)	-24(5)
C(60)	60(6)	104(8)	172(12)	25(8)	-25(7)	-26(5)
C(61)	63(5)	109(8)	132(9)	1(7)	-47(6)	-20(5)
C(62)	55(4)	87(6)	84(6)	-1(4)	-28(4)	-14(4)

X-ray Data Collection, Structure Solution and Refinement for [Nd(L)₂(NO₃)][2(NO₃)], Nd 2:1.

A pale blue crystal of approximate dimensions 1.0 x 1.0 x 0.5 mm was mounted in a quartz capillary tube containing a small amount of its original mother liquor which was sealed using sealing wax to provide a solvent saturated environment. The crystal-containing capillary was then secured with mounting putty in a MiTeGen Reusable Base™ before being transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at room temperature (20

sec/frame scan time for a sphere of diffraction data) using MoK α radiation. The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The centrosymmetric triclinic space group $P\bar{1}$ was assigned and later determined to be correct.

The structure was solved by direct methods and refined on F^2 by full-matrix least-squares techniques. There were several disordered ethanol molecules of solvation present which were included using multiple components with partial site-occupancy-factors. Additionally, C(8), C(9), and a number of oxygen atoms pertaining to nitrate groups were disordered and included using multiple components with partial site-occupancy-factors. All hydrogen atoms except those pertaining to solvent molecules were included using a riding model. Relatively high thermal motion throughout the model prevented universal anisotropic refinement.

Table S5. Crystal data and structure refinement for [Nd(L)₂(NO₃)][2(NO₃)], **Nd 2:1**.

Empirical formula	[C ₆₂ H ₅₄ N ₂ NdO ₆ P ₄ (NO ₃)][2(NO ₃)]•4(CH ₃ CH ₂ OH)	
Formula weight	1561.49	
Temperature	293(2) K	
Wavelength	0.71073 Å	
Crystal system	Triclinic	
Space group	P-1	
Unit cell dimensions	a = 13.5331(16) Å	α = 85.189(5)°.
	b = 15.671(2) Å	β = 77.361(5)°.
	c = 19.644(3) Å	γ = 80.731(5)°.
Volume	4006.8(9) Å ³	
Z	2	
Density (calculated)	1.294 mg/m ³	
Absorption coefficient	0.794 mm ⁻¹	

F(000)	1610
Crystal size	1.0 x 1.0 x 0.5
Theta range for data collection	2.864 to 26.372°.
Index ranges	-16 ≤ h ≤ 16, -19 ≤ k ≤ 19, -24 ≤ l ≤ 24
Reflections collected	130763
Independent reflections	16355 [R(int) = 0.0511]
Completeness to theta = 25.242°	99.7 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.7457 and 0.5562
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	16355 / 0 / 738
Goodness-of-fit on F ²	1.070
Final R indices [I>2sigma(I)]	R1 = 0.0822, wR2 = 0.2063
R indices (all data)	R1 = 0.0991, wR2 = 0.2234
Largest diff. peak and hole	1.862 and -0.949 e.Å ⁻³

Table S6. Atomic coordinates (x 10⁴) and equivalent isotropic displacement parameters (Å² x 10³) for [Nd(L)₂(NO₃)][2(NO₃)], **Nd 2:1**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Nd(1)	5114(1)	7240(1)	7366(1)	42(1)
P(1)	2714(1)	8603(1)	8078(1)	46(1)
P(2)	6230(1)	6753(1)	8963(1)	49(1)
P(3)	7364(1)	8182(1)	6530(1)	48(1)
P(4)	3935(1)	7050(1)	5855(1)	42(1)
N(1)	4878(4)	8691(3)	8677(3)	45(1)
N(2)	5167(4)	8738(3)	5970(3)	38(1)
N(3)	5289(9)	5336(4)	7445(4)	80(3)
O(1)	5292(3)	8456(3)	8026(2)	45(1)
O(2)	3397(3)	7764(3)	7900(3)	51(1)
O(3)	5817(4)	6637(3)	8339(3)	59(1)

O(4)	4755(3)	8576(3)	6635(2)	40(1)
O(5)	6749(3)	7449(3)	6702(2)	52(1)
O(6)	4520(4)	6810(3)	6426(3)	52(1)
O(7)	5997(5)	5734(4)	7098(3)	74(2)
O(8)	4482(5)	5807(4)	7737(3)	71(2)
O(9)	5629(11)	4547(8)	7461(7)	70(3)
O(9A)	5071(12)	4565(9)	7537(8)	83(4)
C(1)	3956(6)	9202(5)	8798(4)	54(2)
C(2)	3548(7)	9472(6)	9472(5)	71(2)
C(3)	4071(8)	9254(7)	9989(5)	82(3)
C(4)	5003(7)	8738(6)	9854(4)	65(2)
C(5)	5416(5)	8450(5)	9186(3)	49(2)
C(6)	3422(6)	9430(5)	8211(4)	53(2)
C(7)	1789(5)	8433(6)	8871(4)	63(2)
C(8)	1830(18)	7520(16)	9129(12)	87(6)
C(9)	1060(20)	7340(20)	9749(15)	112(8)
C(8A)	2018(17)	7800(14)	9333(11)	79(5)
C(9A)	1310(20)	7680(20)	9990(17)	123(9)
C(10)	400(13)	8156(11)	10032(9)	134(5)
C(11)	257(10)	8893(9)	9713(7)	105(4)
C(12)	955(8)	9034(7)	9101(5)	80(3)
C(13)	2019(5)	9024(5)	7412(4)	52(2)
C(14)	2052(8)	9847(6)	7102(6)	84(3)
C(15)	1484(10)	10130(8)	6590(7)	111(4)
C(16)	920(8)	9566(9)	6385(7)	99(4)
C(17)	907(7)	8760(7)	6682(5)	82(3)
C(18)	1471(6)	8483(6)	7192(5)	63(2)
C(19)	6405(6)	7865(5)	9019(4)	57(2)
C(20)	7466(6)	6094(6)	8922(4)	63(2)
C(21)	7794(8)	5541(7)	8391(6)	84(3)
C(22)	8753(12)	4998(10)	8368(8)	121(4)
C(23)	9299(12)	5064(10)	8853(8)	122(4)
C(24)	9014(12)	5628(10)	9370(8)	126(5)
C(25)	8046(9)	6181(7)	9416(6)	90(3)
C(26)	5354(6)	6475(5)	9746(4)	58(2)
C(27)	4497(7)	6143(6)	9685(5)	73(2)

C(28)	3808(8)	5930(8)	10272(7)	99(3)
C(29)	3945(10)	6061(9)	10918(7)	109(4)
C(30)	4792(11)	6380(9)	10991(5)	108(4)
C(31)	5513(8)	6600(7)	10398(4)	83(3)
C(32)	6057(5)	9086(4)	5801(3)	45(1)
C(33)	6464(5)	9286(5)	5112(4)	53(2)
C(34)	5990(6)	9127(5)	4593(4)	60(2)
C(35)	5098(6)	8780(5)	4773(3)	51(2)
C(36)	4677(5)	8588(4)	5463(3)	40(1)
C(37)	6561(5)	9202(4)	6390(4)	48(2)
C(38)	8047(6)	8282(6)	7205(4)	63(2)
C(39)	8019(8)	9022(7)	7516(5)	82(3)
C(40)	8555(10)	8991(9)	8079(7)	104(4)
C(41)	9065(9)	8230(8)	8263(6)	96(3)
C(42)	9079(9)	7521(8)	7974(6)	96(3)
C(43)	8581(7)	7512(6)	7425(5)	73(2)
C(44)	8293(6)	8012(5)	5734(4)	58(2)
C(45)	8284(8)	7347(7)	5353(5)	84(3)
C(46)	9007(11)	7259(9)	4694(7)	112(4)
C(47)	9662(10)	7834(9)	4519(7)	105(4)
C(48)	9677(9)	8471(8)	4879(6)	93(3)
C(50)	3691(5)	8219(4)	5675(4)	47(1)
C(49)	8986(6)	8583(7)	5506(5)	80(3)
C(51)	4656(6)	6578(4)	5063(4)	51(2)
C(52)	5561(7)	6044(6)	5084(5)	76(2)
C(53)	6127(9)	5657(8)	4487(7)	101(4)
C(54)	5800(11)	5791(8)	3872(7)	107(4)
C(55)	4903(12)	6339(8)	3853(5)	105(4)
C(56)	4324(8)	6736(6)	4439(4)	75(2)
C(57)	2703(5)	6691(5)	6092(4)	54(2)
C(58)	2501(7)	6190(6)	6713(5)	69(2)
C(59)	1578(8)	5871(9)	6895(6)	101(4)
C(60)	854(9)	6069(8)	6501(8)	107(4)
C(61)	1042(8)	6570(8)	5895(7)	98(3)
C(62)	1985(7)	6889(6)	5686(5)	75(2)
N(4)	2387(12)	11658(11)	9330(9)	140(5)

O(10)	2750(18)	12252(17)	9192(12)	277(10)
O(11)	2112(16)	11399(14)	8844(11)	104(6)
O(12)	2290(18)	11207(16)	10038(12)	123(7)
O(11A)	1975(13)	11141(11)	9037(9)	80(4)
O(12A)	2460(20)	11535(19)	9814(17)	142(9)
N(5)	12191(7)	8830(6)	4067(5)	86(2)
O(13)	12252(11)	8041(10)	3996(8)	108(4)
O(14)	12550(16)	9029(12)	4467(10)	108(5)
O(15)	11838(12)	9221(11)	3588(9)	99(5)
O(13A)	13040(30)	8680(30)	4140(20)	187(15)
O(14A)	12060(20)	9204(19)	4659(15)	126(9)
O(15A)	11623(15)	9430(12)	3807(10)	74(5)
O(16)	4250(30)	11130(30)	7130(20)	216(16)
C(64)	5670(20)	10630(20)	8124(16)	117(8)
O(16A)	4809(18)	10984(13)	7040(11)	110(6)
C(64A)	5100(30)	11210(30)	8130(20)	160(13)
C(63)	5330(20)	11420(20)	7430(18)	242(12)
O(17)	7760(20)	11517(17)	8492(14)	169(9)
O(17A)	8470(40)	12730(40)	7670(30)	300(20)
C(65)	8420(30)	11650(30)	7640(20)	312(18)
C(66)	9570(30)	11340(20)	7760(18)	271(14)
O(18)	12080(30)	6300(20)	3740(18)	219(13)
C(67)	11940(30)	5290(30)	4190(20)	174(15)
C(68)	11430(30)	5000(30)	3550(20)	181(16)
O(18A)	2610(20)	11805(17)	7442(14)	162(8)
C(67A)	2400(30)	12550(30)	7030(20)	166(14)
C(68A)	1590(40)	13080(30)	7260(30)	192(17)
O(19)	1752(18)	4088(15)	8796(12)	280(10)
C(69)	2600(20)	4569(18)	8472(14)	201(9)
C(70)	2190(20)	5424(19)	8759(14)	211(10)

Table S7. Bond lengths [\AA] and angles [$^\circ$] for $[\text{Nd}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Nd 2:1**.

Nd(1)-O(6)	2.356(4)
Nd(1)-O(5)	2.368(4)
Nd(1)-O(2)	2.374(4)
Nd(1)-O(3)	2.377(5)
Nd(1)-O(1)	2.463(4)
Nd(1)-O(4)	2.482(4)
Nd(1)-O(7)	2.510(6)
Nd(1)-O(8)	2.525(6)
Nd(1)-N(3)	2.948(6)
P(1)-O(2)	1.500(5)
P(1)-C(13)	1.795(7)
P(1)-C(6)	1.800(7)
P(1)-C(7)	1.802(7)
P(2)-O(3)	1.490(5)
P(2)-C(26)	1.795(8)
P(2)-C(20)	1.807(8)
P(2)-C(19)	1.811(9)
P(3)-O(5)	1.499(5)
P(3)-C(44)	1.789(8)
P(3)-C(38)	1.803(8)
P(3)-C(37)	1.820(7)
P(4)-O(6)	1.500(5)
P(4)-C(51)	1.792(7)
P(4)-C(57)	1.797(8)
P(4)-C(50)	1.828(7)
N(1)-O(1)	1.335(7)
N(1)-C(1)	1.355(9)
N(1)-C(5)	1.357(9)
N(2)-O(4)	1.323(6)
N(2)-C(32)	1.366(8)
N(2)-C(36)	1.366(8)
N(3)-O(9)	1.248(14)
N(3)-O(8)	1.266(11)
N(3)-O(7)	1.267(11)

N(3)-O(9A)	1.279(16)
C(1)-C(2)	1.391(11)
C(1)-C(6)	1.480(10)
C(2)-C(3)	1.351(13)
C(3)-C(4)	1.370(13)
C(4)-C(5)	1.391(10)
C(5)-C(19)	1.485(10)
C(7)-C(8A)	1.33(2)
C(7)-C(12)	1.369(12)
C(7)-C(8)	1.47(2)
C(8)-C(9)	1.46(4)
C(9)-C(10)	1.51(3)
C(8A)-C(9A)	1.45(4)
C(9A)-C(10)	1.32(3)
C(10)-C(11)	1.273(19)
C(11)-C(12)	1.383(16)
C(13)-C(18)	1.365(11)
C(13)-C(14)	1.382(12)
C(14)-C(15)	1.397(14)
C(15)-C(16)	1.393(17)
C(16)-C(17)	1.346(15)
C(17)-C(18)	1.389(12)
C(20)-C(21)	1.364(13)
C(20)-C(25)	1.403(14)
C(21)-C(22)	1.426(17)
C(22)-C(23)	1.346(19)
C(23)-C(24)	1.354(19)
C(24)-C(25)	1.440(18)
C(26)-C(27)	1.377(12)
C(26)-C(31)	1.378(12)
C(27)-C(28)	1.369(14)
C(28)-C(29)	1.359(17)
C(29)-C(30)	1.360(18)
C(30)-C(31)	1.405(14)
C(32)-C(33)	1.377(9)
C(32)-C(37)	1.501(9)

C(33)-C(34)	1.375(11)
C(34)-C(35)	1.368(11)
C(35)-C(36)	1.378(9)
C(36)-C(50)	1.501(9)
C(38)-C(39)	1.347(13)
C(38)-C(43)	1.390(12)
C(39)-C(40)	1.442(16)
C(40)-C(41)	1.342(16)
C(41)-C(42)	1.284(15)
C(42)-C(43)	1.394(14)
C(44)-C(45)	1.336(13)
C(44)-C(49)	1.378(12)
C(45)-C(46)	1.445(16)
C(46)-C(47)	1.336(17)
C(47)-C(48)	1.276(16)
C(48)-C(49)	1.379(14)
C(51)-C(52)	1.374(11)
C(51)-C(56)	1.386(11)
C(52)-C(53)	1.381(14)
C(53)-C(54)	1.365(17)
C(54)-C(55)	1.375(18)
C(55)-C(56)	1.375(14)
C(57)-C(62)	1.371(11)
C(57)-C(58)	1.393(11)
C(58)-C(59)	1.382(13)
C(59)-C(60)	1.361(17)
C(60)-C(61)	1.369(17)
C(61)-C(62)	1.410(13)
N(4)-O(12A)	0.98(3)
N(4)-O(10)	1.11(3)
N(4)-O(11)	1.22(2)
N(4)-O(11A)	1.29(2)
N(4)-O(12)	1.50(3)
O(10)-O(12A)	1.62(4)
N(5)-O(14)	1.098(18)
N(5)-O(13A)	1.17(4)

N(5)-O(15)	1.224(17)
N(5)-O(13)	1.244(16)
N(5)-O(15A)	1.26(2)
N(5)-O(14A)	1.31(3)
O(13A)-O(14A)	1.63(5)
O(16)-C(63)	1.83(5)
C(64)-C(63)	1.84(4)
O(16A)-C(63)	1.42(3)
C(64A)-C(63)	1.37(4)
O(17)-C(65)	1.73(4)
O(17A)-C(65)	1.71(6)
C(65)-C(66)	1.62(4)
O(18)-C(67)	1.77(5)
C(67)-C(68)	1.68(6)
O(18A)-C(67A)	1.40(4)
C(67A)-C(68A)	1.29(5)
O(19)-C(69)	1.48(3)
C(69)-C(70)	1.47(3)

O(6)-Nd(1)-O(5)	95.27(18)
O(6)-Nd(1)-O(2)	89.03(17)
O(5)-Nd(1)-O(2)	151.96(16)
O(6)-Nd(1)-O(3)	140.48(16)
O(5)-Nd(1)-O(3)	91.80(18)
O(2)-Nd(1)-O(3)	102.39(19)
O(6)-Nd(1)-O(1)	146.51(15)
O(5)-Nd(1)-O(1)	84.65(16)
O(2)-Nd(1)-O(1)	76.82(15)
O(3)-Nd(1)-O(1)	72.83(16)
O(6)-Nd(1)-O(4)	73.90(15)
O(5)-Nd(1)-O(4)	76.40(15)
O(2)-Nd(1)-O(4)	78.25(15)
O(3)-Nd(1)-O(4)	145.23(16)
O(1)-Nd(1)-O(4)	73.57(14)
O(6)-Nd(1)-O(7)	72.42(18)
O(5)-Nd(1)-O(7)	76.1(2)

O(2)-Nd(1)-O(7)	131.2(2)
O(3)-Nd(1)-O(7)	71.73(19)
O(1)-Nd(1)-O(7)	138.74(18)
O(4)-Nd(1)-O(7)	133.71(17)
O(6)-Nd(1)-O(8)	72.72(17)
O(5)-Nd(1)-O(8)	126.5(2)
O(2)-Nd(1)-O(8)	81.2(2)
O(3)-Nd(1)-O(8)	71.92(18)
O(1)-Nd(1)-O(8)	132.81(17)
O(4)-Nd(1)-O(8)	140.78(17)
O(7)-Nd(1)-O(8)	50.5(2)
O(6)-Nd(1)-N(3)	71.29(17)
O(5)-Nd(1)-N(3)	101.3(3)
O(2)-Nd(1)-N(3)	106.3(3)
O(3)-Nd(1)-N(3)	69.19(18)
O(1)-Nd(1)-N(3)	141.68(16)
O(4)-Nd(1)-N(3)	144.74(16)
O(7)-Nd(1)-N(3)	25.3(2)
O(8)-Nd(1)-N(3)	25.3(2)
O(2)-P(1)-C(13)	112.9(3)
O(2)-P(1)-C(6)	112.0(3)
C(13)-P(1)-C(6)	106.9(3)
O(2)-P(1)-C(7)	109.0(4)
C(13)-P(1)-C(7)	107.5(4)
C(6)-P(1)-C(7)	108.4(4)
O(3)-P(2)-C(26)	110.1(4)
O(3)-P(2)-C(20)	110.4(4)
C(26)-P(2)-C(20)	110.3(4)
O(3)-P(2)-C(19)	111.9(3)
C(26)-P(2)-C(19)	106.9(4)
C(20)-P(2)-C(19)	107.2(4)
O(5)-P(3)-C(44)	110.6(3)
O(5)-P(3)-C(38)	111.5(3)
C(44)-P(3)-C(38)	107.4(4)
O(5)-P(3)-C(37)	111.4(3)
C(44)-P(3)-C(37)	106.0(3)

C(38)-P(3)-C(37)	109.7(4)
O(6)-P(4)-C(51)	109.5(3)
O(6)-P(4)-C(57)	110.7(3)
C(51)-P(4)-C(57)	110.3(3)
O(6)-P(4)-C(50)	112.5(3)
C(51)-P(4)-C(50)	107.4(3)
C(57)-P(4)-C(50)	106.3(3)
O(1)-N(1)-C(1)	118.6(5)
O(1)-N(1)-C(5)	119.1(6)
C(1)-N(1)-C(5)	122.1(6)
O(4)-N(2)-C(32)	119.5(5)
O(4)-N(2)-C(36)	119.8(5)
C(32)-N(2)-C(36)	120.7(5)
O(9)-N(3)-O(8)	135.7(11)
O(9)-N(3)-O(7)	107.9(11)
O(8)-N(3)-O(7)	116.0(6)
O(8)-N(3)-O(9A)	104.8(11)
O(7)-N(3)-O(9A)	138.6(11)
O(9)-N(3)-Nd(1)	163.5(10)
O(8)-N(3)-Nd(1)	58.4(3)
O(7)-N(3)-Nd(1)	57.7(3)
O(9A)-N(3)-Nd(1)	162.6(10)
N(1)-O(1)-Nd(1)	132.4(4)
P(1)-O(2)-Nd(1)	140.0(3)
P(2)-O(3)-Nd(1)	149.9(3)
N(2)-O(4)-Nd(1)	127.7(3)
P(3)-O(5)-Nd(1)	136.5(3)
P(4)-O(6)-Nd(1)	148.6(3)
N(3)-O(7)-Nd(1)	97.1(5)
N(3)-O(8)-Nd(1)	96.4(5)
N(1)-C(1)-C(2)	118.3(7)
N(1)-C(1)-C(6)	118.4(6)
C(2)-C(1)-C(6)	123.2(7)
C(3)-C(2)-C(1)	121.0(8)
C(2)-C(3)-C(4)	119.7(8)
C(3)-C(4)-C(5)	120.1(8)

N(1)-C(5)-C(4)	118.7(7)
N(1)-C(5)-C(19)	119.0(6)
C(4)-C(5)-C(19)	122.3(7)
C(1)-C(6)-P(1)	112.7(5)
C(8A)-C(7)-C(12)	116.2(12)
C(12)-C(7)-C(8)	121.3(12)
C(8A)-C(7)-P(1)	119.5(11)
C(12)-C(7)-P(1)	122.6(7)
C(8)-C(7)-P(1)	113.7(11)
C(9)-C(8)-C(7)	116(2)
C(8)-C(9)-C(10)	112(2)
C(7)-C(8A)-C(9A)	120(2)
C(10)-C(9A)-C(8A)	114(3)
C(11)-C(10)-C(9A)	123(2)
C(11)-C(10)-C(9)	127.9(19)
C(10)-C(11)-C(12)	116.6(14)
C(7)-C(12)-C(11)	121.7(10)
C(18)-C(13)-C(14)	119.5(8)
C(18)-C(13)-P(1)	117.3(6)
C(14)-C(13)-P(1)	123.1(6)
C(13)-C(14)-C(15)	119.7(9)
C(16)-C(15)-C(14)	119.2(10)
C(17)-C(16)-C(15)	120.7(10)
C(16)-C(17)-C(18)	119.9(10)
C(13)-C(18)-C(17)	120.9(9)
C(5)-C(19)-P(2)	112.2(5)
C(21)-C(20)-C(25)	123.2(9)
C(21)-C(20)-P(2)	117.0(7)
C(25)-C(20)-P(2)	119.7(7)
C(20)-C(21)-C(22)	117.4(11)
C(23)-C(22)-C(21)	119.4(15)
C(22)-C(23)-C(24)	124.7(16)
C(23)-C(24)-C(25)	117.6(15)
C(20)-C(25)-C(24)	117.5(11)
C(27)-C(26)-C(31)	120.0(8)
C(27)-C(26)-P(2)	118.4(6)

C(31)-C(26)-P(2)	121.6(7)
C(28)-C(27)-C(26)	120.0(10)
C(29)-C(28)-C(27)	120.8(11)
C(28)-C(29)-C(30)	120.2(10)
C(29)-C(30)-C(31)	120.2(11)
C(26)-C(31)-C(30)	118.8(10)
N(2)-C(32)-C(33)	119.5(6)
N(2)-C(32)-C(37)	117.1(6)
C(33)-C(32)-C(37)	123.4(6)
C(34)-C(33)-C(32)	120.6(7)
C(35)-C(34)-C(33)	119.0(7)
C(34)-C(35)-C(36)	120.8(7)
N(2)-C(36)-C(35)	119.4(6)
N(2)-C(36)-C(50)	118.7(5)
C(35)-C(36)-C(50)	121.9(6)
C(32)-C(37)-P(3)	106.8(5)
C(39)-C(38)-C(43)	120.4(8)
C(39)-C(38)-P(3)	124.6(7)
C(43)-C(38)-P(3)	114.9(7)
C(38)-C(39)-C(40)	118.2(10)
C(41)-C(40)-C(39)	118.6(12)
C(42)-C(41)-C(40)	123.3(13)
C(41)-C(42)-C(43)	120.7(12)
C(38)-C(43)-C(42)	118.8(9)
C(45)-C(44)-C(49)	120.6(9)
C(45)-C(44)-P(3)	119.4(7)
C(49)-C(44)-P(3)	119.9(7)
C(44)-C(45)-C(46)	117.8(11)
C(47)-C(46)-C(45)	117.5(13)
C(48)-C(47)-C(46)	125.2(14)
C(47)-C(48)-C(49)	118.9(13)
C(36)-C(50)-P(4)	110.8(4)
C(44)-C(49)-C(48)	120.0(11)
C(52)-C(51)-C(56)	119.7(8)
C(52)-C(51)-P(4)	118.4(6)
C(56)-C(51)-P(4)	121.9(6)

C(51)-C(52)-C(53)	120.0(10)
C(54)-C(53)-C(52)	121.0(11)
C(53)-C(54)-C(55)	118.6(10)
C(54)-C(55)-C(56)	121.7(11)
C(55)-C(56)-C(51)	119.0(10)
C(62)-C(57)-C(58)	120.5(8)
C(62)-C(57)-P(4)	122.4(6)
C(58)-C(57)-P(4)	117.1(6)
C(59)-C(58)-C(57)	118.6(9)
C(60)-C(59)-C(58)	121.6(11)
C(59)-C(60)-C(61)	120.1(11)
C(60)-C(61)-C(62)	119.7(10)
C(57)-C(62)-C(61)	119.5(9)
O(12A)-N(4)-O(10)	101(3)
O(10)-N(4)-O(11)	113(2)
O(12A)-N(4)-O(11A)	120(3)
O(10)-N(4)-O(11A)	139(2)
O(10)-N(4)-O(12)	123(2)
O(11)-N(4)-O(12)	124(2)
N(4)-O(10)-O(12A)	36.4(15)
N(4)-O(12A)-O(10)	42.2(19)
O(14)-N(5)-O(15)	133.4(15)
O(14)-N(5)-O(13)	117.6(15)
O(15)-N(5)-O(13)	108.2(13)
O(13A)-N(5)-O(15A)	138(2)
O(13A)-N(5)-O(14A)	82(2)
O(15A)-N(5)-O(14A)	94.1(17)
N(5)-O(13A)-O(14A)	53(2)
N(5)-O(14A)-O(13A)	45.0(16)
C(64A)-C(63)-O(16A)	113(3)
O(16)-C(63)-C(64)	110(2)
C(66)-C(65)-O(17A)	95(3)
C(66)-C(65)-O(17)	99(3)
C(68)-C(67)-O(18)	90(3)
C(68A)-C(67A)-O(18A)	117(4)
C(70)-C(69)-O(19)	100(2)

Table S8. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Nd}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Nd 2:1**. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^{*2} U^{11} + \dots + 2 h k a^{*} b^{*} U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Nd(1)	35(1)	49(1)	41(1)	1(1)	-8(1)	-4(1)
P(1)	32(1)	51(1)	51(1)	-4(1)	-4(1)	-4(1)
P(2)	51(1)	52(1)	44(1)	9(1)	-17(1)	-8(1)
P(3)	33(1)	66(1)	44(1)	8(1)	-7(1)	-9(1)
P(4)	43(1)	41(1)	45(1)	-3(1)	-16(1)	-4(1)
N(1)	45(3)	46(3)	45(3)	1(2)	-8(2)	-14(2)
N(2)	38(3)	31(2)	40(3)	1(2)	-4(2)	2(2)
N(3)	164(9)	30(3)	57(4)	8(3)	-56(5)	-11(4)
O(1)	45(2)	55(3)	35(2)	-2(2)	-5(2)	-9(2)
O(2)	37(2)	50(3)	62(3)	-5(2)	-1(2)	-7(2)
O(3)	79(4)	51(3)	53(3)	7(2)	-32(3)	-11(2)
O(4)	39(2)	42(2)	34(2)	1(2)	-5(2)	-1(2)
O(5)	37(2)	59(3)	53(3)	5(2)	-4(2)	-3(2)
O(6)	61(3)	43(2)	58(3)	-1(2)	-31(2)	-3(2)
O(7)	96(5)	57(3)	64(3)	-9(3)	-22(3)	15(3)
O(8)	96(5)	66(4)	62(3)	13(3)	-27(3)	-34(3)
C(1)	53(4)	52(4)	57(4)	-8(3)	-7(3)	-10(3)
C(2)	62(5)	79(6)	68(5)	-27(4)	-8(4)	1(4)
C(3)	81(6)	106(7)	57(5)	-30(5)	3(4)	-17(6)
C(4)	75(5)	77(5)	47(4)	-6(4)	-17(4)	-15(4)
C(5)	51(4)	54(4)	46(3)	1(3)	-12(3)	-21(3)
C(6)	49(4)	48(4)	62(4)	-12(3)	-13(3)	1(3)
C(7)	38(4)	85(6)	58(4)	-11(4)	5(3)	-7(3)
C(13)	33(3)	59(4)	60(4)	2(3)	-6(3)	-3(3)
C(14)	90(7)	73(6)	105(7)	12(5)	-48(6)	-27(5)
C(15)	112(9)	92(8)	139(11)	47(7)	-64(8)	-22(7)
C(16)	68(6)	124(9)	114(9)	28(7)	-49(6)	-18(6)
C(17)	60(5)	105(8)	86(6)	7(6)	-29(5)	-18(5)
C(18)	51(4)	68(5)	79(5)	-3(4)	-27(4)	-11(4)
C(19)	48(4)	80(5)	44(4)	12(3)	-14(3)	-12(3)

C(20)	54(4)	70(5)	62(5)	10(4)	-13(4)	-3(4)
C(26)	59(4)	64(4)	48(4)	11(3)	-15(3)	-5(3)
C(27)	57(5)	82(6)	81(6)	10(5)	-17(4)	-17(4)
C(28)	70(6)	109(9)	111(9)	14(7)	-4(6)	-26(6)
C(29)	98(9)	123(10)	91(8)	23(7)	15(7)	-37(8)
C(30)	123(10)	145(11)	54(5)	21(6)	-7(6)	-41(9)
C(31)	91(7)	108(8)	53(5)	22(5)	-18(4)	-35(6)
C(32)	40(3)	42(3)	50(3)	2(3)	-8(3)	-3(3)
C(33)	45(4)	59(4)	51(4)	9(3)	-4(3)	-8(3)
C(34)	59(4)	72(5)	43(4)	9(3)	-5(3)	-3(4)
C(35)	59(4)	52(4)	41(3)	-2(3)	-16(3)	5(3)
C(36)	41(3)	38(3)	42(3)	-4(2)	-14(2)	6(2)
C(37)	45(3)	52(4)	49(4)	4(3)	-9(3)	-14(3)
C(38)	49(4)	92(6)	52(4)	17(4)	-13(3)	-27(4)
C(49)	41(4)	114(8)	74(6)	25(5)	-2(4)	-8(4)
C(51)	59(4)	46(4)	50(4)	-7(3)	-15(3)	-9(3)
C(52)	66(5)	74(6)	81(6)	-5(5)	-9(4)	-1(4)
C(53)	80(7)	94(8)	108(9)	-21(7)	8(6)	13(6)
C(54)	128(10)	90(8)	83(8)	-28(6)	18(7)	2(7)
C(55)	159(12)	98(8)	61(6)	-22(5)	-29(7)	-7(8)
C(56)	94(7)	73(5)	61(5)	-21(4)	-23(5)	-1(5)
C(57)	47(4)	52(4)	62(4)	-6(3)	-9(3)	-3(3)
C(58)	56(5)	71(5)	79(6)	6(4)	-10(4)	-11(4)
C(59)	71(6)	123(9)	101(8)	33(7)	-6(6)	-23(6)
C(60)	67(6)	101(8)	149(12)	10(8)	-11(7)	-22(6)
C(61)	58(6)	111(9)	135(10)	5(7)	-39(6)	-19(6)
C(62)	60(5)	76(6)	94(7)	9(5)	-27(5)	-15(4)

X-ray Data Collection, Structure Solution and Refinement for $[Am(L)_2(NO_3)][2(NO_3)]$, Am 2:1.

A yellow crystal of approximate dimensions $0.5 \times 0.2 \times 0.1$ mm was mounted in a radiological fume hood in a 0.3 mm quartz capillary tube that was covered with a clear coat of colorless nail polish and contained a small amount of original mother liquor. The capillary was sealed using sealing wax to provide a solvent saturated environment and was secured with super glue in a MiTeGen Reusable BaseTM before being completely encapsulated in polyester heat-shrink tubing to create a non-dispersible configuration. The sample configuration was then transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at room temperature (45 sec/frame scan time for a sphere of diffraction data) using AgK α radiation. The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The centrosymmetric triclinic space group *P-1* was assigned and later determined to be correct.

The initial structure was solved by direct methods using Pu in place of Am, since Am is not recognized by APEX3. The structure was refined on F² by full-matrix least-squares techniques using Am, the scattering factors for which were taken from the International Tables for Crystallography Volume C.⁶ Relatively high thermal motion throughout the model prevented universal anisotropic refinement, and other attempts to collect higher quality data at room temperature were unsuccessful. There were several high residual peaks in solvent accessible voids that could not be identified but were likely ethanol and chloroform molecules of solvation.

The SQUEEZE routine in the PLATON⁵ program package was used to account for the electrons in the solvent accessible voids. All hydrogen atoms were included using a riding model.

Table S9. Crystal data and structure refinement for [Am(L)₂(NO₃)][2(NO₃)], **Am 2:1**.

Empirical formula	[C ₆₂ H ₅₄ AmN ₂ O ₆ P ₄ (NO ₃)][2(NO ₃)]	
Formula weight	1475.98	
Temperature	293(2) K	
Wavelength	0.56086 Å	
Crystal system	Triclinic	
Space group	P-1	
Unit cell dimensions	a = 13.720(4) Å	α= 67.013(6)°.
	b = 17.293(5) Å	β= 81.140(6)°.
	c = 17.989(5) Å	γ = 80.468(7)°.
Volume	3856(2) Å ³	
Z	2	
Density (calculated)	1.271 mg/m ³	
Absorption coefficient	1.382 mm ⁻¹	
F(000)	1472	
Crystal size	0.5 x 0.2 x 0.1 mm	
Theta range for data collection	1.950 to 20.520°.	
Index ranges	-17 ≤ h ≤ 17, -21 ≤ k ≤ 21, -22 ≤ l ≤ 22	
Reflections collected	50293	
Independent reflections	15565 [R(int) = 0.1360]	
Completeness to theta = 19.665°	99.0 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.7445 and 0.6426	
Refinement method	Full-matrix least-squares on F ²	
Data / restraints / parameters	15565 / 0 / 514	
Goodness-of-fit on F ²	1.086	
Final R indices [I>2sigma(I)]	R1 = 0.1253, wR2 = 0.2579	
R indices (all data)	R1 = 0.1841, wR2 = 0.2906	
Largest diff. peak and hole	3.326 and -2.574 e.Å ⁻³	

Table S10. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Am}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Am 2:1**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Am(1)	589(1)	2318(1)	3470(1)	43(1)
P(1)	-554(4)	1847(3)	1974(3)	44(1)
P(2)	2494(3)	3674(3)	2234(2)	35(1)
P(3)	1885(3)	2114(3)	5209(3)	42(1)
P(4)	-1888(3)	3349(3)	3854(3)	41(1)
N(1)	314(10)	3744(7)	1493(7)	35(3)
N(2)	155(8)	3781(6)	4420(6)	26(2)
O(1)	-1(8)	3557(6)	2266(5)	36(2)
O(2)	97(9)	1752(6)	2587(7)	49(3)
O(3)	2061(9)	2876(7)	2777(7)	50(3)
O(4)	572(7)	3629(6)	3755(5)	33(2)
O(5)	1345(9)	1896(6)	4695(7)	46(3)
O(6)	-1117(9)	2622(7)	3908(7)	56(3)
C(1)	-152(12)	3470(10)	1022(10)	42(4)
C(2)	164(13)	3668(11)	238(9)	49(4)
C(3)	936(17)	4134(13)	-119(11)	68(6)
C(4)	1380(14)	4423(11)	349(11)	57(5)
C(5)	1073(13)	4215(10)	1155(9)	43(4)
C(6)	-993(12)	2969(11)	1426(9)	47(4)
C(7)	123(15)	1445(11)	1229(11)	55(5)
C(8)	-315(19)	1492(15)	556(14)	81(6)
C(9)	210(30)	1111(19)	52(19)	117(10)
C(10)	1150(30)	780(20)	170(20)	139(12)
C(11)	1670(30)	690(20)	810(20)	146(13)
C(12)	1080(20)	1051(16)	1337(16)	94(8)
C(13)	-1646(14)	1331(11)	2436(12)	54(5)
C(14)	-1687(18)	869(14)	3262(14)	76(6)
C(15)	-2520(20)	441(16)	3668(17)	96(8)
C(16)	-3180(20)	469(17)	3214(17)	96(8)
C(17)	-3230(20)	908(18)	2421(18)	112(9)

C(18)	-2390(20)	1361(17)	2013(17)	95(8)
C(19)	1565(12)	4498(10)	1676(10)	44(4)
C(20)	3388(13)	3443(11)	1514(11)	51(4)
C(21)	3635(19)	2572(16)	1603(15)	88(7)
C(22)	4370(20)	2390(20)	1029(18)	115(10)
C(23)	4810(20)	3002(19)	439(18)	111(9)
C(24)	4569(18)	3811(15)	353(14)	79(6)
C(25)	3880(15)	4042(13)	901(12)	65(5)
C(26)	3097(11)	4126(10)	2773(8)	38(4)
C(27)	2797(16)	4923(13)	2794(12)	66(5)
C(28)	3303(18)	5211(15)	3221(13)	79(6)
C(29)	4069(18)	4707(14)	3630(14)	79(6)
C(30)	4320(18)	3932(14)	3636(13)	77(6)
C(31)	3826(14)	3633(12)	3209(11)	58(5)
C(32)	705(13)	3623(9)	5041(9)	38(4)
C(33)	265(13)	3785(11)	5697(10)	49(4)
C(34)	-705(14)	4124(11)	5749(11)	55(5)
C(35)	-1241(12)	4311(11)	5096(11)	51(4)
C(36)	-818(11)	4131(9)	4442(9)	35(3)
C(37)	1747(11)	3229(12)	4956(8)	50(5)
C(38)	1365(13)	1659(11)	6235(10)	49(4)
C(39)	1750(20)	1694(17)	6866(17)	103(8)
C(40)	1350(30)	1280(20)	7670(20)	128(11)
C(41)	520(20)	881(18)	7769(19)	108(9)
C(42)	100(20)	850(19)	7143(19)	116(10)
C(43)	530(18)	1235(14)	6364(14)	77(6)
C(44)	3178(13)	1778(10)	5071(11)	49(4)
C(45)	3485(15)	1366(12)	4551(11)	60(5)
C(46)	4460(20)	1118(16)	4393(16)	92(7)
C(47)	5120(20)	1251(17)	4792(16)	102(8)
C(48)	4870(20)	1685(17)	5303(17)	105(9)
C(49)	3848(19)	1952(15)	5441(15)	87(7)
C(50)	-1365(11)	4285(9)	3733(10)	39(4)
C(51)	-2647(13)	3618(13)	3038(11)	53(5)
C(52)	-2721(17)	4409(14)	2421(13)	75(6)
C(53)	-3270(20)	4555(19)	1749(17)	103(8)

C(54)	-3710(20)	3903(18)	1813(17)	101(8)
C(55)	-3680(20)	3192(17)	2366(16)	94(8)
C(56)	-3143(17)	2993(14)	3020(13)	74(6)
C(57)	-2682(13)	3093(11)	4783(11)	49(4)
C(58)	-3530(15)	3649(12)	4870(12)	60(5)
C(59)	-4059(18)	3459(15)	5603(14)	78(6)
C(60)	-3800(20)	2788(17)	6224(17)	99(8)
C(61)	-3000(20)	2217(17)	6186(16)	94(8)
C(62)	-2438(18)	2355(15)	5436(14)	78(6)
N(3)	1200(30)	460(30)	4060(20)	177(13)
N(4)	7390(17)	3466(16)	-309(14)	94(6)
N(5)	-6804(19)	3809(17)	6352(17)	103(7)
O(7)	1842(11)	1015(9)	3495(8)	71(4)
O(8)	193(12)	836(10)	4361(9)	84(4)
O(9)	1508(19)	-227(17)	4145(15)	146(8)
O(10)	6879(18)	3786(15)	-888(14)	139(8)
O(11)	7490(20)	2703(18)	2(16)	161(9)
O(12)	7520(20)	3902(17)	-13(16)	155(9)
O(13)	-6343(19)	4210(16)	6631(15)	147(8)
O(14)	-6860(20)	4171(19)	5720(19)	176(11)
O(15)	-6600(30)	3080(20)	6720(20)	207(13)

Table S11. Bond lengths [Å] and angles [°] for $[\text{Am}(\mathbf{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Am 2:1**.

Am(1)-O(3)	2.344(11)
Am(1)-O(5)	2.380(11)
Am(1)-O(2)	2.387(10)
Am(1)-O(6)	2.392(12)
Am(1)-O(1)	2.506(10)
Am(1)-O(4)	2.506(9)
Am(1)-O(8)	2.534(15)
Am(1)-O(7)	2.589(14)
Am(1)-N(3)	2.98(4)

Am(1)-N(1)	3.482(11)
Am(1)-N(2)	3.498(10)
Am(1)-P(4)	3.665(4)
P(1)-O(2)	1.468(12)
P(1)-C(13)	1.792(19)
P(1)-C(7)	1.809(18)
P(1)-C(6)	1.844(17)
P(2)-O(3)	1.500(12)
P(2)-C(20)	1.758(19)
P(2)-C(26)	1.814(15)
P(2)-C(19)	1.827(16)
P(3)-O(5)	1.456(12)
P(3)-C(37)	1.78(2)
P(3)-C(38)	1.784(18)
P(3)-C(44)	1.784(18)
P(4)-O(6)	1.486(12)
P(4)-C(57)	1.788(18)
P(4)-C(51)	1.799(17)
P(4)-C(50)	1.801(15)
N(1)-O(1)	1.317(14)
N(1)-C(5)	1.35(2)
N(1)-C(1)	1.389(19)
N(2)-O(4)	1.346(13)
N(2)-C(32)	1.359(18)
N(2)-C(36)	1.373(18)
C(1)-C(2)	1.33(2)
C(1)-C(6)	1.48(2)
C(2)-C(3)	1.36(3)
C(3)-C(4)	1.38(3)
C(4)-C(5)	1.37(2)
C(5)-C(19)	1.50(2)
C(7)-C(12)	1.38(3)
C(7)-C(8)	1.40(3)
C(8)-C(9)	1.37(3)
C(9)-C(10)	1.32(4)
C(10)-C(11)	1.39(4)

C(11)-C(12)	1.41(4)
C(13)-C(18)	1.35(3)
C(13)-C(14)	1.38(3)
C(14)-C(15)	1.41(3)
C(15)-C(16)	1.30(3)
C(16)-C(17)	1.33(3)
C(17)-C(18)	1.44(4)
C(20)-C(25)	1.36(3)
C(20)-C(21)	1.44(3)
C(21)-C(22)	1.42(4)
C(22)-C(23)	1.32(4)
C(23)-C(24)	1.34(3)
C(24)-C(25)	1.39(3)
C(26)-C(31)	1.33(2)
C(26)-C(27)	1.38(2)
C(27)-C(28)	1.38(3)
C(28)-C(29)	1.36(3)
C(29)-C(30)	1.32(3)
C(30)-C(31)	1.38(3)
C(32)-C(33)	1.35(2)
C(32)-C(37)	1.49(2)
C(33)-C(34)	1.37(2)
C(34)-C(35)	1.38(2)
C(35)-C(36)	1.35(2)
C(36)-C(50)	1.49(2)
C(38)-C(39)	1.35(3)
C(38)-C(43)	1.40(3)
C(39)-C(40)	1.41(4)
C(40)-C(41)	1.39(4)
C(41)-C(42)	1.36(4)
C(42)-C(43)	1.38(3)
C(44)-C(49)	1.35(3)
C(44)-C(45)	1.36(2)
C(45)-C(46)	1.36(3)
C(46)-C(47)	1.33(3)
C(47)-C(48)	1.37(3)

C(48)-C(49)	1.42(3)
C(51)-C(56)	1.38(3)
C(51)-C(52)	1.39(3)
C(52)-C(53)	1.44(3)
C(53)-C(54)	1.32(3)
C(54)-C(55)	1.24(3)
C(55)-C(56)	1.38(3)
C(57)-C(62)	1.39(3)
C(57)-C(58)	1.41(3)
C(58)-C(59)	1.35(3)
C(59)-C(60)	1.30(3)
C(60)-C(61)	1.36(3)
C(61)-C(62)	1.40(3)
N(3)-O(9)	1.15(4)
N(3)-O(7)	1.40(4)
N(3)-O(8)	1.54(4)
N(4)-O(12)	1.13(3)
N(4)-O(11)	1.21(3)
N(4)-O(10)	1.24(3)
N(5)-O(14)	1.07(3)
N(5)-O(15)	1.18(3)
N(5)-O(13)	1.28(3)
O(3)-Am(1)-O(5)	87.6(4)
O(3)-Am(1)-O(2)	102.9(4)
O(5)-Am(1)-O(2)	141.5(3)
O(3)-Am(1)-O(6)	146.3(4)
O(5)-Am(1)-O(6)	101.6(4)
O(2)-Am(1)-O(6)	90.0(4)
O(3)-Am(1)-O(1)	76.8(4)
O(5)-Am(1)-O(1)	144.8(3)
O(2)-Am(1)-O(1)	73.4(3)
O(6)-Am(1)-O(1)	77.4(4)
O(3)-Am(1)-O(4)	75.1(4)
O(5)-Am(1)-O(4)	73.2(3)
O(2)-Am(1)-O(4)	145.2(3)

O(6)-Am(1)-O(4)	76.7(4)
O(1)-Am(1)-O(4)	72.3(3)
O(3)-Am(1)-O(8)	133.1(5)
O(5)-Am(1)-O(8)	71.9(4)
O(2)-Am(1)-O(8)	74.2(4)
O(6)-Am(1)-O(8)	80.2(5)
O(1)-Am(1)-O(8)	140.3(4)
O(4)-Am(1)-O(8)	132.7(4)
O(3)-Am(1)-O(7)	75.1(4)
O(5)-Am(1)-O(7)	75.2(4)
O(2)-Am(1)-O(7)	72.0(4)
O(6)-Am(1)-O(7)	138.5(4)
O(1)-Am(1)-O(7)	128.5(4)
O(4)-Am(1)-O(7)	137.0(4)
O(8)-Am(1)-O(7)	59.2(5)
O(3)-Am(1)-N(3)	102.7(8)
O(5)-Am(1)-N(3)	71.2(7)
O(2)-Am(1)-N(3)	70.3(7)
O(6)-Am(1)-N(3)	111.0(8)
O(1)-Am(1)-N(3)	142.6(7)
O(4)-Am(1)-N(3)	144.5(7)
O(8)-Am(1)-N(3)	31.2(8)
O(7)-Am(1)-N(3)	28.0(7)
O(3)-Am(1)-N(1)	66.6(3)
O(5)-Am(1)-N(1)	149.3(3)
O(2)-Am(1)-N(1)	64.6(3)
O(6)-Am(1)-N(1)	92.4(4)
O(1)-Am(1)-N(1)	17.2(3)
O(4)-Am(1)-N(1)	83.8(3)
O(8)-Am(1)-N(1)	138.1(4)
O(7)-Am(1)-N(1)	111.4(4)
N(3)-Am(1)-N(1)	128.7(7)
O(3)-Am(1)-N(2)	90.6(3)
O(5)-Am(1)-N(2)	64.6(3)
O(2)-Am(1)-N(2)	150.0(3)
O(6)-Am(1)-N(2)	65.5(4)

O(1)-Am(1)-N(2)	84.0(3)
O(4)-Am(1)-N(2)	17.7(3)
O(8)-Am(1)-N(2)	115.5(4)
O(7)-Am(1)-N(2)	137.9(4)
N(3)-Am(1)-N(2)	133.2(7)
N(1)-Am(1)-N(2)	97.8(3)
O(3)-Am(1)-P(4)	131.5(3)
O(5)-Am(1)-P(4)	105.2(3)
O(2)-Am(1)-P(4)	95.3(3)
O(6)-Am(1)-P(4)	14.8(3)
O(1)-Am(1)-P(4)	65.9(2)
O(4)-Am(1)-P(4)	65.0(2)
O(8)-Am(1)-P(4)	95.0(4)
O(7)-Am(1)-P(4)	153.2(3)
N(3)-Am(1)-P(4)	125.8(8)
N(1)-Am(1)-P(4)	82.2(2)
N(2)-Am(1)-P(4)	56.77(19)
O(2)-P(1)-C(13)	111.1(8)
O(2)-P(1)-C(7)	109.8(8)
C(13)-P(1)-C(7)	111.3(8)
O(2)-P(1)-C(6)	111.2(6)
C(13)-P(1)-C(6)	106.1(8)
C(7)-P(1)-C(6)	107.2(8)
O(2)-P(1)-Am(1)	19.9(4)
C(13)-P(1)-Am(1)	110.8(6)
C(7)-P(1)-Am(1)	125.1(7)
C(6)-P(1)-Am(1)	93.0(4)
O(3)-P(2)-C(20)	108.5(8)
O(3)-P(2)-C(26)	113.3(7)
C(20)-P(2)-C(26)	107.9(7)
O(3)-P(2)-C(19)	112.6(7)
C(20)-P(2)-C(19)	107.3(8)
C(26)-P(2)-C(19)	107.0(7)
O(3)-P(2)-Am(1)	22.0(5)
C(20)-P(2)-Am(1)	124.7(6)
C(26)-P(2)-Am(1)	115.1(5)

C(19)-P(2)-Am(1)	92.0(5)
O(5)-P(3)-C(37)	111.2(6)
O(5)-P(3)-C(38)	108.9(8)
C(37)-P(3)-C(38)	106.4(7)
O(5)-P(3)-C(44)	109.9(7)
C(37)-P(3)-C(44)	107.5(7)
C(38)-P(3)-C(44)	112.9(8)
O(5)-P(3)-Am(1)	19.4(4)
C(37)-P(3)-Am(1)	92.9(5)
C(38)-P(3)-Am(1)	122.5(6)
C(44)-P(3)-Am(1)	111.5(6)
O(6)-P(4)-C(57)	109.0(8)
O(6)-P(4)-C(51)	113.1(8)
C(57)-P(4)-C(51)	108.0(8)
O(6)-P(4)-C(50)	112.4(7)
C(57)-P(4)-C(50)	106.1(7)
C(51)-P(4)-C(50)	107.9(8)
O(6)-P(4)-Am(1)	24.4(5)
C(57)-P(4)-Am(1)	128.1(6)
C(51)-P(4)-Am(1)	112.7(5)
C(50)-P(4)-Am(1)	90.6(5)
O(1)-N(1)-C(5)	120.0(11)
O(1)-N(1)-C(1)	119.9(12)
C(5)-N(1)-C(1)	120.0(13)
O(1)-N(1)-Am(1)	34.3(6)
C(5)-N(1)-Am(1)	113.8(9)
C(1)-N(1)-Am(1)	115.7(9)
O(4)-N(2)-C(32)	120.2(11)
O(4)-N(2)-C(36)	118.8(11)
C(32)-N(2)-C(36)	120.9(12)
O(4)-N(2)-Am(1)	34.5(5)
C(32)-N(2)-Am(1)	116.6(8)
C(36)-N(2)-Am(1)	112.8(7)
N(1)-O(1)-Am(1)	128.5(8)
P(1)-O(2)-Am(1)	148.0(7)
P(2)-O(3)-Am(1)	144.1(7)

N(2)-O(4)-Am(1)	127.9(7)
P(3)-O(5)-Am(1)	148.8(6)
P(4)-O(6)-Am(1)	140.8(7)
C(2)-C(1)-N(1)	119.6(15)
C(2)-C(1)-C(6)	123.1(14)
N(1)-C(1)-C(6)	117.3(14)
C(1)-C(2)-C(3)	121.8(16)
C(2)-C(3)-C(4)	118.3(17)
C(5)-C(4)-C(3)	120.5(18)
N(1)-C(5)-C(4)	119.7(16)
N(1)-C(5)-C(19)	119.0(13)
C(4)-C(5)-C(19)	121.3(16)
N(1)-C(5)-Am(1)	49.1(7)
C(4)-C(5)-Am(1)	148.5(11)
C(19)-C(5)-Am(1)	77.5(8)
C(1)-C(6)-P(1)	111.4(11)
C(1)-C(6)-Am(1)	81.5(8)
P(1)-C(6)-Am(1)	61.2(4)
C(12)-C(7)-C(8)	119(2)
C(12)-C(7)-P(1)	119.8(16)
C(8)-C(7)-P(1)	121.5(17)
C(9)-C(8)-C(7)	118(3)
C(10)-C(9)-C(8)	119(3)
C(9)-C(10)-C(11)	128(4)
C(10)-C(11)-C(12)	111(4)
C(7)-C(12)-C(11)	125(3)
C(18)-C(13)-C(14)	120(2)
C(18)-C(13)-P(1)	123.0(18)
C(14)-C(13)-P(1)	116.6(16)
C(13)-C(14)-C(15)	120(2)
C(16)-C(15)-C(14)	116(3)
C(15)-C(16)-C(17)	129(3)
C(16)-C(17)-C(18)	115(3)
C(13)-C(18)-C(17)	120(3)
C(5)-C(19)-P(2)	111.6(11)
C(5)-C(19)-Am(1)	81.9(9)

P(2)-C(19)-Am(1)	61.9(5)
C(25)-C(20)-C(21)	118.8(19)
C(25)-C(20)-P(2)	123.3(15)
C(21)-C(20)-P(2)	117.8(16)
C(22)-C(21)-C(20)	118(2)
C(23)-C(22)-C(21)	120(3)
C(22)-C(23)-C(24)	122(3)
C(23)-C(24)-C(25)	121(3)
C(20)-C(25)-C(24)	120(2)
C(31)-C(26)-C(27)	119.4(16)
C(31)-C(26)-P(2)	116.9(14)
C(27)-C(26)-P(2)	123.5(14)
C(28)-C(27)-C(26)	119(2)
C(29)-C(28)-C(27)	120(2)
C(30)-C(29)-C(28)	120(2)
C(29)-C(30)-C(31)	121(2)
C(26)-C(31)-C(30)	120.3(19)
C(33)-C(32)-N(2)	118.6(15)
C(33)-C(32)-C(37)	125.2(14)
N(2)-C(32)-C(37)	116.0(12)
C(32)-C(33)-C(34)	122.5(16)
C(33)-C(34)-C(35)	117.9(15)
C(36)-C(35)-C(34)	120.4(16)
C(35)-C(36)-N(2)	119.6(13)
C(35)-C(36)-C(50)	123.0(14)
N(2)-C(36)-C(50)	117.3(12)
C(35)-C(36)-Am(1)	148.8(11)
N(2)-C(36)-Am(1)	49.8(6)
C(50)-C(36)-Am(1)	75.1(7)
C(32)-C(37)-P(3)	115.1(12)
C(32)-C(37)-Am(1)	83.4(8)
P(3)-C(37)-Am(1)	61.9(5)
C(39)-C(38)-C(43)	121(2)
C(39)-C(38)-P(3)	123.0(18)
C(43)-C(38)-P(3)	116.2(15)
C(38)-C(39)-C(40)	121(3)

C(41)-C(40)-C(39)	116(3)
C(42)-C(41)-C(40)	124(3)
C(41)-C(42)-C(43)	118(3)
C(42)-C(43)-C(38)	120(2)
C(49)-C(44)-C(45)	120(2)
C(49)-C(44)-P(3)	121.6(16)
C(45)-C(44)-P(3)	118.2(14)
C(46)-C(45)-C(44)	122(2)
C(47)-C(46)-C(45)	118(3)
C(46)-C(47)-C(48)	124(3)
C(47)-C(48)-C(49)	117(3)
C(44)-C(49)-C(48)	120(2)
C(36)-C(50)-P(4)	110.6(11)
C(36)-C(50)-Am(1)	84.4(8)
P(4)-C(50)-Am(1)	63.3(4)
C(56)-C(51)-C(52)	119.5(18)
C(56)-C(51)-P(4)	117.9(16)
C(52)-C(51)-P(4)	122.5(15)
C(51)-C(52)-C(53)	119(2)
C(54)-C(53)-C(52)	115(3)
C(55)-C(54)-C(53)	128(3)
C(54)-C(55)-C(56)	121(3)
C(51)-C(56)-C(55)	117(2)
C(62)-C(57)-C(58)	119.7(18)
C(62)-C(57)-P(4)	119.5(16)
C(58)-C(57)-P(4)	120.7(14)
C(59)-C(58)-C(57)	118.4(19)
C(60)-C(59)-C(58)	122(3)
C(59)-C(60)-C(61)	123(3)
C(60)-C(61)-C(62)	118(3)
C(57)-C(62)-C(61)	118(2)
O(9)-N(3)-O(7)	110(4)
O(9)-N(3)-O(8)	131(4)
O(7)-N(3)-O(8)	118(3)
O(9)-N(3)-Am(1)	168(3)
O(7)-N(3)-Am(1)	60.2(16)

O(8)-N(3)-Am(1)	58.2(14)
O(12)-N(4)-O(11)	125(3)
O(12)-N(4)-O(10)	116(3)
O(11)-N(4)-O(10)	117(3)
O(14)-N(5)-O(15)	133(4)
O(14)-N(5)-O(13)	108(3)
O(15)-N(5)-O(13)	107(3)
N(3)-O(7)-Am(1)	91.8(19)
N(3)-O(8)-Am(1)	90.7(17)
N(3)-O(9)-Am(1)	9(2)

Table S12. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Am}(\mathbf{L})_2(\text{NO}_3)][\text{2}(\text{NO}_3)]$, **Am 2:1**. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^*{}^2 U^{11} + \dots + 2 h k a^* b^* U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Am(1)	39(1)	50(1)	44(1)	-22(1)	-1(1)	-8(1)
P(1)	51(3)	43(2)	42(2)	-21(2)	-3(2)	-10(2)
P(2)	28(2)	49(2)	34(2)	-20(2)	-3(2)	-8(2)
P(3)	47(3)	35(2)	44(2)	-13(2)	-12(2)	-3(2)
P(4)	26(2)	57(3)	47(2)	-28(2)	-3(2)	-7(2)
N(1)	41(8)	34(6)	33(6)	-11(5)	-13(6)	-6(6)
O(1)	38(6)	49(6)	24(5)	-16(4)	2(4)	-9(5)
O(2)	70(9)	35(6)	49(7)	-21(5)	-8(6)	-8(5)
O(3)	43(7)	57(7)	51(7)	-15(6)	-11(5)	-13(6)
O(4)	36(6)	43(6)	20(5)	-10(4)	4(4)	-12(5)
O(5)	55(8)	30(5)	52(7)	-16(5)	0(6)	-6(5)
O(6)	41(7)	50(7)	69(8)	-15(6)	-6(6)	0(6)
C(1)	30(9)	41(8)	58(10)	-21(8)	-15(8)	7(7)
C(2)	52(11)	75(12)	28(8)	-22(8)	-1(7)	-24(9)
C(3)	85(16)	74(13)	29(9)	0(9)	-8(9)	-14(12)
C(4)	52(12)	55(11)	49(10)	-8(8)	4(9)	-8(9)

C(5)	53(11)	45(9)	25(7)	-11(6)	0(7)	2(8)
C(6)	40(10)	78(12)	44(9)	-42(9)	-27(8)	5(8)
C(7)	70(14)	54(10)	49(10)	-29(8)	-5(9)	-7(9)
C(13)	50(12)	55(11)	67(12)	-34(9)	-1(9)	-6(9)
C(20)	49(11)	61(11)	53(10)	-28(9)	-17(9)	-10(9)
C(26)	28(9)	59(10)	27(7)	-14(7)	2(6)	-19(7)
C(32)	58(11)	35(8)	32(8)	-23(6)	-5(7)	-9(7)
C(33)	49(11)	63(11)	44(9)	-27(8)	-16(8)	-8(9)
C(34)	54(12)	62(11)	63(11)	-45(10)	16(9)	-8(9)
C(35)	26(9)	68(11)	80(13)	-45(10)	-13(9)	-5(8)
C(36)	36(9)	33(7)	49(9)	-27(7)	-12(7)	-5(6)
C(37)	16(8)	112(15)	17(7)	-19(8)	-5(6)	-6(8)
C(38)	47(11)	51(10)	51(10)	-21(8)	-6(8)	-4(8)
C(44)	47(11)	38(9)	61(11)	-19(8)	-20(9)	10(7)
C(50)	25(8)	45(9)	55(10)	-31(8)	5(7)	0(7)
C(51)	34(10)	87(13)	60(11)	-52(10)	-5(8)	-4(9)
C(57)	37(10)	61(11)	59(11)	-27(9)	-3(8)	-24(8)

X-ray Data Collection, Structure Solution and Refinement for [Eu(L)₂(NO₃)][2(NO₃)], Eu 2:1.

A colorless crystal of approximate dimensions 0.576 x 0.454 x 0.350 mm was mounted in a quartz capillary tube containing a small amount of its original mother liquor which was sealed using sealing wax to provide a solvent saturated environment. The crystal-containing capillary was then secured with mounting putty in a MiTeGen Reusable Base™ before being transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at 80 K (20 sec/frame scan time for a sphere of diffraction data) using AgK α radiation. The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were

carried out using the SHELXL⁴ program. The centrosymmetric triclinic space group *P-1* was assigned and later determined to be correct.

The structure was solved by direct methods and refined on F² by full-matrix least-squares techniques. There were four disordered chloroform molecules of solvation present, only some of which could be satisfactorily modeled using multiple components with partial site-occupancy-factors. Hydrogen atoms were included using a riding model except in the case of solvent molecules for which they were not included.

Table S13. Crystal data and structure refinement for [Eu(L)₂(NO₃)][2(NO₃)], **Eu 2:1**.

Empirical formula	[C ₆₂ H ₅₄ N ₂ EuO ₆ P ₄ (NO ₃)][2(NO ₃)•4(CHCl ₄)		
Formula weight	1862.41		
Temperature	83(2) K		
Wavelength	0.56086 Å		
Crystal system	Triclinic		
Space group	P-1		
Unit cell dimensions	a = 13.2172(7) Å	α= 83.232(2)°.	
	b = 15.2557(9) Å	β= 77.334(2)°.	
	c = 19.8448(12) Å	γ = 82.073(2)°.	
Volume	3850.9(4) Å ³		
Z	2		
Density (calculated)	1.606 mg/m ³		
Absorption coefficient	0.725 mm ⁻¹		
F(000)	1872		
Crystal size	0.576 x 0.454 x 0.350 mm ³		
Theta range for data collection	1.857 to 22.164°.		
Index ranges	-17 ≤ h ≤ 17, -20 ≤ k ≤ 20, -26 ≤ l ≤ 26		
Reflections collected	164656		
Independent reflections	19449 [R(int) = 0.0328]		

Completeness to theta = 19.665°	99.8 %
Absorption correction	Numerical
Max. and min. transmission	0.8944 and 0.6399
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	19449 / 0 / 864
Goodness-of-fit on F ²	1.041
Final R indices [I>2sigma(I)]	R1 = 0.0874, wR2 = 0.2283
R indices (all data)	R1 = 0.0936, wR2 = 0.2352
Largest diff. peak and hole	6.895 and -4.195 e.Å ⁻³

Table S14. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Eu}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Eu 2:1**. U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U(eq)
Eu(1)	4828(1)	7833(1)	2620(1)	15(1)
P(1)	2493(1)	6928(1)	3504(1)	20(1)
P(2)	6027(1)	8011(1)	4102(1)	16(1)
P(3)	7244(1)	6496(1)	1942(1)	16(1)
P(4)	3628(1)	8324(1)	1083(1)	19(1)
N(1)	4737(4)	6278(3)	4032(2)	16(1)
N(2)	5013(4)	6381(3)	1375(2)	17(1)
O(1)	5146(3)	6475(3)	3366(2)	17(1)
O(2)	3191(3)	7656(3)	3291(2)	20(1)
O(3)	5486(3)	8260(3)	3506(2)	22(1)
O(4)	4626(3)	6601(3)	2019(2)	17(1)
O(5)	6549(3)	7340(3)	2112(2)	19(1)
O(6)	4033(4)	8464(3)	1706(2)	23(1)
N(3)	4741(5)	9761(4)	2553(3)	29(1)
O(7)	5525(4)	9288(3)	2221(2)	29(1)
O(8)	3980(4)	9361(3)	2882(2)	26(1)
O(9)	4722(6)	10558(3)	2552(3)	49(2)
C(1)	3810(4)	5909(4)	4203(3)	18(1)

C(2)	3405(5)	5666(4)	4897(3)	22(1)
C(3)	3913(5)	5803(4)	5402(3)	22(1)
C(4)	4837(5)	6184(4)	5218(3)	21(1)
C(5)	5255(4)	6414(3)	4526(3)	15(1)
C(6)	3252(5)	5857(4)	3633(3)	23(1)
C(7)	1736(5)	6876(5)	2871(3)	26(1)
C(8)	1660(6)	6097(5)	2594(4)	32(1)
C(9)	1126(6)	6140(6)	2055(4)	38(2)
C(10)	665(6)	6950(6)	1808(4)	39(2)
C(11)	707(6)	7718(6)	2104(4)	34(2)
C(12)	1254(5)	7688(5)	2624(4)	31(1)
C(13)	1635(5)	7128(5)	4320(3)	27(1)
C(14)	1777(6)	7821(6)	4669(4)	38(2)
C(15)	1164(8)	7945(9)	5331(5)	60(3)
C(16)	434(8)	7394(9)	5634(5)	62(3)
C(17)	294(6)	6707(8)	5296(5)	63(4)
C(18)	892(5)	6543(6)	4619(5)	44(2)
C(19)	6258(4)	6813(3)	4293(3)	17(1)
C(20)	7284(5)	8408(4)	3907(3)	21(1)
C(21)	7550(5)	8941(4)	3287(4)	27(1)
C(22)	8500(6)	9287(5)	3127(5)	38(2)
C(23)	9178(6)	9111(5)	3584(5)	41(2)
C(24)	8914(6)	8570(5)	4198(5)	37(2)
C(25)	7976(5)	8218(4)	4356(4)	28(1)
C(26)	5240(5)	8462(4)	4864(3)	21(1)
C(27)	4336(5)	9036(4)	4793(4)	27(1)
C(28)	3703(6)	9392(5)	5385(5)	41(2)
C(29)	3969(8)	9170(5)	6021(4)	46(2)
C(30)	4849(8)	8604(5)	6093(4)	42(2)
C(31)	5494(6)	8241(4)	5513(3)	31(1)
C(32)	5964(4)	5885(4)	1233(3)	20(1)
C(33)	6368(5)	5643(5)	571(4)	28(1)
C(34)	5791(6)	5882(5)	54(4)	32(1)
C(35)	4828(5)	6357(5)	215(3)	27(1)
C(36)	4434(4)	6613(4)	880(3)	19(1)
C(37)	6523(4)	5624(4)	1814(3)	19(1)

C(38)	7937(4)	6053(4)	2613(3)	19(1)
C(39)	8008(6)	5165(5)	2863(4)	32(2)
C(40)	8564(7)	4866(5)	3379(4)	38(2)
C(41)	9048(5)	5439(5)	3649(4)	30(1)
C(42)	8984(5)	6332(5)	3405(4)	28(1)
C(43)	8428(5)	6636(4)	2882(3)	24(1)
C(44)	8186(4)	6711(4)	1163(3)	20(1)
C(45)	8179(5)	7572(5)	843(4)	31(1)
C(46)	8931(6)	7760(6)	252(4)	42(2)
C(47)	9689(6)	7099(6)	-13(4)	37(2)
C(48)	9707(5)	6241(5)	306(4)	31(1)
C(49)	8959(5)	6040(5)	897(3)	26(1)
C(50)	3414(5)	7175(4)	1064(3)	21(1)
C(51)	2388(5)	8973(5)	1064(3)	28(1)
C(52)	1803(6)	8859(6)	585(4)	38(2)
C(53)	853(7)	9388(7)	578(5)	52(2)
C(54)	524(7)	10028(7)	1030(6)	59(3)
C(55)	1081(7)	10130(6)	1510(6)	53(2)
C(56)	2023(6)	9600(5)	1542(4)	36(2)
C(57)	4547(5)	8606(4)	308(3)	24(1)
C(58)	5490(5)	8881(5)	369(4)	28(1)
C(59)	6226(6)	9093(6)	-222(4)	38(2)
C(60)	6020(7)	9039(6)	-865(4)	45(2)
C(61)	5087(8)	8770(7)	-929(4)	48(2)
C(62)	4350(6)	8556(6)	-344(4)	38(2)
N(4)	2195(5)	3932(5)	4220(4)	44(2)
O(10)	3012(6)	3487(6)	4144(5)	71(2)
O(11)	1838(9)	4153(7)	4821(5)	84(3)
O(12)	1757(5)	4278(4)	3721(3)	42(1)
N(5)	7685(4)	3549(4)	418(4)	32(1)
O(13)	7396(5)	2786(4)	517(4)	48(2)
O(14)	7669(4)	3987(4)	-146(3)	36(1)
O(15)	7989(4)	3850(4)	897(3)	38(1)
C(63)	1175(14)	3348(12)	1649(10)	97(5)
Cl(1)	2211(5)	2556(4)	1302(3)	91(2)
Cl(1B)	1538(14)	2403(11)	1463(9)	101(4)

Cl(2)	1265(3)	3597(2)	2485(2)	70(1)
Cl(3)	1463(3)	4293(2)	1007(2)	71(1)
C(64)	2159(11)	760(10)	3497(7)	74(3)
Cl(4)	1583(4)	153(3)	4265(3)	102(1)
Cl(5)	1131(4)	992(3)	2998(2)	94(1)
Cl(6)	2364(2)	1818(2)	3703(2)	65(1)
C(65)	4158(9)	13671(8)	2533(6)	60(3)
Cl(7)	4414(5)	12684(4)	2159(3)	123(2)
Cl(8)	5128(3)	13897(2)	2937(2)	69(1)
Cl(9)	3874(2)	14560(2)	1910(1)	47(1)
Cl(11)	-2890(20)	11270(20)	2139(16)	508(16)
C(66)	-2512(11)	10861(9)	1570(7)	54(3)
Cl(10)	-1717(8)	11424(7)	1262(5)	103(3)
Cl(12)	-2295(17)	9995(15)	1273(11)	205(8)
C(66A)	-2120(20)	12250(20)	2095(15)	21(5)
Cl(1A)	-2302(7)	12565(6)	2911(5)	92(2)
Cl(2A)	-736(12)	11946(10)	1942(8)	152(5)

Table S15. Bond lengths [Å] and angles [°] for $[\text{Eu}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Eu 2:1**.

Eu(1)-O(2)	2.311(4)
Eu(1)-O(3)	2.319(4)
Eu(1)-O(5)	2.335(4)
Eu(1)-O(6)	2.338(4)
Eu(1)-O(4)	2.415(4)
Eu(1)-O(1)	2.444(4)
Eu(1)-O(7)	2.497(5)
Eu(1)-O(8)	2.505(4)
Eu(1)-N(3)	2.917(5)
Eu(1)-P(1)	3.5689(15)
P(1)-O(2)	1.505(4)
P(1)-C(7)	1.782(6)
P(1)-C(13)	1.795(7)

P(1)-C(6)	1.818(6)
P(2)-O(3)	1.497(4)
P(2)-C(26)	1.790(6)
P(2)-C(20)	1.793(6)
P(2)-C(19)	1.820(5)
P(3)-O(5)	1.503(4)
P(3)-C(44)	1.788(6)
P(3)-C(38)	1.795(6)
P(3)-C(37)	1.811(6)
P(4)-O(6)	1.498(5)
P(4)-C(57)	1.787(6)
P(4)-C(51)	1.800(7)
P(4)-C(50)	1.821(6)
N(1)-O(1)	1.328(6)
N(1)-C(5)	1.362(7)
N(1)-C(1)	1.376(7)
N(2)-O(4)	1.332(6)
N(2)-C(36)	1.358(7)
N(2)-C(32)	1.365(7)
N(3)-O(9)	1.212(7)
N(3)-O(8)	1.264(8)
N(3)-O(7)	1.279(8)
C(1)-C(2)	1.389(8)
C(1)-C(6)	1.495(8)
C(2)-C(3)	1.372(9)
C(3)-C(4)	1.383(8)
C(4)-C(5)	1.387(8)
C(5)-C(19)	1.494(7)
C(7)-C(8)	1.392(11)
C(7)-C(12)	1.399(9)
C(8)-C(9)	1.395(10)
C(9)-C(10)	1.386(12)
C(10)-C(11)	1.385(13)
C(11)-C(12)	1.378(9)
C(13)-C(14)	1.384(12)
C(13)-C(18)	1.406(10)

C(14)-C(15)	1.403(11)
C(15)-C(16)	1.361(18)
C(16)-C(17)	1.363(18)
C(17)-C(18)	1.431(14)
C(20)-C(25)	1.392(9)
C(20)-C(21)	1.397(8)
C(21)-C(22)	1.386(10)
C(22)-C(23)	1.390(12)
C(23)-C(24)	1.393(12)
C(24)-C(25)	1.376(10)
C(26)-C(31)	1.392(9)
C(26)-C(27)	1.403(8)
C(27)-C(28)	1.402(10)
C(28)-C(29)	1.376(14)
C(29)-C(30)	1.374(14)
C(30)-C(31)	1.397(10)
C(32)-C(33)	1.378(9)
C(32)-C(37)	1.490(8)
C(33)-C(34)	1.395(10)
C(34)-C(35)	1.367(10)
C(35)-C(36)	1.390(8)
C(36)-C(50)	1.492(8)
C(38)-C(39)	1.385(9)
C(38)-C(43)	1.386(9)
C(39)-C(40)	1.386(10)
C(40)-C(41)	1.370(11)
C(41)-C(42)	1.388(10)
C(42)-C(43)	1.400(9)
C(44)-C(45)	1.389(9)
C(44)-C(49)	1.404(8)
C(45)-C(46)	1.391(10)
C(46)-C(47)	1.382(12)
C(47)-C(48)	1.384(12)
C(48)-C(49)	1.392(9)
C(51)-C(52)	1.389(11)
C(51)-C(56)	1.391(11)

C(52)-C(53)	1.398(11)
C(53)-C(54)	1.365(17)
C(54)-C(55)	1.359(17)
C(55)-C(56)	1.397(11)
C(57)-C(62)	1.389(9)
C(57)-C(58)	1.402(9)
C(58)-C(59)	1.386(10)
C(59)-C(60)	1.376(12)
C(60)-C(61)	1.387(13)
C(61)-C(62)	1.381(10)
N(4)-O(10)	1.183(10)
N(4)-O(11)	1.251(12)
N(4)-O(12)	1.279(9)
N(5)-O(14)	1.237(9)
N(5)-O(13)	1.255(8)
N(5)-O(15)	1.262(8)
C(63)-Cl(1B)	1.52(2)
C(63)-Cl(1)	1.772(19)
C(63)-Cl(2)	1.777(19)
C(63)-Cl(3)	1.831(19)
C(64)-Cl(4)	1.764(15)
C(64)-Cl(6)	1.781(15)
C(64)-Cl(5)	1.825(15)
C(65)-Cl(7)	1.715(13)
C(65)-Cl(8)	1.745(12)
C(65)-Cl(9)	1.783(12)
Cl(11)-C(66)	1.32(3)
Cl(11)-C(66A)	1.90(4)
Cl(11)-Cl(10)	2.07(3)
C(66)-Cl(10)	1.435(17)
C(66)-Cl(12)	1.48(2)
Cl(10)-Cl(12)	2.41(2)
C(66A)-Cl(1A)	1.70(3)
C(66A)-Cl(2A)	1.80(3)

O(2)-Eu(1)-O(3) 95.05(16)

O(2)-Eu(1)-O(5)	154.34(14)
O(3)-Eu(1)-O(5)	87.01(15)
O(2)-Eu(1)-O(6)	88.80(16)
O(3)-Eu(1)-O(6)	139.91(15)
O(5)-Eu(1)-O(6)	105.99(16)
O(2)-Eu(1)-O(4)	86.95(14)
O(3)-Eu(1)-O(4)	145.58(14)
O(5)-Eu(1)-O(4)	77.37(14)
O(6)-Eu(1)-O(4)	74.39(14)
O(2)-Eu(1)-O(1)	76.96(14)
O(3)-Eu(1)-O(1)	74.23(14)
O(5)-Eu(1)-O(1)	79.04(14)
O(6)-Eu(1)-O(1)	144.72(14)
O(4)-Eu(1)-O(1)	72.79(13)
O(2)-Eu(1)-O(7)	125.15(16)
O(3)-Eu(1)-O(7)	71.81(15)
O(5)-Eu(1)-O(7)	79.79(16)
O(6)-Eu(1)-O(7)	73.50(16)
O(4)-Eu(1)-O(7)	133.18(14)
O(1)-Eu(1)-O(7)	140.58(15)
O(2)-Eu(1)-O(8)	73.82(16)
O(3)-Eu(1)-O(8)	71.61(15)
O(5)-Eu(1)-O(8)	130.42(15)
O(6)-Eu(1)-O(8)	71.28(15)
O(4)-Eu(1)-O(8)	140.66(14)
O(1)-Eu(1)-O(8)	132.17(14)
O(7)-Eu(1)-O(8)	51.37(17)
O(2)-Eu(1)-N(3)	99.35(17)
O(3)-Eu(1)-N(3)	68.70(15)
O(5)-Eu(1)-N(3)	105.17(17)
O(6)-Eu(1)-N(3)	71.30(15)
O(4)-Eu(1)-N(3)	144.91(14)
O(1)-Eu(1)-N(3)	142.30(14)
O(7)-Eu(1)-N(3)	25.87(17)
O(8)-Eu(1)-N(3)	25.53(17)
O(2)-Eu(1)-P(1)	16.54(11)

O(3)-Eu(1)-P(1)	103.60(12)
O(5)-Eu(1)-P(1)	139.09(10)
O(6)-Eu(1)-P(1)	91.08(12)
O(4)-Eu(1)-P(1)	71.83(10)
O(1)-Eu(1)-P(1)	66.67(10)
O(7)-Eu(1)-P(1)	141.12(13)
O(8)-Eu(1)-P(1)	90.06(12)
N(3)-Eu(1)-P(1)	115.55(13)
O(2)-P(1)-C(7)	111.2(3)
O(2)-P(1)-C(13)	109.4(3)
C(7)-P(1)-C(13)	109.3(3)
O(2)-P(1)-C(6)	111.0(3)
C(7)-P(1)-C(6)	109.4(3)
C(13)-P(1)-C(6)	106.6(3)
O(2)-P(1)-Eu(1)	25.94(17)
C(7)-P(1)-Eu(1)	107.1(2)
C(13)-P(1)-Eu(1)	131.9(2)
C(6)-P(1)-Eu(1)	89.5(2)
O(3)-P(2)-C(26)	109.6(3)
O(3)-P(2)-C(20)	110.6(3)
C(26)-P(2)-C(20)	110.2(3)
O(3)-P(2)-C(19)	112.3(3)
C(26)-P(2)-C(19)	107.7(3)
C(20)-P(2)-C(19)	106.3(3)
O(5)-P(3)-C(44)	109.2(3)
O(5)-P(3)-C(38)	112.6(3)
C(44)-P(3)-C(38)	107.9(3)
O(5)-P(3)-C(37)	112.4(2)
C(44)-P(3)-C(37)	108.3(3)
C(38)-P(3)-C(37)	106.2(3)
O(6)-P(4)-C(57)	110.1(3)
O(6)-P(4)-C(51)	111.6(3)
C(57)-P(4)-C(51)	109.6(3)
O(6)-P(4)-C(50)	112.6(3)
C(57)-P(4)-C(50)	107.0(3)
C(51)-P(4)-C(50)	105.8(3)

O(1)-N(1)-C(5)	119.8(4)
O(1)-N(1)-C(1)	118.7(5)
C(5)-N(1)-C(1)	121.5(5)
O(4)-N(2)-C(36)	119.6(5)
O(4)-N(2)-C(32)	119.1(5)
C(36)-N(2)-C(32)	121.2(5)
N(1)-O(1)-Eu(1)	129.4(3)
P(1)-O(2)-Eu(1)	137.5(3)
P(2)-O(3)-Eu(1)	149.3(3)
N(2)-O(4)-Eu(1)	132.4(3)
P(3)-O(5)-Eu(1)	140.5(2)
P(4)-O(6)-Eu(1)	147.8(3)
O(9)-N(3)-O(8)	121.2(7)
O(9)-N(3)-O(7)	121.8(7)
O(8)-N(3)-O(7)	117.0(5)
O(9)-N(3)-Eu(1)	177.1(5)
O(8)-N(3)-Eu(1)	58.7(3)
O(7)-N(3)-Eu(1)	58.4(3)
N(3)-O(7)-Eu(1)	95.7(4)
N(3)-O(8)-Eu(1)	95.8(4)
N(1)-C(1)-C(2)	118.8(5)
N(1)-C(1)-C(6)	117.7(5)
C(2)-C(1)-C(6)	123.4(5)
C(3)-C(2)-C(1)	120.6(6)
C(2)-C(3)-C(4)	119.6(5)
C(3)-C(4)-C(5)	120.1(5)
N(1)-C(5)-C(4)	119.4(5)
N(1)-C(5)-C(19)	117.9(5)
C(4)-C(5)-C(19)	122.6(5)
C(1)-C(6)-P(1)	107.5(4)
C(8)-C(7)-C(12)	120.1(6)
C(8)-C(7)-P(1)	123.8(5)
C(12)-C(7)-P(1)	116.0(5)
C(7)-C(8)-C(9)	119.3(7)
C(10)-C(9)-C(8)	120.1(8)
C(11)-C(10)-C(9)	120.4(7)

C(12)-C(11)-C(10)	120.0(7)
C(11)-C(12)-C(7)	120.0(7)
C(14)-C(13)-C(18)	121.2(7)
C(14)-C(13)-P(1)	118.8(5)
C(18)-C(13)-P(1)	119.9(7)
C(13)-C(14)-C(15)	119.4(9)
C(16)-C(15)-C(14)	120.9(11)
C(15)-C(16)-C(17)	120.0(9)
C(16)-C(17)-C(18)	121.9(9)
C(13)-C(18)-C(17)	116.5(10)
C(5)-C(19)-P(2)	111.3(4)
C(25)-C(20)-C(21)	119.9(6)
C(25)-C(20)-P(2)	122.3(5)
C(21)-C(20)-P(2)	117.8(5)
C(22)-C(21)-C(20)	119.7(7)
C(21)-C(22)-C(23)	120.1(7)
C(22)-C(23)-C(24)	120.0(7)
C(25)-C(24)-C(23)	120.1(7)
C(24)-C(25)-C(20)	120.2(7)
C(31)-C(26)-C(27)	120.5(6)
C(31)-C(26)-P(2)	121.6(5)
C(27)-C(26)-P(2)	117.9(5)
C(28)-C(27)-C(26)	118.8(7)
C(29)-C(28)-C(27)	120.0(8)
C(30)-C(29)-C(28)	121.4(7)
C(29)-C(30)-C(31)	119.9(8)
C(26)-C(31)-C(30)	119.5(7)
N(2)-C(32)-C(33)	119.8(6)
N(2)-C(32)-C(37)	117.7(5)
C(33)-C(32)-C(37)	122.6(5)
C(32)-C(33)-C(34)	119.9(6)
C(35)-C(34)-C(33)	119.1(6)
C(34)-C(35)-C(36)	120.6(6)
N(2)-C(36)-C(35)	119.4(5)
N(2)-C(36)-C(50)	118.5(5)
C(35)-C(36)-C(50)	122.1(5)

C(32)-C(37)-P(3)	111.7(4)
C(39)-C(38)-C(43)	119.4(6)
C(39)-C(38)-P(3)	123.1(5)
C(43)-C(38)-P(3)	117.5(5)
C(38)-C(39)-C(40)	120.1(7)
C(41)-C(40)-C(39)	120.9(7)
C(40)-C(41)-C(42)	119.8(6)
C(41)-C(42)-C(43)	119.5(6)
C(38)-C(43)-C(42)	120.3(6)
C(45)-C(44)-C(49)	120.2(6)
C(45)-C(44)-P(3)	118.7(5)
C(49)-C(44)-P(3)	121.1(5)
C(44)-C(45)-C(46)	119.5(7)
C(47)-C(46)-C(45)	120.5(7)
C(46)-C(47)-C(48)	120.4(6)
C(47)-C(48)-C(49)	120.0(7)
C(48)-C(49)-C(44)	119.5(6)
C(36)-C(50)-P(4)	110.0(4)
C(52)-C(51)-C(56)	120.2(7)
C(52)-C(51)-P(4)	121.5(6)
C(56)-C(51)-P(4)	118.4(6)
C(51)-C(52)-C(53)	119.7(9)
C(54)-C(53)-C(52)	119.5(9)
C(55)-C(54)-C(53)	121.0(8)
C(54)-C(55)-C(56)	120.9(9)
C(51)-C(56)-C(55)	118.5(9)
C(62)-C(57)-C(58)	119.8(6)
C(62)-C(57)-P(4)	121.7(5)
C(58)-C(57)-P(4)	118.5(5)
C(59)-C(58)-C(57)	120.0(7)
C(60)-C(59)-C(58)	119.6(7)
C(59)-C(60)-C(61)	120.8(7)
C(62)-C(61)-C(60)	120.1(8)
C(61)-C(62)-C(57)	119.8(7)
O(10)-N(4)-O(11)	115.1(9)
O(10)-N(4)-O(12)	124.0(9)

O(11)-N(4)-O(12)	120.2(8)
O(14)-N(5)-O(13)	119.2(7)
O(14)-N(5)-O(15)	121.6(6)
O(13)-N(5)-O(15)	119.2(7)
Cl(1B)-C(63)-Cl(2)	117.2(13)
Cl(1)-C(63)-Cl(2)	111.9(10)
Cl(1B)-C(63)-Cl(3)	120.6(13)
Cl(1)-C(63)-Cl(3)	99.0(9)
Cl(2)-C(63)-Cl(3)	110.6(10)
Cl(4)-C(64)-Cl(6)	108.8(8)
Cl(4)-C(64)-Cl(5)	104.1(7)
Cl(6)-C(64)-Cl(5)	105.0(7)
Cl(7)-C(65)-Cl(8)	114.7(7)
Cl(7)-C(65)-Cl(9)	109.9(7)
Cl(8)-C(65)-Cl(9)	111.0(7)
C(66)-Cl(11)-Cl(10)	43.4(11)
Cl(11)-C(66)-Cl(10)	97.2(17)
Cl(11)-C(66)-Cl(12)	145(2)
Cl(10)-C(66)-Cl(12)	111.4(13)
C(66)-Cl(10)-Cl(11)	39.4(10)
C(66)-Cl(10)-Cl(12)	34.9(8)
Cl(11)-Cl(10)-Cl(12)	72.9(10)
C(66)-Cl(12)-Cl(10)	33.8(8)
Cl(1A)-C(66A)-Cl(2A)	98.4(16)
Cl(1A)-C(66A)-Cl(11)	108.0(18)
Cl(2A)-C(66A)-Cl(11)	113.1(19)

Table S16. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Eu}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Eu 2:1**.
 The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^{*2} U^{11} + \dots + 2 h k a^{*} b^{*} U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Eu(1)	14(1)	14(1)	14(1)	2(1)	-2(1)	0(1)
P(1)	16(1)	25(1)	19(1)	8(1)	-7(1)	-4(1)
P(2)	16(1)	13(1)	19(1)	0(1)	-6(1)	-1(1)
P(3)	12(1)	17(1)	18(1)	0(1)	-1(1)	-1(1)
P(4)	21(1)	19(1)	16(1)	4(1)	-4(1)	0(1)
N(1)	14(2)	14(2)	18(2)	4(2)	-5(2)	-1(2)
N(2)	16(2)	17(2)	17(2)	0(2)	-3(2)	-3(2)
O(1)	19(2)	17(2)	14(2)	2(1)	-4(1)	-2(1)
O(2)	16(2)	20(2)	22(2)	4(2)	-3(2)	-1(2)
O(3)	24(2)	18(2)	25(2)	0(2)	-13(2)	-1(2)
O(4)	15(2)	19(2)	15(2)	1(1)	-2(1)	-1(1)
O(5)	14(2)	16(2)	25(2)	0(2)	-1(2)	0(1)
O(6)	30(2)	18(2)	21(2)	4(2)	-10(2)	1(2)
N(3)	52(4)	20(2)	17(2)	1(2)	-16(2)	0(2)
O(7)	38(3)	27(2)	22(2)	4(2)	-7(2)	-12(2)
O(8)	35(2)	21(2)	21(2)	-1(2)	-10(2)	6(2)
O(9)	101(5)	17(2)	30(3)	0(2)	-15(3)	-13(3)
C(1)	19(3)	16(2)	21(3)	4(2)	-8(2)	-2(2)
C(2)	20(3)	21(3)	24(3)	7(2)	-4(2)	-4(2)
C(3)	20(3)	24(3)	19(3)	4(2)	-3(2)	0(2)
C(4)	21(3)	23(3)	18(3)	0(2)	-6(2)	1(2)
C(5)	15(2)	11(2)	19(2)	1(2)	-4(2)	1(2)
C(6)	25(3)	22(3)	26(3)	6(2)	-12(2)	-9(2)
C(7)	20(3)	36(3)	24(3)	9(2)	-10(2)	-8(2)
C(8)	32(3)	36(4)	30(3)	7(3)	-12(3)	-9(3)
C(9)	38(4)	48(4)	33(4)	4(3)	-14(3)	-18(3)
C(10)	29(3)	64(5)	27(3)	17(3)	-15(3)	-17(3)
C(11)	26(3)	47(4)	30(3)	14(3)	-13(3)	-9(3)
C(12)	24(3)	41(4)	27(3)	11(3)	-11(2)	-7(3)
C(13)	13(2)	42(4)	22(3)	13(3)	-3(2)	0(2)

C(14)	28(3)	55(5)	26(3)	-5(3)	1(3)	4(3)
C(15)	55(6)	87(8)	27(4)	-11(4)	4(4)	16(5)
C(16)	48(5)	81(8)	33(4)	13(5)	12(4)	28(5)
C(17)	15(3)	90(8)	58(6)	56(6)	5(3)	11(4)
C(18)	14(3)	62(5)	47(4)	34(4)	-9(3)	-5(3)
C(19)	15(2)	14(2)	21(2)	0(2)	-7(2)	1(2)
C(20)	19(3)	17(2)	26(3)	-2(2)	-4(2)	-2(2)
C(21)	27(3)	21(3)	29(3)	5(2)	-3(2)	-3(2)
C(22)	27(3)	29(3)	49(4)	10(3)	4(3)	-5(3)
C(23)	20(3)	32(4)	67(5)	3(4)	-3(3)	-8(3)
C(24)	24(3)	34(4)	56(5)	-3(3)	-16(3)	-2(3)
C(25)	24(3)	24(3)	37(3)	0(3)	-11(3)	-2(2)
C(26)	22(3)	16(2)	24(3)	-5(2)	-4(2)	-2(2)
C(27)	20(3)	21(3)	39(4)	-5(2)	-2(2)	-1(2)
C(28)	33(4)	25(3)	56(5)	-13(3)	12(3)	-2(3)
C(29)	63(5)	26(3)	39(4)	-13(3)	18(4)	-5(3)
C(30)	70(6)	28(3)	26(3)	-5(3)	-1(3)	-5(4)
C(31)	45(4)	23(3)	24(3)	-5(2)	-7(3)	0(3)
C(32)	16(2)	17(2)	27(3)	-3(2)	-4(2)	-3(2)
C(33)	22(3)	29(3)	32(3)	-11(3)	-3(2)	-1(2)
C(34)	32(3)	41(4)	24(3)	-15(3)	-4(3)	0(3)
C(35)	27(3)	35(3)	21(3)	-8(2)	-6(2)	-4(3)
C(36)	18(2)	20(3)	19(3)	0(2)	-3(2)	-1(2)
C(37)	16(2)	14(2)	24(3)	-2(2)	-3(2)	1(2)
C(38)	12(2)	25(3)	17(2)	2(2)	0(2)	-2(2)
C(39)	37(4)	25(3)	38(4)	10(3)	-19(3)	-9(3)
C(40)	44(4)	30(3)	41(4)	13(3)	-19(3)	-7(3)
C(41)	20(3)	41(4)	26(3)	6(3)	-5(2)	0(3)
C(42)	19(3)	37(3)	31(3)	-2(3)	-8(2)	-3(2)
C(43)	18(3)	26(3)	29(3)	-2(2)	-7(2)	0(2)
C(44)	13(2)	27(3)	19(3)	0(2)	-3(2)	-2(2)
C(45)	22(3)	34(3)	31(3)	8(3)	1(3)	2(3)
C(46)	30(4)	47(4)	39(4)	21(3)	2(3)	-2(3)
C(47)	23(3)	57(5)	25(3)	4(3)	4(2)	-6(3)
C(48)	20(3)	46(4)	27(3)	-13(3)	3(2)	-3(3)
C(49)	23(3)	29(3)	25(3)	-5(2)	0(2)	-2(2)

C(50)	20(3)	22(3)	20(3)	2(2)	-6(2)	-3(2)
C(51)	22(3)	28(3)	27(3)	10(2)	-1(2)	3(2)
C(52)	30(3)	46(4)	34(4)	13(3)	-11(3)	4(3)
C(53)	33(4)	61(6)	54(5)	23(4)	-16(4)	9(4)
C(54)	32(4)	57(6)	76(7)	14(5)	-11(4)	16(4)
C(55)	38(4)	38(4)	67(6)	0(4)	12(4)	11(3)
C(56)	30(3)	31(3)	38(4)	3(3)	5(3)	1(3)
C(57)	24(3)	25(3)	19(3)	4(2)	-2(2)	-3(2)
C(58)	22(3)	31(3)	28(3)	6(2)	-5(2)	-2(2)
C(59)	27(3)	43(4)	40(4)	3(3)	0(3)	-10(3)
C(60)	46(5)	55(5)	28(4)	6(3)	9(3)	-18(4)
C(61)	57(5)	67(6)	20(3)	-3(3)	1(3)	-28(5)
C(62)	41(4)	53(5)	21(3)	-1(3)	-3(3)	-23(4)
N(4)	30(3)	39(4)	53(4)	22(3)	4(3)	-6(3)
O(10)	54(4)	83(6)	76(5)	10(4)	-24(4)	-1(4)
O(11)	117(8)	90(7)	56(5)	-20(5)	-37(5)	-9(6)
O(12)	40(3)	42(3)	49(3)	23(3)	-26(3)	-16(2)
N(5)	19(2)	27(3)	48(4)	-9(3)	-5(2)	3(2)
O(13)	52(4)	32(3)	64(4)	5(3)	-24(3)	-11(3)
O(14)	38(3)	34(3)	33(3)	-8(2)	-3(2)	0(2)
O(15)	30(3)	42(3)	44(3)	-7(2)	-13(2)	0(2)

**X-ray Data Collection, Structure Solution and Refinement for [Nd(L)₂(NO₃)][2(NO₃)], Nd
2:1.**

A pale blue crystal of approximate dimensions 0.512 x 0.327 x 0.246 mm was mounted in a quartz capillary tube containing a small amount of its original mother liquor which was sealed using sealing wax to provide a solvent saturated environment. The crystal-containing capillary was then secured with mounting putty in a MiTeGen Reusable Base™ before being transferred

to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at 80 K (20 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The centrosymmetric triclinic space group *P-1* was assigned and later determined to be correct.

The structure was solved by direct methods and refined on F² by full-matrix least-squares techniques. There were four disordered chloroform molecules of solvation present, only some of which could be satisfactorily modeled using multiple components with partial site-occupancy-factors. Hydrogen atoms were included using a riding model except in the case of solvent molecules for which they were not included.

Table S17. Crystal data and structure refinement for [Nd(L)₂(NO₃)][2(NO₃)], **Nd 2:1**.

Empirical formula	[C ₆₂ H ₅₄ N ₂ NdO ₆ P ₄ (NO ₃)][2(NO ₃)]•4(CHCl ₄)		
Formula weight	1854.69		
Temperature	80(2) K		
Wavelength	0.56086 Å		
Crystal system	Triclinic		
Space group	<i>P-1</i>		
Unit cell dimensions	a = 13.2192(6) Å	α= 83.252(2)°.	
	b = 15.2343(8) Å	β= 77.4165(19)°.	
	c = 19.8616(10) Å	γ = 82.345(2)°.	
Volume	3852.7(3) Å ³		
Z	2		
Density (calculated)	1.599 mg/m ³		
Absorption coefficient	0.650 mm ⁻¹		

F(000)	1866
Crystal size	0.512 x 0.327 x 0.246 mm ³
Theta range for data collection	1.895 to 22.060°.
Index ranges	-17 ≤ h ≤ 17, -20 ≤ k ≤ 20, -26 ≤ l ≤ 26
Reflections collected	116391
Independent reflections	19365 [R(int) = 0.0334]
Completeness to theta = 19.665°	99.9 %
Absorption correction	Numerical
Max. and min. transmission	0.9426 and 0.6951
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	19365 / 0 / 868
Goodness-of-fit on F ²	1.031
Final R indices [I>2sigma(I)]	R1 = 0.0832, wR2 = 0.2135
R indices (all data)	R1 = 0.0897, wR2 = 0.2199
Largest diff. peak and hole	6.319 and -4.344 e.Å ⁻³

Table S18. Atomic coordinates (x 10⁴) and equivalent isotropic displacement parameters (Å² x 10³) for [Nd(L)₂(NO₃)][2(NO₃)], Nd 2:1. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Nd(1)	4824(1)	7851(1)	2611(1)	13(1)
P(1)	2471(1)	6938(1)	3522(1)	18(1)
P(2)	6041(1)	8021(1)	4105(1)	14(1)
P(3)	7250(1)	6497(1)	1915(1)	14(1)
P(4)	3593(1)	8331(1)	1070(1)	18(1)
N(1)	4726(3)	6284(3)	4039(2)	14(1)
N(2)	5002(3)	6387(3)	1356(2)	15(1)
O(1)	5130(3)	6482(2)	3372(2)	15(1)
O(2)	3167(3)	7672(3)	3311(2)	17(1)
O(3)	5530(3)	8281(3)	3499(2)	20(1)
O(4)	4620(3)	6606(3)	2000(2)	15(1)
O(5)	6568(3)	7346(3)	2082(2)	18(1)

O(6)	3989(3)	8486(3)	1693(2)	21(1)
N(3)	4708(5)	9807(3)	2562(3)	28(1)
O(7)	5497(4)	9351(3)	2219(2)	28(1)
O(8)	3967(4)	9395(3)	2889(2)	25(1)
O(9)	4681(6)	10603(3)	2569(3)	47(2)
C(1)	3794(4)	5916(3)	4212(3)	16(1)
C(2)	3395(4)	5668(4)	4902(3)	20(1)
C(3)	3914(4)	5807(4)	5411(3)	20(1)
C(4)	4839(4)	6186(4)	5221(3)	18(1)
C(5)	5246(4)	6418(3)	4530(3)	14(1)
C(6)	3228(4)	5872(4)	3647(3)	21(1)
C(7)	1713(4)	6884(4)	2890(3)	23(1)
C(8)	1628(5)	6105(4)	2614(3)	28(1)
C(9)	1092(6)	6146(5)	2078(4)	33(1)
C(10)	641(5)	6955(5)	1829(3)	33(2)
C(11)	693(5)	7729(5)	2114(3)	30(1)
C(12)	1241(5)	7699(4)	2638(3)	26(1)
C(13)	1625(4)	7125(4)	4340(3)	24(1)
C(14)	1773(5)	7816(5)	4691(3)	34(2)
C(15)	1176(7)	7929(7)	5356(4)	53(2)
C(16)	447(7)	7382(8)	5659(4)	59(3)
C(17)	293(5)	6691(7)	5316(5)	57(3)
C(18)	876(5)	6547(5)	4641(4)	37(2)
C(19)	6251(4)	6825(3)	4294(3)	15(1)
C(20)	7301(4)	8413(4)	3926(3)	18(1)
C(21)	7587(5)	8951(4)	3313(3)	24(1)
C(22)	8541(5)	9298(5)	3168(4)	34(2)
C(23)	9202(5)	9112(5)	3637(4)	36(2)
C(24)	8927(5)	8561(5)	4241(4)	32(1)
C(25)	7983(5)	8208(4)	4387(3)	24(1)
C(26)	5234(4)	8468(4)	4861(3)	18(1)
C(27)	4342(5)	9041(4)	4778(3)	24(1)
C(28)	3691(5)	9392(4)	5360(4)	34(2)
C(29)	3934(6)	9162(5)	6007(4)	38(2)
C(30)	4815(7)	8602(5)	6087(4)	36(2)
C(31)	5473(5)	8246(4)	5512(3)	26(1)

C(32)	5951(4)	5898(4)	1211(3)	18(1)
C(33)	6349(5)	5663(4)	545(3)	23(1)
C(34)	5771(5)	5907(5)	34(3)	27(1)
C(35)	4802(5)	6375(4)	197(3)	23(1)
C(36)	4416(4)	6620(4)	863(3)	17(1)
C(37)	6515(4)	5631(3)	1788(3)	17(1)
C(38)	7930(4)	6056(4)	2589(3)	18(1)
C(39)	8011(6)	5154(4)	2828(4)	31(1)
C(40)	8568(6)	4854(5)	3347(4)	36(2)
C(41)	9023(5)	5439(5)	3635(3)	28(1)
C(42)	8940(5)	6336(5)	3405(3)	26(1)
C(43)	8395(4)	6640(4)	2881(3)	22(1)
C(44)	8200(4)	6702(4)	1139(3)	18(1)
C(45)	8180(5)	7553(5)	804(3)	29(1)
C(46)	8938(6)	7734(5)	211(4)	39(2)
C(47)	9706(5)	7076(5)	-36(3)	34(2)
C(48)	9726(5)	6229(5)	294(3)	28(1)
C(49)	8976(4)	6034(4)	889(3)	23(1)
C(50)	3395(4)	7177(4)	1055(3)	18(1)
C(51)	2353(5)	8967(4)	1044(3)	25(1)
C(52)	1759(5)	8831(5)	575(4)	33(2)
C(53)	814(6)	9352(6)	568(4)	45(2)
C(54)	486(7)	10005(7)	1012(5)	51(2)
C(55)	1051(6)	10130(6)	1482(5)	44(2)
C(56)	1995(5)	9603(5)	1509(4)	31(1)
C(57)	4517(5)	8618(4)	297(3)	22(1)
C(58)	5464(5)	8880(4)	359(3)	27(1)
C(59)	6204(6)	9084(5)	-231(4)	36(2)
C(60)	5982(7)	9047(6)	-876(4)	44(2)
C(61)	5052(7)	8796(6)	-938(4)	44(2)
C(62)	4312(6)	8577(5)	-353(3)	34(2)
N(4)	2200(5)	3940(4)	4228(4)	37(1)
O(10)	3008(5)	3468(5)	4144(4)	57(2)
O(11)	1817(7)	4161(5)	4824(4)	67(2)
O(12)	1765(4)	4267(4)	3722(3)	37(1)
N(5)	7680(4)	3557(4)	442(3)	27(1)

O(13)	7391(5)	2788(4)	540(3)	40(1)
O(14)	7677(4)	3987(3)	-129(2)	31(1)
O(15)	7971(4)	3873(3)	917(3)	33(1)
C(63)	1241(10)	13367(9)	1607(7)	72(3)
Cl(2)	1285(2)	13602(2)	2462(1)	58(1)
Cl(1)	2345(5)	12545(4)	1311(3)	68(1)
Cl(3)	1481(2)	14308(2)	1003(1)	42(1)
Cl(1B)	1871(8)	12486(6)	1335(5)	76(2)
Cl(3A)	59(17)	13941(14)	1552(11)	94(5)
C(64)	2090(9)	789(8)	3491(6)	61(2)
Cl(4)	1558(3)	152(2)	4253(2)	76(1)
Cl(5)	1096(3)	1017(2)	2986(2)	74(1)
Cl(6)	2354(2)	1824(2)	3695(1)	51(1)
C(65)	4145(8)	13691(7)	2544(5)	51(2)
Cl(7)	4455(4)	12707(3)	2161(3)	106(1)
Cl(8)	5126(2)	13927(2)	2946(1)	60(1)
Cl(9)	3863(2)	14571(1)	1928(1)	40(1)
Cl(11)	-2836(10)	11291(9)	2111(7)	262(5)
C(66)	-2152(18)	12272(16)	2102(12)	63(5)
Cl(10)	-748(7)	11943(6)	1888(5)	119(2)
Cl(12)	-2317(5)	12585(4)	2917(3)	74(2)
C(66A)	-2499(10)	10822(9)	1584(7)	27(2)
Cl(1A)	-2323(14)	9944(13)	1283(10)	159(6)
Cl(2A)	-1730(7)	11419(6)	1277(5)	99(2)

Table S19. Bond lengths [Å] and angles [°] for $[\text{Nd}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Nd 2:1**.

Nd(1)-O(2)	2.356(4)
Nd(1)-O(3)	2.359(4)
Nd(1)-O(6)	2.378(4)
Nd(1)-O(5)	2.381(4)
Nd(1)-O(4)	2.440(4)
Nd(1)-O(1)	2.470(4)

Nd(1)-O(7)	2.538(4)
Nd(1)-O(8)	2.539(4)
Nd(1)-N(3)	2.956(5)
P(1)-O(2)	1.506(4)
P(1)-C(7)	1.783(6)
P(1)-C(13)	1.790(6)
P(1)-C(6)	1.813(6)
P(2)-O(3)	1.493(4)
P(2)-C(26)	1.790(6)
P(2)-C(20)	1.793(6)
P(2)-C(19)	1.813(5)
P(3)-O(5)	1.502(4)
P(3)-C(44)	1.788(6)
P(3)-C(38)	1.789(6)
P(3)-C(37)	1.810(5)
P(4)-O(6)	1.497(4)
P(4)-C(57)	1.791(6)
P(4)-C(51)	1.797(6)
P(4)-C(50)	1.817(6)
N(1)-O(1)	1.330(6)
N(1)-C(5)	1.357(7)
N(1)-C(1)	1.380(7)
N(2)-O(4)	1.334(6)
N(2)-C(36)	1.363(7)
N(2)-C(32)	1.363(7)
N(3)-O(9)	1.210(7)
N(3)-O(8)	1.252(8)
N(3)-O(7)	1.282(8)
C(1)-C(2)	1.385(8)
C(1)-C(6)	1.490(7)
C(2)-C(3)	1.388(8)
C(3)-C(4)	1.380(8)
C(4)-C(5)	1.384(7)
C(5)-C(19)	1.500(7)
C(7)-C(8)	1.392(9)
C(7)-C(12)	1.402(8)

C(8)-C(9)	1.392(9)
C(9)-C(10)	1.382(10)
C(10)-C(11)	1.382(11)
C(11)-C(12)	1.386(8)
C(13)-C(14)	1.385(10)
C(13)-C(18)	1.402(9)
C(14)-C(15)	1.400(10)
C(15)-C(16)	1.353(16)
C(16)-C(17)	1.378(17)
C(17)-C(18)	1.418(12)
C(20)-C(21)	1.395(8)
C(20)-C(25)	1.400(8)
C(21)-C(22)	1.392(9)
C(22)-C(23)	1.392(11)
C(23)-C(24)	1.389(11)
C(24)-C(25)	1.384(9)
C(26)-C(31)	1.390(8)
C(26)-C(27)	1.399(8)
C(27)-C(28)	1.397(9)
C(28)-C(29)	1.386(12)
C(29)-C(30)	1.376(11)
C(30)-C(31)	1.394(9)
C(32)-C(33)	1.384(8)
C(32)-C(37)	1.488(8)
C(33)-C(34)	1.387(9)
C(34)-C(35)	1.372(9)
C(35)-C(36)	1.387(8)
C(36)-C(50)	1.494(7)
C(38)-C(43)	1.389(8)
C(38)-C(39)	1.398(8)
C(39)-C(40)	1.395(9)
C(40)-C(41)	1.376(10)
C(41)-C(42)	1.387(10)
C(42)-C(43)	1.394(8)
C(44)-C(45)	1.386(8)
C(44)-C(49)	1.398(8)

C(45)-C(46)	1.396(9)
C(46)-C(47)	1.380(11)
C(47)-C(48)	1.377(10)
C(48)-C(49)	1.398(8)
C(51)-C(56)	1.382(9)
C(51)-C(52)	1.392(9)
C(52)-C(53)	1.390(9)
C(53)-C(54)	1.371(14)
C(54)-C(55)	1.359(13)
C(55)-C(56)	1.399(10)
C(57)-C(62)	1.386(9)
C(57)-C(58)	1.397(9)
C(58)-C(59)	1.387(9)
C(59)-C(60)	1.384(12)
C(60)-C(61)	1.370(13)
C(61)-C(62)	1.386(10)
N(4)-O(10)	1.197(9)
N(4)-O(11)	1.249(10)
N(4)-O(12)	1.282(8)
N(5)-O(14)	1.242(7)
N(5)-O(15)	1.258(7)
N(5)-O(13)	1.260(8)
C(63)-Cl(1B)	1.568(16)
C(63)-Cl(3A)	1.71(2)
C(63)-Cl(3)	1.774(13)
C(63)-Cl(2)	1.790(13)
C(63)-Cl(1)	1.831(14)
C(64)-Cl(4)	1.761(12)
C(64)-Cl(6)	1.769(12)
C(64)-Cl(5)	1.794(12)
C(65)-Cl(7)	1.721(11)
C(65)-Cl(8)	1.758(10)
C(65)-Cl(9)	1.764(10)
Cl(11)-C(66A)	1.301(17)
Cl(11)-C(66)	1.84(3)
Cl(11)-Cl(2A)	1.967(15)

C(66)-Cl(12)	1.70(2)
C(66)-Cl(10)	1.83(2)
C(66A)-Cl(2A)	1.443(16)
C(66A)-Cl(1A)	1.50(2)
Cl(1A)-Cl(2A)	2.47(2)

O(2)-Nd(1)-O(3)	95.19(14)
O(2)-Nd(1)-O(6)	88.66(14)
O(3)-Nd(1)-O(6)	140.43(14)
O(2)-Nd(1)-O(5)	154.26(13)
O(3)-Nd(1)-O(5)	86.62(14)
O(6)-Nd(1)-O(5)	106.21(14)
O(2)-Nd(1)-O(4)	87.90(13)
O(3)-Nd(1)-O(4)	145.22(13)
O(6)-Nd(1)-O(4)	74.14(13)
O(5)-Nd(1)-O(4)	76.56(13)
O(2)-Nd(1)-O(1)	76.19(13)
O(3)-Nd(1)-O(1)	74.00(13)
O(6)-Nd(1)-O(1)	144.28(13)
O(5)-Nd(1)-O(1)	79.70(13)
O(4)-Nd(1)-O(1)	73.17(12)
O(2)-Nd(1)-O(7)	123.65(15)
O(3)-Nd(1)-O(7)	71.37(14)
O(6)-Nd(1)-O(7)	73.83(15)
O(5)-Nd(1)-O(7)	81.28(15)
O(4)-Nd(1)-O(7)	133.67(13)
O(1)-Nd(1)-O(7)	141.18(14)
O(2)-Nd(1)-O(8)	73.24(14)
O(3)-Nd(1)-O(8)	71.89(14)
O(6)-Nd(1)-O(8)	71.65(14)
O(5)-Nd(1)-O(8)	130.99(14)
O(4)-Nd(1)-O(8)	141.00(14)
O(1)-Nd(1)-O(8)	131.14(13)
O(7)-Nd(1)-O(8)	50.44(16)
O(2)-Nd(1)-N(3)	98.14(15)
O(3)-Nd(1)-N(3)	68.75(14)

O(6)-Nd(1)-N(3)	71.71(14)
O(5)-Nd(1)-N(3)	106.37(15)
O(4)-Nd(1)-N(3)	145.12(13)
O(1)-Nd(1)-N(3)	141.66(13)
O(7)-Nd(1)-N(3)	25.57(16)
O(8)-Nd(1)-N(3)	24.90(16)
O(2)-P(1)-C(7)	111.1(3)
O(2)-P(1)-C(13)	109.4(3)
C(7)-P(1)-C(13)	109.7(3)
O(2)-P(1)-C(6)	111.0(2)
C(7)-P(1)-C(6)	109.3(3)
C(13)-P(1)-C(6)	106.2(3)
O(3)-P(2)-C(26)	109.7(3)
O(3)-P(2)-C(20)	110.0(3)
C(26)-P(2)-C(20)	110.6(3)
O(3)-P(2)-C(19)	112.4(2)
C(26)-P(2)-C(19)	107.5(3)
C(20)-P(2)-C(19)	106.5(2)
O(5)-P(3)-C(44)	109.3(2)
O(5)-P(3)-C(38)	112.4(2)
C(44)-P(3)-C(38)	107.9(2)
O(5)-P(3)-C(37)	112.3(2)
C(44)-P(3)-C(37)	108.3(3)
C(38)-P(3)-C(37)	106.4(3)
O(6)-P(4)-C(57)	109.9(3)
O(6)-P(4)-C(51)	111.5(3)
C(57)-P(4)-C(51)	109.6(3)
O(6)-P(4)-C(50)	112.7(2)
C(57)-P(4)-C(50)	106.9(3)
C(51)-P(4)-C(50)	106.0(3)
O(1)-N(1)-C(5)	119.9(4)
O(1)-N(1)-C(1)	118.6(4)
C(5)-N(1)-C(1)	121.5(4)
O(4)-N(2)-C(36)	119.5(4)
O(4)-N(2)-C(32)	119.1(4)
C(36)-N(2)-C(32)	121.3(5)

N(1)-O(1)-Nd(1)	130.0(3)
P(1)-O(2)-Nd(1)	136.9(2)
P(2)-O(3)-Nd(1)	148.9(2)
N(2)-O(4)-Nd(1)	132.7(3)
P(3)-O(5)-Nd(1)	140.2(2)
P(4)-O(6)-Nd(1)	147.0(2)
O(9)-N(3)-O(8)	121.4(6)
O(9)-N(3)-O(7)	121.4(6)
O(8)-N(3)-O(7)	117.2(5)
O(9)-N(3)-Nd(1)	176.9(4)
O(8)-N(3)-Nd(1)	58.6(3)
O(7)-N(3)-Nd(1)	58.7(3)
N(3)-O(7)-Nd(1)	95.7(3)
N(3)-O(8)-Nd(1)	96.5(3)
N(1)-C(1)-C(2)	118.8(5)
N(1)-C(1)-C(6)	117.8(5)
C(2)-C(1)-C(6)	123.2(5)
C(1)-C(2)-C(3)	120.3(5)
C(4)-C(3)-C(2)	119.4(5)
C(3)-C(4)-C(5)	120.2(5)
N(1)-C(5)-C(4)	119.7(5)
N(1)-C(5)-C(19)	117.8(4)
C(4)-C(5)-C(19)	122.5(5)
C(1)-C(6)-P(1)	108.1(4)
C(8)-C(7)-C(12)	119.9(6)
C(8)-C(7)-P(1)	124.0(5)
C(12)-C(7)-P(1)	115.9(5)
C(9)-C(8)-C(7)	119.5(6)
C(10)-C(9)-C(8)	120.0(7)
C(9)-C(10)-C(11)	121.0(6)
C(10)-C(11)-C(12)	119.6(6)
C(11)-C(12)-C(7)	120.0(6)
C(14)-C(13)-C(18)	120.7(7)
C(14)-C(13)-P(1)	118.6(5)
C(18)-C(13)-P(1)	120.6(6)
C(13)-C(14)-C(15)	119.5(8)

C(16)-C(15)-C(14)	121.2(10)
C(15)-C(16)-C(17)	119.8(8)
C(16)-C(17)-C(18)	121.3(8)
C(13)-C(18)-C(17)	117.5(9)
C(5)-C(19)-P(2)	112.2(3)
C(21)-C(20)-C(25)	120.1(5)
C(21)-C(20)-P(2)	117.9(4)
C(25)-C(20)-P(2)	122.0(4)
C(22)-C(21)-C(20)	119.8(6)
C(21)-C(22)-C(23)	119.9(6)
C(24)-C(23)-C(22)	120.4(6)
C(25)-C(24)-C(23)	120.1(6)
C(24)-C(25)-C(20)	119.8(6)
C(31)-C(26)-C(27)	120.8(6)
C(31)-C(26)-P(2)	121.4(4)
C(27)-C(26)-P(2)	117.8(4)
C(28)-C(27)-C(26)	119.1(6)
C(29)-C(28)-C(27)	119.7(7)
C(30)-C(29)-C(28)	121.1(6)
C(29)-C(30)-C(31)	120.0(7)
C(26)-C(31)-C(30)	119.4(6)
N(2)-C(32)-C(33)	119.4(5)
N(2)-C(32)-C(37)	117.9(5)
C(33)-C(32)-C(37)	122.7(5)
C(32)-C(33)-C(34)	120.0(5)
C(35)-C(34)-C(33)	119.5(6)
C(34)-C(35)-C(36)	120.1(6)
N(2)-C(36)-C(35)	119.5(5)
N(2)-C(36)-C(50)	118.3(5)
C(35)-C(36)-C(50)	122.1(5)
C(32)-C(37)-P(3)	111.9(4)
C(43)-C(38)-C(39)	119.3(5)
C(43)-C(38)-P(3)	118.0(4)
C(39)-C(38)-P(3)	122.7(5)
C(40)-C(39)-C(38)	119.6(6)
C(41)-C(40)-C(39)	120.8(6)

C(40)-C(41)-C(42)	120.1(6)
C(41)-C(42)-C(43)	119.7(6)
C(38)-C(43)-C(42)	120.6(6)
C(45)-C(44)-C(49)	120.3(5)
C(45)-C(44)-P(3)	118.5(4)
C(49)-C(44)-P(3)	121.2(4)
C(44)-C(45)-C(46)	119.3(6)
C(47)-C(46)-C(45)	120.5(7)
C(48)-C(47)-C(46)	120.4(6)
C(47)-C(48)-C(49)	120.0(6)
C(44)-C(49)-C(48)	119.5(6)
C(36)-C(50)-P(4)	110.6(4)
C(56)-C(51)-C(52)	120.1(6)
C(56)-C(51)-P(4)	118.1(5)
C(52)-C(51)-P(4)	121.8(5)
C(53)-C(52)-C(51)	119.5(7)
C(54)-C(53)-C(52)	119.8(8)
C(55)-C(54)-C(53)	121.1(7)
C(54)-C(55)-C(56)	120.2(8)
C(51)-C(56)-C(55)	119.2(7)
C(62)-C(57)-C(58)	120.0(6)
C(62)-C(57)-P(4)	121.3(5)
C(58)-C(57)-P(4)	118.7(5)
C(59)-C(58)-C(57)	119.9(6)
C(60)-C(59)-C(58)	119.2(7)
C(61)-C(60)-C(59)	121.0(7)
C(60)-C(61)-C(62)	120.3(7)
C(61)-C(62)-C(57)	119.5(7)
O(10)-N(4)-O(11)	117.8(8)
O(10)-N(4)-O(12)	122.2(7)
O(11)-N(4)-O(12)	119.9(7)
O(14)-N(5)-O(15)	121.2(6)
O(14)-N(5)-O(13)	118.8(6)
O(15)-N(5)-O(13)	120.0(6)
Cl(1B)-C(63)-Cl(3A)	135.7(12)
Cl(1B)-C(63)-Cl(2)	117.8(9)

Cl(3A)-C(63)-Cl(2)	98.2(10)
Cl(3)-C(63)-Cl(2)	111.0(7)
Cl(3)-C(63)-Cl(1)	103.3(7)
Cl(2)-C(63)-Cl(1)	108.6(7)
Cl(4)-C(64)-Cl(6)	110.0(6)
Cl(4)-C(64)-Cl(5)	106.5(6)
Cl(6)-C(64)-Cl(5)	107.3(6)
Cl(7)-C(65)-Cl(8)	112.8(6)
Cl(7)-C(65)-Cl(9)	110.0(6)
Cl(8)-C(65)-Cl(9)	110.7(5)
C(66A)-Cl(11)-Cl(2A)	47.2(8)
Cl(12)-C(66)-Cl(10)	102.0(12)
Cl(12)-C(66)-Cl(11)	110.6(13)
Cl(10)-C(66)-Cl(11)	108.7(13)
Cl(11)-C(66A)-Cl(2A)	91.5(11)
Cl(11)-C(66A)-Cl(1A)	149.9(14)
Cl(2A)-C(66A)-Cl(1A)	114.6(12)
C(66A)-Cl(1A)-Cl(2A)	32.0(7)
C(66A)-Cl(2A)-Cl(11)	41.4(7)
C(66A)-Cl(2A)-Cl(1A)	33.4(7)
Cl(11)-Cl(2A)-Cl(1A)	74.0(7)

Table S20. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Nd}(\text{L})_2(\text{NO}_3)][2(\text{NO}_3)]$, **Nd 2:1**. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^*{}^2 U^{11} + \dots + 2 h k a^* b^* U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Nd(1)	12(1)	12(1)	12(1)	1(1)	-1(1)	1(1)
P(1)	13(1)	22(1)	18(1)	6(1)	-6(1)	-3(1)
P(2)	15(1)	13(1)	16(1)	0(1)	-5(1)	-1(1)
P(3)	10(1)	15(1)	16(1)	-1(1)	-1(1)	0(1)
P(4)	20(1)	18(1)	14(1)	3(1)	-4(1)	2(1)

N(1)	14(2)	12(2)	16(2)	2(2)	-3(2)	1(2)
N(2)	15(2)	14(2)	14(2)	-1(2)	-2(2)	-3(2)
O(1)	17(2)	16(2)	12(2)	1(1)	-2(1)	-1(1)
O(2)	12(2)	18(2)	20(2)	2(1)	-2(1)	0(1)
O(3)	26(2)	15(2)	21(2)	0(1)	-12(2)	-1(2)
O(4)	15(2)	18(2)	12(2)	-2(1)	-1(1)	-1(1)
O(5)	12(2)	16(2)	23(2)	-2(1)	1(1)	0(1)
O(6)	28(2)	18(2)	17(2)	2(1)	-8(2)	1(2)
N(3)	50(3)	20(2)	14(2)	0(2)	-13(2)	-1(2)
O(7)	38(2)	26(2)	22(2)	6(2)	-6(2)	-13(2)
O(8)	33(2)	17(2)	24(2)	-2(2)	-8(2)	5(2)
O(9)	97(5)	18(2)	26(2)	-2(2)	-9(3)	-14(3)
C(1)	15(2)	14(2)	20(2)	3(2)	-7(2)	-1(2)
C(2)	16(2)	17(2)	24(3)	6(2)	-4(2)	-1(2)
C(3)	21(3)	21(3)	16(2)	3(2)	-2(2)	-2(2)
C(4)	20(2)	17(2)	16(2)	0(2)	-5(2)	2(2)
C(5)	13(2)	11(2)	17(2)	-1(2)	-3(2)	1(2)
C(6)	22(3)	18(2)	24(3)	4(2)	-12(2)	-6(2)
C(7)	19(3)	31(3)	19(3)	6(2)	-9(2)	-5(2)
C(8)	28(3)	29(3)	28(3)	4(2)	-9(2)	-7(2)
C(9)	32(3)	40(4)	29(3)	0(3)	-11(3)	-13(3)
C(10)	25(3)	52(4)	24(3)	10(3)	-10(2)	-12(3)
C(11)	22(3)	40(4)	27(3)	9(3)	-12(2)	-3(3)
C(12)	23(3)	30(3)	25(3)	6(2)	-9(2)	-3(2)
C(13)	12(2)	36(3)	20(3)	10(2)	-2(2)	0(2)
C(14)	26(3)	47(4)	23(3)	-5(3)	0(2)	7(3)
C(15)	51(5)	71(6)	27(4)	-8(4)	0(3)	18(4)
C(16)	44(5)	81(7)	29(4)	13(4)	11(3)	28(5)
C(17)	14(3)	79(7)	53(5)	49(5)	7(3)	10(3)
C(18)	14(3)	50(4)	41(4)	29(3)	-7(3)	-4(3)
C(19)	14(2)	14(2)	18(2)	0(2)	-5(2)	1(2)
C(20)	16(2)	15(2)	23(3)	-3(2)	-1(2)	-2(2)
C(21)	23(3)	20(3)	25(3)	2(2)	0(2)	-2(2)
C(22)	26(3)	26(3)	43(4)	7(3)	5(3)	-3(2)
C(23)	17(3)	28(3)	62(5)	0(3)	0(3)	-7(2)
C(24)	22(3)	27(3)	49(4)	-4(3)	-15(3)	-1(2)

C(25)	20(3)	22(3)	32(3)	-1(2)	-7(2)	-2(2)
C(26)	19(2)	15(2)	21(3)	-5(2)	-3(2)	-3(2)
C(27)	20(3)	17(3)	34(3)	-4(2)	-4(2)	0(2)
C(28)	28(3)	21(3)	48(4)	-8(3)	8(3)	-1(2)
C(29)	47(4)	22(3)	36(4)	-10(3)	15(3)	-3(3)
C(30)	56(5)	27(3)	22(3)	-5(2)	-2(3)	-6(3)
C(31)	37(3)	19(3)	22(3)	-2(2)	-6(2)	-1(2)
C(32)	14(2)	16(2)	23(3)	-5(2)	-2(2)	-1(2)
C(33)	20(3)	24(3)	26(3)	-10(2)	-1(2)	0(2)
C(34)	26(3)	34(3)	22(3)	-13(2)	-2(2)	0(2)
C(35)	25(3)	28(3)	18(3)	-7(2)	-5(2)	-5(2)
C(36)	16(2)	19(2)	16(2)	-2(2)	-3(2)	-2(2)
C(37)	13(2)	14(2)	23(3)	-2(2)	-3(2)	1(2)
C(38)	12(2)	23(3)	16(2)	-1(2)	0(2)	0(2)
C(39)	38(4)	23(3)	34(3)	7(3)	-18(3)	-8(3)
C(40)	42(4)	31(3)	38(4)	13(3)	-20(3)	-5(3)
C(41)	18(3)	40(4)	24(3)	4(3)	-5(2)	-1(2)
C(42)	18(3)	34(3)	28(3)	-4(2)	-7(2)	0(2)
C(43)	16(2)	23(3)	27(3)	-4(2)	-5(2)	1(2)
C(44)	11(2)	24(3)	19(2)	-3(2)	-2(2)	-2(2)
C(45)	20(3)	31(3)	28(3)	6(2)	3(2)	2(2)
C(46)	28(3)	43(4)	35(4)	17(3)	4(3)	-3(3)
C(47)	22(3)	55(4)	22(3)	0(3)	4(2)	-8(3)
C(48)	17(3)	38(3)	26(3)	-12(3)	2(2)	-2(2)
C(49)	18(3)	24(3)	23(3)	-6(2)	1(2)	-1(2)
C(50)	16(2)	20(2)	18(2)	-1(2)	-4(2)	-1(2)
C(51)	22(3)	27(3)	21(3)	6(2)	-4(2)	4(2)
C(52)	30(3)	41(4)	26(3)	6(3)	-10(3)	7(3)
C(53)	32(4)	57(5)	43(4)	9(4)	-16(3)	13(3)
C(54)	33(4)	57(5)	57(5)	5(4)	-13(4)	20(4)
C(55)	35(4)	38(4)	50(5)	-6(3)	2(3)	14(3)
C(56)	28(3)	28(3)	30(3)	2(3)	0(2)	5(2)
C(57)	24(3)	22(3)	19(3)	3(2)	-1(2)	-1(2)
C(58)	24(3)	26(3)	28(3)	4(2)	-3(2)	-1(2)
C(59)	29(3)	36(4)	39(4)	5(3)	4(3)	-9(3)
C(60)	48(4)	44(4)	31(4)	1(3)	11(3)	-13(3)

C(61)	63(5)	47(4)	22(3)	-3(3)	2(3)	-20(4)
C(62)	42(4)	42(4)	18(3)	1(3)	-3(3)	-17(3)
N(4)	28(3)	32(3)	43(3)	15(3)	2(2)	-7(2)
O(10)	42(3)	64(4)	63(4)	4(3)	-20(3)	1(3)
O(11)	88(6)	73(5)	47(4)	-19(3)	-23(4)	-8(4)
O(12)	35(3)	37(3)	41(3)	20(2)	-21(2)	-15(2)
N(5)	18(2)	26(3)	35(3)	-8(2)	-2(2)	3(2)
O(13)	44(3)	29(3)	50(3)	4(2)	-16(3)	-8(2)
O(14)	32(2)	31(2)	28(2)	-6(2)	-2(2)	1(2)
O(15)	27(2)	39(3)	35(3)	-7(2)	-11(2)	1(2)

X-ray Data Collection, Structure Solution and Refinement for Nd(**L**)(NO₃)₃, **Nd 1:1.**

A pale blue crystal of approximate dimensions 0.246 x 0.133 x 0.084 mm was mounted in Krytox™ grease on a MiTeGen Reusable Base™ before being transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at room temperature (35 sec/frame scan time for a sphere of diffraction data) using MoK α radiation. The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The non-centrosymmetric monoclinic space group *Cc* was assigned and later determined to be correct.

The structure was solved by direct methods and refined on F² by full-matrix least-squares techniques as a 2-component (3:97) inversion twin. All hydrogen atoms were included using a riding model.

Table S21. Crystal data and structure refinement for Nd(L)(NO₃)₃, **Nd 1:1.**

Empirical formula	C ₃₁ H ₂₇ NNdO ₃ P ₂ (NO ₃) ₃	
Formula weight	853.74	
Temperature	293(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	Cc	
Unit cell dimensions	a = 12.1971(6) Å	α= 90°.
	b = 19.1392(9) Å	β= 91.7723(18)°.
	c = 14.8031(8) Å	γ = 90°.
Volume	3454.0(3) Å ³	
Z	4	
Density (calculated)	1.642 mg/m ³	
Absorption coefficient	1.663 mm ⁻¹	
F(000)	1708	
Crystal size	0.246 x 0.133 x 0.084 mm ³	
Theta range for data collection	3.350 to 28.378°.	
Index ranges	-16 ≤ h ≤ 14, -25 ≤ k ≤ 25, -19 ≤ l ≤ 18	
Reflections collected	28402	
Independent reflections	7120 [R(int) = 0.0581]	
Completeness to theta = 25.242°	98.7 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.7457 and 0.6394	
Refinement method	Full-matrix least-squares on F ²	
Data / restraints / parameters	7120 / 2 / 452	
Goodness-of-fit on F ²	1.122	
Final R indices [I>2sigma(I)]	R1 = 0.0493, wR2 = 0.0732	
R indices (all data)	R1 = 0.0636, wR2 = 0.0762	
Absolute structure parameter	0.027(16)	
Largest diff. peak and hole	1.683 and -2.080 e.Å ⁻³	

Table S22. Atomic coordinates ($x \times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for Nd(L)(NO₃)₃, **Nd 1:1**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Nd(1)	2448(1)	7717(1)	7508(1)	30(1)
P(1)	1436(2)	9537(1)	7304(1)	32(1)
P(2)	214(2)	7019(1)	8766(1)	30(1)
O(1)	2040(5)	8352(2)	8869(3)	36(1)
O(2)	1923(5)	8853(3)	7020(4)	44(2)
O(3)	702(5)	7329(3)	7940(4)	43(1)
O(4)	1290(6)	7537(3)	6101(4)	55(2)
O(5)	2232(8)	6651(4)	6526(6)	65(2)
O(6)	962(9)	6540(5)	5476(6)	102(3)
O(7)	4061(6)	6839(3)	7775(5)	64(2)
O(8)	2998(8)	6968(5)	8858(6)	73(3)
O(9)	4381(7)	6272(3)	9014(5)	70(2)
O(10)	3907(5)	7996(3)	6387(4)	44(2)
O(11)	4228(5)	8392(3)	7734(4)	46(2)
O(12)	5404(7)	8560(4)	6692(5)	77(2)
N(4)	4535(7)	8318(3)	6933(5)	43(2)
N(1)	1149(5)	8613(3)	9237(4)	28(1)
N(2)	1477(10)	6897(5)	6012(7)	59(3)
N(3)	3831(8)	6675(3)	8557(6)	50(2)
C(1)	930(7)	9315(4)	9125(5)	32(2)
C(2)	52(8)	9586(4)	9568(5)	46(2)
C(3)	-598(9)	9185(5)	10093(6)	49(3)
C(4)	-360(8)	8478(4)	10181(6)	40(2)
C(5)	504(7)	8198(4)	9752(5)	31(2)
C(6)	1647(8)	9714(4)	8513(5)	36(2)
C(7)	2115(7)	10247(4)	6759(5)	39(2)
C(8)	2912(9)	10095(6)	6165(7)	62(3)
C(9)	3474(11)	10647(8)	5736(7)	82(4)
C(10)	3182(13)	11328(7)	5911(9)	85(4)
C(11)	2396(12)	11467(6)	6484(8)	73(3)

C(12)	1841(10)	10945(4)	6914(7)	57(3)
C(13)	3(8)	9548(4)	7050(5)	35(2)
C(14)	-439(9)	9034(5)	6487(6)	52(2)
C(15)	-1535(11)	9018(5)	6289(7)	69(3)
C(16)	-2237(9)	9508(6)	6664(8)	64(3)
C(17)	-1814(9)	10025(5)	7203(7)	56(3)
C(18)	-701(8)	10049(5)	7397(6)	45(2)
C(19)	780(7)	7436(3)	9780(5)	33(2)
C(20)	423(7)	6097(3)	8839(5)	31(2)
C(21)	942(10)	5771(5)	9549(7)	58(3)
C(22)	1066(11)	5044(5)	9556(8)	77(4)
C(23)	690(10)	4669(5)	8836(8)	65(3)
C(24)	172(12)	4984(5)	8124(8)	75(4)
C(25)	63(11)	5706(5)	8113(7)	66(3)
C(26)	-1222(7)	7167(4)	8744(5)	37(2)
C(27)	-1945(8)	6762(5)	9232(6)	51(2)
C(28)	-3035(10)	6921(6)	9225(8)	69(3)
C(29)	-3440(11)	7469(7)	8741(10)	82(4)
C(30)	-2738(10)	7884(6)	8258(9)	76(4)
C(31)	-1641(9)	7736(5)	8259(7)	60(3)

Table S23. Bond lengths [Å] and angles [°] for Nd(L)(NO₃)₃, **Nd 1:1**.

Nd(1)-O(3)	2.362(6)
Nd(1)-O(2)	2.373(5)
Nd(1)-O(1)	2.419(5)
Nd(1)-O(4)	2.504(7)
Nd(1)-O(5)	2.514(7)
Nd(1)-O(10)	2.527(5)
Nd(1)-O(8)	2.534(8)
Nd(1)-O(11)	2.540(6)
Nd(1)-O(7)	2.609(7)
Nd(1)-N(2)	2.934(10)

Nd(1)-N(4)	2.942(7)
P(1)-O(2)	1.502(5)
P(1)-C(13)	1.776(10)
P(1)-C(7)	1.797(8)
P(1)-C(6)	1.832(8)
P(2)-O(3)	1.499(5)
P(2)-C(26)	1.774(9)
P(2)-C(20)	1.786(7)
P(2)-C(19)	1.816(8)
O(1)-N(1)	1.328(7)
O(4)-N(2)	1.253(12)
O(5)-N(2)	1.267(14)
O(6)-N(2)	1.208(13)
O(7)-N(3)	1.239(10)
O(8)-N(3)	1.255(11)
O(9)-N(3)	1.213(10)
O(10)-N(4)	1.258(10)
O(11)-N(4)	1.261(8)
O(12)-N(4)	1.221(9)
N(1)-C(5)	1.366(9)
N(1)-C(1)	1.379(9)
C(1)-C(2)	1.374(11)
C(1)-C(6)	1.489(10)
C(2)-C(3)	1.364(12)
C(3)-C(4)	1.389(11)
C(4)-C(5)	1.358(11)
C(5)-C(19)	1.499(10)
C(7)-C(8)	1.362(12)
C(7)-C(12)	1.399(11)
C(8)-C(9)	1.419(15)
C(9)-C(10)	1.378(17)
C(10)-C(11)	1.326(18)
C(11)-C(12)	1.374(13)
C(13)-C(14)	1.388(12)
C(13)-C(18)	1.395(12)
C(14)-C(15)	1.360(15)

C(15)-C(16)	1.396(15)
C(16)-C(17)	1.363(15)
C(17)-C(18)	1.381(14)
C(20)-C(21)	1.362(13)
C(20)-C(25)	1.370(12)
C(21)-C(22)	1.400(13)
C(22)-C(23)	1.354(15)
C(23)-C(24)	1.354(16)
C(24)-C(25)	1.388(14)
C(26)-C(27)	1.392(11)
C(26)-C(31)	1.393(13)
C(27)-C(28)	1.364(15)
C(28)-C(29)	1.356(17)
C(29)-C(30)	1.383(17)
C(30)-C(31)	1.367(15)

O(3)-Nd(1)-O(2)	97.6(2)
O(3)-Nd(1)-O(1)	73.84(19)
O(2)-Nd(1)-O(1)	74.48(17)
O(3)-Nd(1)-O(4)	72.0(2)
O(2)-Nd(1)-O(4)	74.5(2)
O(1)-Nd(1)-O(4)	129.6(2)
O(3)-Nd(1)-O(5)	79.8(2)
O(2)-Nd(1)-O(5)	123.0(3)
O(1)-Nd(1)-O(5)	150.3(2)
O(4)-Nd(1)-O(5)	50.3(3)
O(3)-Nd(1)-O(10)	154.5(2)
O(2)-Nd(1)-O(10)	78.23(19)
O(1)-Nd(1)-O(10)	127.49(18)
O(4)-Nd(1)-O(10)	82.7(2)
O(5)-Nd(1)-O(10)	81.6(2)
O(3)-Nd(1)-O(8)	80.1(3)
O(2)-Nd(1)-O(8)	145.2(3)
O(1)-Nd(1)-O(8)	71.5(2)
O(4)-Nd(1)-O(8)	135.0(3)
O(5)-Nd(1)-O(8)	91.0(3)

O(10)-Nd(1)-O(8)	117.6(3)
O(3)-Nd(1)-O(11)	153.96(18)
O(2)-Nd(1)-O(11)	78.2(2)
O(1)-Nd(1)-O(11)	80.33(18)
O(4)-Nd(1)-O(11)	129.61(19)
O(5)-Nd(1)-O(11)	124.3(2)
O(10)-Nd(1)-O(11)	50.42(18)
O(8)-Nd(1)-O(11)	88.9(3)
O(3)-Nd(1)-O(7)	116.0(2)
O(2)-Nd(1)-O(7)	146.3(2)
O(1)-Nd(1)-O(7)	111.8(2)
O(4)-Nd(1)-O(7)	116.2(2)
O(5)-Nd(1)-O(7)	68.3(3)
O(10)-Nd(1)-O(7)	72.1(2)
O(8)-Nd(1)-O(7)	48.4(2)
O(11)-Nd(1)-O(7)	70.8(2)
O(3)-Nd(1)-N(2)	71.9(2)
O(2)-Nd(1)-N(2)	99.3(3)
O(1)-Nd(1)-N(2)	144.0(3)
O(4)-Nd(1)-N(2)	25.1(3)
O(5)-Nd(1)-N(2)	25.4(3)
O(10)-Nd(1)-N(2)	83.9(2)
O(8)-Nd(1)-N(2)	112.6(3)
O(11)-Nd(1)-N(2)	134.0(2)
O(7)-Nd(1)-N(2)	93.1(3)
O(3)-Nd(1)-N(4)	175.0(2)
O(2)-Nd(1)-N(4)	77.4(2)
O(1)-Nd(1)-N(4)	104.29(19)
O(4)-Nd(1)-N(4)	106.5(2)
O(5)-Nd(1)-N(4)	103.1(2)
O(10)-Nd(1)-N(4)	25.17(19)
O(8)-Nd(1)-N(4)	103.8(3)
O(11)-Nd(1)-N(4)	25.26(18)
O(7)-Nd(1)-N(4)	69.0(2)
N(2)-Nd(1)-N(4)	108.9(2)
O(2)-P(1)-C(13)	110.3(4)

O(2)-P(1)-C(7)	109.9(4)
C(13)-P(1)-C(7)	111.1(4)
O(2)-P(1)-C(6)	113.0(4)
C(13)-P(1)-C(6)	108.2(4)
C(7)-P(1)-C(6)	104.2(4)
O(3)-P(2)-C(26)	109.8(4)
O(3)-P(2)-C(20)	112.4(3)
C(26)-P(2)-C(20)	107.3(4)
O(3)-P(2)-C(19)	110.5(3)
C(26)-P(2)-C(19)	107.2(4)
C(20)-P(2)-C(19)	109.5(4)
N(1)-O(1)-Nd(1)	136.3(5)
P(1)-O(2)-Nd(1)	144.9(3)
P(2)-O(3)-Nd(1)	136.8(4)
N(2)-O(4)-Nd(1)	97.0(7)
N(2)-O(5)-Nd(1)	96.1(6)
N(3)-O(7)-Nd(1)	96.5(5)
N(3)-O(8)-Nd(1)	99.8(6)
N(4)-O(10)-Nd(1)	96.2(4)
N(4)-O(11)-Nd(1)	95.5(5)
O(12)-N(4)-O(10)	120.9(7)
O(12)-N(4)-O(11)	121.2(8)
O(10)-N(4)-O(11)	117.9(7)
O(12)-N(4)-Nd(1)	179.2(6)
O(10)-N(4)-Nd(1)	58.6(4)
O(11)-N(4)-Nd(1)	59.2(4)
O(1)-N(1)-C(5)	120.2(5)
O(1)-N(1)-C(1)	118.4(5)
C(5)-N(1)-C(1)	121.4(6)
O(6)-N(2)-O(4)	122.0(13)
O(6)-N(2)-O(5)	122.4(11)
O(4)-N(2)-O(5)	115.5(9)
O(6)-N(2)-Nd(1)	171.0(8)
O(4)-N(2)-Nd(1)	57.9(5)
O(5)-N(2)-Nd(1)	58.4(5)
O(9)-N(3)-O(7)	123.0(9)

O(9)-N(3)-O(8)	121.6(9)
O(7)-N(3)-O(8)	115.4(8)
C(2)-C(1)-N(1)	117.5(7)
C(2)-C(1)-C(6)	125.3(7)
N(1)-C(1)-C(6)	117.2(6)
C(3)-C(2)-C(1)	122.3(7)
C(2)-C(3)-C(4)	118.6(8)
C(5)-C(4)-C(3)	120.2(8)
C(4)-C(5)-N(1)	120.1(7)
C(4)-C(5)-C(19)	123.2(7)
N(1)-C(5)-C(19)	116.7(6)
C(1)-C(6)-P(1)	115.6(6)
C(8)-C(7)-C(12)	119.2(8)
C(8)-C(7)-P(1)	118.5(7)
C(12)-C(7)-P(1)	122.2(7)
C(7)-C(8)-C(9)	119.6(10)
C(10)-C(9)-C(8)	119.2(11)
C(11)-C(10)-C(9)	120.4(11)
C(10)-C(11)-C(12)	121.8(12)
C(11)-C(12)-C(7)	119.6(10)
C(14)-C(13)-C(18)	118.4(9)
C(14)-C(13)-P(1)	118.7(7)
C(18)-C(13)-P(1)	122.9(7)
C(15)-C(14)-C(13)	120.4(9)
C(14)-C(15)-C(16)	120.7(10)
C(17)-C(16)-C(15)	119.7(11)
C(16)-C(17)-C(18)	119.9(10)
C(17)-C(18)-C(13)	120.9(9)
C(5)-C(19)-P(2)	109.0(6)
C(21)-C(20)-C(25)	119.0(8)
C(21)-C(20)-P(2)	124.1(6)
C(25)-C(20)-P(2)	116.8(7)
C(20)-C(21)-C(22)	120.6(10)
C(23)-C(22)-C(21)	119.2(11)
C(22)-C(23)-C(24)	121.1(9)
C(23)-C(24)-C(25)	119.6(10)

C(20)-C(25)-C(24)	120.4(10)
C(27)-C(26)-C(31)	118.3(9)
C(27)-C(26)-P(2)	122.9(7)
C(31)-C(26)-P(2)	118.7(7)
C(28)-C(27)-C(26)	120.4(10)
C(29)-C(28)-C(27)	121.0(11)
C(28)-C(29)-C(30)	119.8(12)
C(31)-C(30)-C(29)	120.2(11)
C(30)-C(31)-C(26)	120.4(10)

Table S24. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for Nd(L)(NO₃)₃, **Nd 1:1**. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^*{}^2 U^{11} + \dots + 2 h k a^* b^* U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Nd(1)	31(1)	26(1)	32(1)	-4(1)	5(1)	3(1)
P(1)	38(1)	28(1)	31(1)	0(1)	-1(1)	2(1)
P(2)	28(1)	31(1)	32(1)	1(1)	0(1)	-1(1)
O(1)	37(4)	33(3)	38(3)	-2(2)	5(3)	2(2)
O(2)	56(5)	38(3)	38(3)	-6(2)	1(3)	11(3)
O(3)	46(4)	47(3)	36(3)	-1(3)	2(3)	-8(3)
O(4)	70(5)	51(4)	45(4)	-5(3)	-3(3)	-1(3)
O(5)	79(7)	43(4)	74(6)	-21(4)	1(5)	9(4)
O(6)	122(9)	96(6)	86(6)	-38(5)	-16(6)	-44(6)
O(7)	59(5)	55(4)	77(5)	4(4)	10(4)	19(3)
O(8)	72(8)	83(6)	63(6)	21(5)	12(5)	38(6)
O(9)	76(6)	51(4)	81(5)	9(4)	-20(4)	21(4)
O(10)	40(4)	55(3)	36(3)	-10(3)	3(3)	-7(3)
O(11)	51(5)	51(3)	36(3)	-11(3)	3(3)	-12(3)
O(12)	53(6)	96(5)	84(5)	-14(4)	27(5)	-30(4)
N(4)	40(5)	40(4)	49(5)	-8(3)	9(4)	-3(3)
N(1)	32(4)	28(3)	24(3)	-2(2)	3(3)	5(3)

N(2)	72(9)	61(6)	46(5)	-19(5)	12(6)	-22(6)
N(3)	55(6)	31(4)	64(6)	3(4)	-3(5)	3(4)
C(1)	40(5)	33(4)	22(4)	-5(3)	-4(3)	4(3)
C(2)	71(8)	32(4)	36(5)	-4(3)	8(5)	15(4)
C(3)	50(7)	59(6)	37(5)	-4(4)	12(5)	21(5)
C(4)	42(6)	40(4)	39(5)	-5(4)	14(4)	6(4)
C(5)	36(5)	35(4)	21(4)	-1(3)	-1(3)	-4(3)
C(6)	51(6)	28(4)	30(4)	-2(3)	-5(4)	-3(4)
C(7)	39(6)	43(5)	35(4)	11(3)	-6(4)	-4(4)
C(8)	54(8)	81(7)	53(6)	6(5)	7(6)	-2(6)
C(9)	59(9)	136(12)	53(7)	18(7)	15(6)	-21(8)
C(10)	83(11)	89(9)	80(9)	39(7)	-22(8)	-35(8)
C(11)	77(10)	63(7)	79(8)	23(6)	-26(7)	-26(6)
C(12)	71(8)	46(5)	53(6)	11(4)	-9(5)	-10(5)
C(13)	48(6)	31(4)	27(4)	3(3)	-1(4)	-3(4)
C(14)	41(7)	50(5)	63(6)	-8(4)	-10(5)	2(4)
C(15)	72(10)	66(6)	66(7)	-10(5)	-29(6)	-14(6)
C(16)	36(7)	83(7)	72(7)	18(6)	-17(5)	-17(6)
C(17)	47(8)	62(6)	58(6)	10(5)	-1(5)	16(5)
C(18)	41(7)	46(5)	48(6)	-4(4)	-4(5)	3(4)
C(19)	39(5)	25(4)	34(4)	4(3)	2(4)	-7(3)
C(20)	25(5)	27(4)	42(4)	-7(3)	-1(4)	0(3)
C(21)	89(10)	35(5)	48(6)	-1(4)	-17(6)	5(5)
C(22)	113(12)	45(5)	72(7)	5(5)	-15(7)	13(6)
C(23)	74(9)	38(5)	83(8)	-11(5)	1(7)	9(5)
C(24)	103(12)	48(6)	71(8)	-22(6)	-24(7)	-4(6)
C(25)	97(10)	47(5)	54(6)	-10(5)	-23(6)	0(5)
C(26)	32(5)	41(4)	38(4)	-8(3)	-1(4)	-1(4)
C(27)	42(7)	64(6)	47(5)	-9(4)	6(5)	-11(5)
C(28)	41(8)	91(8)	76(8)	-24(7)	13(6)	-12(6)
C(29)	29(7)	102(10)	113(10)	-40(8)	-9(7)	9(6)
C(30)	51(8)	62(7)	115(10)	-8(6)	-24(7)	31(6)
C(31)	48(7)	58(5)	73(7)	0(5)	-8(5)	7(5)

X-ray Data Collection, Structure Solution and Refinement for Am(L)(NO₃)₃, Am 1:1.

A yellow crystal of approximate dimensions 0.3 x 0.3 x 0.1 mm was mounted in a radiological fume hood in a 0.3 mm quartz capillary tube that was covered with a clear coat of colorless nail polish and contained a small amount of original mother liquor. The capillary was sealed using sealing wax to provide a solvent saturated environment and was secured with super glue in a MiTeGen Reusable Base™ before being completely encapsulated in polyester heat-shrink tubing to create a non-dispersible configuration. The sample configuration was then transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at room temperature (10 sec/frame scan time for a sphere of diffraction data) using AgK α radiation. The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The chiral monoclinic space group *Cc* was assigned and later determined to be correct.

The initial structure was solved by direct methods using Pu in place of Am, since Am is not recognized by APEX3. The structure was refined on F^2 by full-matrix least-squares techniques using Am, the scattering factors for which were taken from the International Tables for Crystallography Volume C.⁶ O(12) was disordered and included using multiple components with partial site-occupancy-factors. All hydrogen atoms were included using a riding model.

Table S25. Crystal data and structure refinement for Am(L)(NO₃)₃, Am 1:1.

Empirical formula



Formula weight	952.50
Temperature	293(2) K
Wavelength	0.56086 Å
Crystal system	Monoclinic
Space group	Cc
Unit cell dimensions	a = 12.216(5) Å b = 19.158(6) Å c = 14.838(5) Å
	α= 90°. β= 91.680(11)°. γ = 90°.
Volume	3471(2) Å ³
Z	4
Density (calculated)	1.823 mg/m ³
Absorption coefficient	2.987 mm ⁻¹
F(000)	1848
Crystal size	0.3 x 0.3 x 0.1
Theta range for data collection	2.633 to 21.954°.
Index ranges	-16 ≤ h ≤ 16, -25 ≤ k ≤ 25, -19 ≤ l ≤ 19
Reflections collected	20103
Independent reflections	8401 [R(int) = 0.0656]
Completeness to theta = 19.665°	99.7 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.7455 and 0.2657
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	8401 / 2 / 451
Goodness-of-fit on F ²	1.036
Final R indices [I>2sigma(I)]	R1 = 0.0485, wR2 = 0.1045
R indices (all data)	R1 = 0.0668, wR2 = 0.1112
Absolute structure parameter	0.060(19)
Largest diff. peak and hole	2.207 and -1.678 e.Å ⁻³

Table S26. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for Am(L)(NO₃)₃, **Am 1:1**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Am(1)	2449(1)	7716(1)	7508(1)	31(1)
P(1)	1437(3)	9534(2)	7301(2)	35(1)
P(2)	210(3)	7019(2)	8759(2)	31(1)
N(1)	1140(10)	8610(5)	9237(7)	33(2)
O(1)	2034(8)	8352(4)	8862(6)	35(2)
O(2)	1907(9)	8848(5)	7013(7)	46(2)
O(3)	677(9)	7335(5)	7930(7)	42(2)
C(1)	927(12)	9313(6)	9128(9)	34(3)
C(2)	35(17)	9583(7)	9559(10)	52(4)
C(3)	-597(15)	9188(8)	10092(12)	53(4)
C(4)	-378(13)	8481(7)	10184(9)	40(3)
C(5)	515(12)	8203(6)	9744(8)	35(3)
C(6)	1636(13)	9717(7)	8515(9)	38(3)
C(7)	2074(13)	10245(7)	6755(10)	42(3)
C(8)	1834(17)	10943(8)	6905(12)	57(4)
C(9)	2380(20)	11468(10)	6497(16)	78(7)
C(10)	3180(20)	11324(14)	5892(16)	88(8)
C(11)	3445(18)	10646(16)	5731(14)	89(8)
C(12)	2906(15)	10104(11)	6161(12)	60(4)
C(13)	20(12)	9542(7)	7048(9)	37(3)
C(14)	-444(16)	9033(8)	6486(12)	57(4)
C(15)	-1553(19)	9014(11)	6287(15)	75(6)
C(16)	-2214(17)	9504(11)	6650(14)	70(5)
C(17)	-1817(15)	10024(10)	7194(13)	62(5)
C(18)	-684(15)	10049(8)	7396(12)	49(4)
C(19)	776(11)	7432(6)	9778(8)	29(3)
C(20)	-1228(13)	7168(7)	8744(10)	40(3)
C(21)	-1651(16)	7734(9)	8247(13)	57(4)
C(22)	-2750(20)	7883(12)	8250(18)	82(7)

C(23)	-3430(20)	7471(15)	8729(19)	83(7)
C(24)	-3065(15)	6932(14)	9219(15)	73(6)
C(25)	-1944(14)	6764(10)	9245(11)	55(4)
C(26)	420(12)	6101(6)	8830(9)	35(3)
C(27)	960(18)	5772(8)	9544(13)	57(5)
C(28)	1090(20)	5045(9)	9530(14)	76(6)
C(29)	679(18)	4666(7)	8831(13)	66(5)
C(30)	150(20)	4987(9)	8112(14)	74(6)
C(31)	37(19)	5706(8)	8100(13)	67(5)
N(2)	3839(11)	6678(6)	8554(11)	48(3)
N(3)	1490(20)	6883(9)	6004(11)	64(5)
N(4)	4542(11)	8324(6)	6928(9)	44(3)
O(4)	3016(15)	6976(10)	8854(12)	77(6)
O(5)	4097(11)	6846(6)	7764(9)	65(3)
O(6)	4386(13)	6270(6)	9003(10)	72(4)
O(7)	2207(16)	6636(7)	6533(10)	75(5)
O(8)	1301(14)	7538(7)	6093(8)	66(4)
O(9)	947(16)	6539(9)	5476(12)	100(6)
O(10)	3895(9)	7997(6)	6378(7)	47(2)
O(11)	4225(9)	8391(5)	7729(7)	48(3)
O(12)	5500(20)	8471(14)	6770(19)	54(7)
O(12A)	5300(20)	8659(14)	6590(18)	55(7)

Table S27. Bond lengths [Å] and angles [°] for Am(L)(NO₃)₃, **Am 1:1**.

Am(1)-O(2)	2.379(10)
Am(1)-O(3)	2.385(10)
Am(1)-O(1)	2.417(9)
Am(1)-O(8)	2.514(13)
Am(1)-O(10)	2.529(10)
Am(1)-O(4)	2.529(15)
Am(1)-O(7)	2.537(12)
Am(1)-O(11)	2.539(10)
Am(1)-O(5)	2.632(12)

Am(1)-N(3)	2.956(17)
Am(1)-N(4)	2.960(13)
Am(1)-N(2)	3.015(13)
P(1)-O(2)	1.501(10)
P(1)-C(13)	1.759(15)
P(1)-C(7)	1.777(14)
P(1)-C(6)	1.844(14)
P(2)-O(3)	1.499(10)
P(2)-C(20)	1.779(16)
P(2)-C(26)	1.780(12)
P(2)-C(19)	1.824(13)
N(1)-O(1)	1.334(14)
N(1)-C(5)	1.339(16)
N(1)-C(1)	1.380(15)
C(1)-C(2)	1.38(2)
C(1)-C(6)	1.490(19)
C(2)-C(3)	1.35(2)
C(3)-C(4)	1.39(2)
C(4)-C(5)	1.39(2)
C(5)-C(19)	1.512(17)
C(7)-C(8)	1.39(2)
C(7)-C(12)	1.39(2)
C(8)-C(9)	1.36(2)
C(9)-C(10)	1.37(4)
C(10)-C(11)	1.36(4)
C(11)-C(12)	1.39(3)
C(13)-C(14)	1.39(2)
C(13)-C(18)	1.41(2)
C(14)-C(15)	1.38(3)
C(15)-C(16)	1.36(3)
C(16)-C(17)	1.36(3)
C(17)-C(18)	1.41(3)
C(20)-C(25)	1.40(2)
C(20)-C(21)	1.40(2)
C(21)-C(22)	1.38(3)
C(22)-C(23)	1.36(4)

C(23)-C(24)	1.33(4)
C(24)-C(25)	1.41(3)
C(26)-C(27)	1.38(2)
C(26)-C(31)	1.39(2)
C(27)-C(28)	1.40(2)
C(28)-C(29)	1.35(3)
C(29)-C(30)	1.38(3)
C(30)-C(31)	1.38(2)
N(2)-O(6)	1.213(17)
N(2)-O(4)	1.249(19)
N(2)-O(5)	1.265(19)
N(3)-O(9)	1.21(2)
N(3)-O(7)	1.25(3)
N(3)-O(8)	1.28(2)
N(4)-O(12)	1.23(3)
N(4)-O(12A)	1.24(3)
N(4)-O(11)	1.268(15)
N(4)-O(10)	1.282(17)

O(2)-Am(1)-O(3)	96.5(4)
O(2)-Am(1)-O(1)	74.5(3)
O(3)-Am(1)-O(1)	73.9(3)
O(2)-Am(1)-O(8)	73.9(4)
O(3)-Am(1)-O(8)	71.9(4)
O(1)-Am(1)-O(8)	129.6(4)
O(2)-Am(1)-O(10)	78.1(4)
O(3)-Am(1)-O(10)	153.6(4)
O(1)-Am(1)-O(10)	127.7(3)
O(8)-Am(1)-O(10)	81.9(5)
O(2)-Am(1)-O(4)	145.3(5)
O(3)-Am(1)-O(4)	81.4(5)
O(1)-Am(1)-O(4)	71.7(5)
O(8)-Am(1)-O(4)	135.8(7)
O(10)-Am(1)-O(4)	117.6(5)
O(2)-Am(1)-O(7)	122.8(5)
O(3)-Am(1)-O(7)	79.0(5)

O(1)-Am(1)-O(7)	149.5(5)
O(8)-Am(1)-O(7)	50.2(5)
O(10)-Am(1)-O(7)	82.4(5)
O(4)-Am(1)-O(7)	91.0(7)
O(2)-Am(1)-O(11)	78.7(4)
O(3)-Am(1)-O(11)	154.2(3)
O(1)-Am(1)-O(11)	80.5(3)
O(8)-Am(1)-O(11)	129.1(4)
O(10)-Am(1)-O(11)	50.7(3)
O(4)-Am(1)-O(11)	88.3(6)
O(7)-Am(1)-O(11)	125.1(5)
O(2)-Am(1)-O(5)	145.8(4)
O(3)-Am(1)-O(5)	117.7(4)
O(1)-Am(1)-O(5)	112.2(4)
O(8)-Am(1)-O(5)	116.1(5)
O(10)-Am(1)-O(5)	71.6(4)
O(4)-Am(1)-O(5)	48.7(5)
O(7)-Am(1)-O(5)	69.0(5)
O(11)-Am(1)-O(5)	70.0(4)
O(2)-Am(1)-N(3)	99.1(5)
O(3)-Am(1)-N(3)	72.0(5)
O(1)-Am(1)-N(3)	144.3(5)
O(8)-Am(1)-N(3)	25.5(5)
O(10)-Am(1)-N(3)	83.3(4)
O(4)-Am(1)-N(3)	112.8(6)
O(7)-Am(1)-N(3)	24.9(5)
O(11)-Am(1)-N(3)	133.7(5)
O(5)-Am(1)-N(3)	93.0(6)
O(2)-Am(1)-N(4)	77.7(4)
O(3)-Am(1)-N(4)	174.1(3)
O(1)-Am(1)-N(4)	104.3(3)
O(8)-Am(1)-N(4)	106.0(5)
O(10)-Am(1)-N(4)	25.5(3)
O(4)-Am(1)-N(4)	103.4(6)
O(7)-Am(1)-N(4)	104.0(5)
O(11)-Am(1)-N(4)	25.2(3)

O(5)-Am(1)-N(4)	68.2(4)
N(3)-Am(1)-N(4)	108.6(5)
O(2)-Am(1)-N(2)	155.2(4)
O(3)-Am(1)-N(2)	99.5(4)
O(1)-Am(1)-N(2)	91.9(4)
O(8)-Am(1)-N(2)	129.3(4)
O(10)-Am(1)-N(2)	95.1(4)
O(4)-Am(1)-N(2)	24.0(4)
O(7)-Am(1)-N(2)	79.1(5)
O(11)-Am(1)-N(2)	78.7(4)
O(5)-Am(1)-N(2)	24.7(4)
N(3)-Am(1)-N(2)	103.7(5)
N(4)-Am(1)-N(2)	86.1(4)
O(2)-P(1)-C(13)	109.2(7)
O(2)-P(1)-C(7)	111.3(7)
C(13)-P(1)-C(7)	109.7(7)
O(2)-P(1)-C(6)	113.8(6)
C(13)-P(1)-C(6)	107.9(7)
C(7)-P(1)-C(6)	104.7(7)
O(2)-P(1)-Am(1)	21.8(4)
C(13)-P(1)-Am(1)	110.4(4)
C(7)-P(1)-Am(1)	127.5(5)
C(6)-P(1)-Am(1)	93.6(4)
O(3)-P(2)-C(20)	109.0(7)
O(3)-P(2)-C(26)	112.9(6)
C(20)-P(2)-C(26)	107.4(6)
O(3)-P(2)-C(19)	111.1(6)
C(20)-P(2)-C(19)	106.9(6)
C(26)-P(2)-C(19)	109.2(6)
O(3)-P(2)-Am(1)	27.8(4)
C(20)-P(2)-Am(1)	134.3(5)
C(26)-P(2)-Am(1)	106.6(5)
C(19)-P(2)-Am(1)	89.5(4)
O(1)-N(1)-C(5)	120.4(10)
O(1)-N(1)-C(1)	117.8(10)
C(5)-N(1)-C(1)	121.7(11)

O(1)-N(1)-Am(1)	28.2(5)
C(5)-N(1)-Am(1)	114.0(7)
C(1)-N(1)-Am(1)	118.7(7)
N(1)-O(1)-Am(1)	136.7(8)
P(1)-O(2)-Am(1)	144.6(6)
P(2)-O(3)-Am(1)	135.2(6)
C(2)-C(1)-N(1)	117.4(12)
C(2)-C(1)-C(6)	124.7(12)
N(1)-C(1)-C(6)	117.8(11)
C(3)-C(2)-C(1)	122.2(13)
C(2)-C(3)-C(4)	119.5(14)
C(3)-C(4)-C(5)	118.6(13)
N(1)-C(5)-C(4)	120.5(12)
N(1)-C(5)-C(19)	117.6(11)
C(4)-C(5)-C(19)	121.7(12)
N(1)-C(5)-Am(1)	49.2(6)
C(4)-C(5)-Am(1)	156.1(10)
C(19)-C(5)-Am(1)	71.8(6)
C(1)-C(6)-P(1)	115.8(10)
C(1)-C(6)-Am(1)	83.9(7)
P(1)-C(6)-Am(1)	60.7(4)
C(8)-C(7)-C(12)	116.8(16)
C(8)-C(7)-P(1)	124.5(13)
C(12)-C(7)-P(1)	118.6(13)
C(9)-C(8)-C(7)	122(2)
C(8)-C(9)-C(10)	121(2)
C(11)-C(10)-C(9)	119(2)
C(10)-C(11)-C(12)	121(2)
C(7)-C(12)-C(11)	121(2)
C(14)-C(13)-C(18)	117.4(15)
C(14)-C(13)-P(1)	120.2(12)
C(18)-C(13)-P(1)	122.4(11)
C(15)-C(14)-C(13)	121.8(17)
C(16)-C(15)-C(14)	119.2(18)
C(15)-C(16)-C(17)	122.3(19)
C(16)-C(17)-C(18)	118.8(18)

C(13)-C(18)-C(17)	120.4(15)
C(5)-C(19)-P(2)	108.8(9)
C(5)-C(19)-Am(1)	87.3(7)
P(2)-C(19)-Am(1)	63.6(4)
C(25)-C(20)-C(21)	118.8(16)
C(25)-C(20)-P(2)	122.5(12)
C(21)-C(20)-P(2)	118.6(13)
C(22)-C(21)-C(20)	120(2)
C(23)-C(22)-C(21)	119(2)
C(24)-C(23)-C(22)	123(2)
C(23)-C(24)-C(25)	120(2)
C(20)-C(25)-C(24)	118.8(19)
C(27)-C(26)-C(31)	119.3(13)
C(27)-C(26)-P(2)	124.1(11)
C(31)-C(26)-P(2)	116.6(11)
C(26)-C(27)-C(28)	119.5(16)
C(29)-C(28)-C(27)	120.4(17)
C(28)-C(29)-C(30)	120.7(15)
C(29)-C(30)-C(31)	119.8(17)
C(30)-C(31)-C(26)	120.1(17)
O(6)-N(2)-O(4)	122.3(17)
O(6)-N(2)-O(5)	121.6(16)
O(4)-N(2)-O(5)	116.0(14)
O(6)-N(2)-Am(1)	177.7(13)
O(4)-N(2)-Am(1)	55.6(9)
O(5)-N(2)-Am(1)	60.4(7)
O(9)-N(3)-O(7)	124.1(19)
O(9)-N(3)-O(8)	120(2)
O(7)-N(3)-O(8)	115.6(16)
O(9)-N(3)-Am(1)	169.1(17)
O(7)-N(3)-Am(1)	58.5(9)
O(8)-N(3)-Am(1)	57.6(9)
O(12)-N(4)-O(11)	118.2(18)
O(12A)-N(4)-O(11)	125.2(17)
O(12)-N(4)-O(10)	124.2(17)
O(12A)-N(4)-O(10)	116.6(17)

O(11)-N(4)-O(10)	116.5(12)
O(12)-N(4)-Am(1)	168.0(17)
O(12A)-N(4)-Am(1)	168.3(17)
O(11)-N(4)-Am(1)	58.5(7)
O(10)-N(4)-Am(1)	58.1(7)
N(2)-O(4)-Am(1)	100.4(11)
N(2)-O(5)-Am(1)	94.9(9)
N(3)-O(7)-Am(1)	96.7(10)
N(3)-O(8)-Am(1)	96.9(12)
N(3)-O(9)-Am(1)	7.8(12)
N(4)-O(10)-Am(1)	96.4(8)
N(4)-O(11)-Am(1)	96.3(8)

Table S28. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $\text{Am}(\mathbf{L})(\text{NO}_3)_3$, **Am 1:1**. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^{*2} U^{11} + \dots + 2 h k a^{*} b^{*} U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Am(1)	32(1)	26(1)	36(1)	-4(1)	4(1)	2(1)
P(1)	45(2)	25(1)	35(2)	0(1)	1(2)	2(1)
P(2)	30(2)	29(1)	35(2)	1(1)	1(1)	-1(1)
N(1)	43(7)	27(4)	28(5)	-1(4)	6(5)	2(4)
O(1)	30(5)	38(4)	37(5)	-1(4)	2(4)	1(4)
O(2)	42(6)	43(5)	52(6)	-1(4)	7(5)	12(4)
O(3)	35(6)	48(5)	41(5)	1(4)	3(4)	-8(4)
C(1)	39(8)	30(5)	32(6)	-5(5)	-4(5)	4(5)
C(2)	84(13)	33(6)	40(8)	-3(5)	7(8)	10(7)
C(3)	56(11)	51(8)	52(9)	-9(7)	21(8)	21(8)
C(4)	43(8)	39(6)	39(7)	-2(5)	10(6)	3(6)
C(5)	39(8)	35(6)	30(6)	-2(5)	2(5)	0(6)
C(6)	44(9)	31(6)	39(7)	6(5)	-5(6)	5(6)
C(7)	39(9)	44(7)	42(8)	8(6)	-3(6)	-4(6)
C(8)	72(13)	43(8)	54(9)	13(7)	-10(8)	-8(8)

C(9)	82(16)	55(10)	93(15)	31(10)	-40(13)	-34(10)
C(10)	84(18)	100(18)	78(15)	36(13)	-19(13)	-39(15)
C(11)	50(13)	150(20)	64(13)	24(14)	12(10)	-14(14)
C(12)	45(11)	78(11)	57(10)	16(9)	-3(8)	4(9)
C(13)	40(8)	36(7)	34(7)	6(5)	-6(6)	-1(6)
C(14)	55(11)	51(8)	65(11)	-10(8)	-7(8)	-2(8)
C(15)	69(15)	66(11)	90(15)	-17(10)	-10(12)	-22(11)
C(16)	43(11)	86(13)	80(13)	14(11)	-10(10)	-20(10)
C(17)	45(11)	66(10)	73(12)	12(9)	4(9)	15(8)
C(18)	52(11)	38(7)	58(10)	-6(7)	0(8)	3(7)
C(19)	31(7)	26(5)	31(6)	2(4)	-2(5)	-2(5)
C(20)	38(8)	42(7)	39(7)	-4(5)	-5(6)	-1(6)
C(21)	41(10)	60(9)	70(11)	6(8)	-12(8)	9(8)
C(22)	76(17)	75(13)	95(17)	-4(12)	-17(13)	26(12)
C(23)	50(13)	101(15)	97(17)	-39(14)	-17(12)	18(12)
C(24)	31(9)	113(17)	76(13)	-34(13)	8(9)	-19(11)
C(25)	44(10)	76(11)	45(8)	-11(8)	6(7)	-10(8)
C(26)	34(7)	24(5)	46(7)	-4(5)	-3(6)	1(5)
C(27)	72(14)	44(8)	53(10)	-4(7)	-31(9)	15(9)
C(28)	103(19)	45(8)	77(13)	12(9)	-28(12)	11(10)
C(29)	86(15)	25(6)	86(13)	-4(7)	7(11)	-1(8)
C(30)	110(20)	43(9)	67(13)	-16(9)	-28(13)	-4(11)
C(31)	93(16)	46(8)	59(11)	-11(8)	-25(10)	-4(9)
N(2)	32(7)	39(6)	74(9)	9(6)	-3(7)	-4(5)
N(3)	97(16)	55(9)	41(8)	-15(7)	13(10)	-30(10)
N(4)	41(7)	39(6)	53(7)	-8(5)	9(6)	-2(5)
O(4)	66(12)	96(11)	71(11)	32(9)	12(9)	57(10)
O(5)	53(8)	57(6)	83(9)	8(6)	3(7)	14(6)
O(6)	76(10)	53(7)	85(10)	9(6)	-20(8)	20(7)
O(7)	114(14)	45(6)	65(9)	-28(6)	2(9)	17(7)
O(8)	105(12)	53(6)	41(6)	-3(5)	-8(7)	6(7)
O(9)	117(15)	94(10)	87(11)	-40(9)	-18(10)	-41(11)
O(10)	45(6)	59(6)	37(5)	-11(5)	6(4)	-10(5)
O(11)	47(7)	52(5)	45(6)	-11(5)	5(5)	-15(5)

X-ray Data Collection, Structure Solution and Refinement for [Eu(L)₂(H₂O)₂][3(NO₃)].

A colorless crystal of approximate dimensions 0.240 x 0.232 x 0.110 mm was mounted in Krytox™ grease on a MiTeGen Reusable Base™ before being transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at room temperature (45 sec/frame scan time for a sphere of diffraction data) using MoK α radiation. The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The chiral monoclinic space group $P2_1$ was assigned and later determined to be correct.

The structure was solved by direct methods and refined on F² by full-matrix least-squares techniques as a 2-component (3:97) inversion twin. There was significant thermal motion in certain atoms that prevented universal anisotropic refinement. N(5), O(17), O(18), and O(19) possess particularly severe thermal motion, and a satisfactory disorder model could not be devised for these atoms. These atoms have been fixed in a trigonal planar geometry, and N(5) bonds to O(15) and O(17) have fixed lengths. There was one cocrystallized water molecule. The hydrogen atoms on the free water and water ligands could not be located and were therefore not included. All other hydrogen atoms were included using a riding model. It was later determined that analogous compounds react with Krytox™ grease which could have contributed to poorer data quality in this case.

Table S29. Crystal data and structure refinement for [Eu(L)₂(H₂O)₂][3(NO₃)].

Empirical formula	[C ₆₂ H ₅₄ EuN ₂ O ₆ P ₄ (H ₂ O) ₂][3(NO ₃)](H ₂ O)	
Formula weight	1438.99	
Temperature	293(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	P2 ₁	
Unit cell dimensions	a = 13.8222(6) Å	α= 90°.
	b = 16.6815(8) Å	β= 113.7618(16)°.
	c = 15.1765(7) Å	γ = 90°.
Volume	3202.7(3) Å ³	
Z	2	
Density (calculated)	1.492 mg/m ³	
Absorption coefficient	1.154 mm ⁻¹	
F(000)	1468	
Crystal size	0.240 x 0.232 x 0.110 mm ³	
Theta range for data collection	2.925 to 27.103°.	
Index ranges	-17 ≤ h ≤ 17, -21 ≤ k ≤ 21, -19 ≤ l ≤ 19	
Reflections collected	67814	
Independent reflections	14136 [R(int) = 0.0665]	
Completeness to theta = 25.242°	99.8 %	
Absorption correction	Numerical	
Max. and min. transmission	0.8704 and 0.7501	
Refinement method	Full-matrix least-squares on F ²	
Data / restraints / parameters	14136 / 3 / 552	
Goodness-of-fit on F ²	1.165	
Final R indices [I>2sigma(I)]	R1 = 0.0664, wR2 = 0.1400	
R indices (all data)	R1 = 0.0739, wR2 = 0.1432	
Absolute structure parameter	0.032(18)	
Largest diff. peak and hole	1.203 and -3.655 e.Å ⁻³	

Table S30. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Eu}(\text{L})_2(\text{H}_2\text{O})_2][3(\text{NO}_3)]$. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
Eu(1)	32804(1)	2550(1)	31862(1)	21(1)
P(1)	35275(2)	2892(2)	34045(2)	31(1)
P(2)	31367(2)	1200(2)	32786(2)	30(1)
P(3)	34471(2)	1582(2)	30846(2)	32(1)
P(4)	30315(2)	2677(2)	29665(2)	29(1)
N(1)	34038(7)	1092(5)	33674(7)	30(2)
N(2)	31911(7)	1076(5)	30059(6)	27(2)
O(1)	33834(6)	1341(5)	32793(5)	32(2)
O(2)	34230(7)	3021(5)	33242(6)	42(2)
O(3)	31765(6)	1972(5)	32584(6)	36(2)
O(4)	32063(6)	1291(5)	30947(5)	30(2)
O(5)	33890(6)	2271(5)	31053(6)	37(2)
O(6)	31329(6)	2902(5)	30482(6)	36(2)
C(1)	34948(8)	1322(7)	34400(8)	35(3)
C(2)	35170(10)	1036(9)	35330(9)	49(3)
C(3)	34486(12)	565(10)	35517(10)	57(4)
C(4)	33559(11)	332(8)	34757(11)	48(3)
C(5)	33331(9)	609(7)	33833(9)	36(3)
C(6)	35690(8)	1857(7)	34189(9)	34(3)
C(7)	35230(10)	3232(7)	35146(8)	38(3)
C(8)	34362(14)	3646(11)	35104(13)	68(4)
C(9)	34294(19)	3959(15)	35928(17)	98(7)
C(10)	35111(19)	3838(14)	36794(18)	99(7)
C(11)	35985(17)	3429(13)	36844(16)	86(6)
C(12)	36067(13)	3120(10)	36038(12)	62(4)
C(13)	36311(10)	3423(8)	33874(8)	39(3)
C(14)	36034(11)	3966(8)	33123(10)	48(3)
C(15)	36814(16)	4414(12)	32993(14)	69(5)
C(16)	37867(15)	4244(11)	33571(13)	71(5)
C(17)	38135(14)	3722(10)	34317(13)	65(4)

C(18)	37370(12)	3301(9)	34458(11)	56(4)
C(19)	32321(8)	400(7)	32977(9)	34(3)
C(20)	31120(11)	1306(8)	33844(9)	42(3)
C(21)	31508(16)	1949(13)	34419(15)	81(5)
C(22)	31310(20)	2042(17)	35270(20)	119(9)
C(23)	30818(19)	1455(16)	35505(19)	106(7)
C(24)	30474(16)	829(13)	34972(15)	80(5)
C(25)	30640(15)	729(12)	34137(14)	71(5)
C(26)	30154(9)	873(7)	31841(8)	35(3)
C(27)	29262(11)	1349(9)	31652(11)	51(3)
C(28)	28326(14)	1142(11)	30881(12)	64(4)
C(29)	28267(13)	513(10)	30325(12)	59(4)
C(30)	29128(12)	60(10)	30499(11)	57(4)
C(31)	30090(11)	222(8)	31244(10)	45(3)
C(32)	32712(9)	714(7)	29911(9)	36(3)
C(33)	32554(11)	477(9)	28988(10)	52(4)
C(34)	31597(13)	601(11)	28235(12)	68(5)
C(35)	30809(12)	951(9)	28411(10)	53(4)
C(36)	30954(9)	1193(7)	29324(8)	32(2)
C(37)	33759(9)	653(7)	30757(9)	36(3)
C(38)	35800(10)	1476(9)	31749(10)	45(3)
C(39)	36407(12)	2165(10)	31936(11)	57(4)
C(40)	37439(15)	2135(12)	32637(13)	73(5)
C(41)	37796(15)	1447(11)	33126(14)	71(5)
C(42)	37207(15)	768(13)	32946(14)	74(5)
C(43)	36180(14)	768(11)	32224(12)	65(4)
C(44)	34564(9)	1719(7)	29705(9)	37(3)
C(45)	33934(11)	2276(8)	29095(10)	51(3)
C(46)	33905(15)	2320(11)	28156(14)	80(5)
C(47)	34587(17)	1833(13)	27932(16)	86(6)
C(48)	35189(15)	1297(12)	28528(14)	78(5)
C(49)	35218(11)	1213(9)	29442(11)	49(3)
C(50)	30110(8)	1603(6)	29546(8)	30(2)
C(51)	30348(9)	3027(7)	28556(8)	36(3)
C(52)	29553(16)	2897(11)	27679(14)	77(5)
C(53)	29650(20)	3143(15)	26841(19)	103(7)

C(54)	30539(16)	3551(12)	26924(15)	82(5)
C(55)	31357(13)	3656(10)	27774(11)	58(4)
C(56)	31255(11)	3391(8)	28602(10)	46(3)
C(57)	29190(9)	3033(8)	29880(10)	41(3)
C(58)	29412(13)	3425(9)	30739(11)	57(4)
C(59)	28563(16)	3633(12)	30970(15)	77(5)
C(60)	27551(16)	3470(12)	30353(14)	77(5)
C(61)	27337(14)	3080(10)	29532(13)	66(4)
C(62)	28157(12)	2848(9)	29265(12)	56(4)
O(7)	33280(6)	3843(5)	31383(6)	36(2)
O(8)	32017(7)	3642(5)	32462(6)	41(2)
N(3)	34569(9)	4138(8)	29860(9)	50(3)
O(9)	33656(10)	4163(8)	29772(9)	73(3)
O(10)	35267(13)	4151(10)	30688(12)	104(5)
O(11)	34836(11)	4110(9)	29188(10)	83(4)
N(4)	27443(10)	644(7)	27695(8)	48(3)
O(12)	28278(12)	290(9)	27998(10)	88(4)
O(13)	27427(11)	1359(9)	27491(10)	89(4)
O(14)	26627(10)	273(8)	27588(9)	74(3)
N(5)	28944(18)	-738(15)	35839(17)	240(20)
O(17)	28360(30)	-1160(20)	35940(30)	274(18)
O(16)	29610(30)	-240(20)	36480(30)	260(16)
O(15)	28880(40)	-670(30)	35100(20)	440(40)
O(18)	38083(13)	1426(10)	35896(12)	110(5)

Table S31. Bond lengths [\AA] and angles [°] for $[\text{Eu}(\text{L})_2(\text{H}_2\text{O})_2][3(\text{NO}_3)]$.

Eu(1)-O(6)	2.334(8)
Eu(1)-O(5)	2.337(8)
Eu(1)-O(3)	2.338(8)
Eu(1)-O(2)	2.358(9)
Eu(1)-O(7)	2.449(8)
Eu(1)-O(8)	2.476(8)
Eu(1)-O(4)	2.498(8)

Eu(1)-O(1)	2.544(8)
P(1)-O(2)	1.483(9)
P(1)-C(7)	1.789(13)
P(1)-C(13)	1.789(13)
P(1)-C(6)	1.805(13)
P(2)-O(3)	1.480(9)
P(2)-C(20)	1.780(12)
P(2)-C(26)	1.797(12)
P(2)-C(19)	1.815(11)
P(3)-O(5)	1.506(8)
P(3)-C(44)	1.802(13)
P(3)-C(38)	1.804(13)
P(3)-C(37)	1.812(13)
P(4)-O(6)	1.498(8)
P(4)-C(51)	1.798(12)
P(4)-C(57)	1.811(12)
P(4)-C(50)	1.812(11)
N(1)-O(1)	1.318(11)
N(1)-C(1)	1.351(14)
N(1)-C(5)	1.361(14)
N(2)-O(4)	1.327(11)
N(2)-C(32)	1.357(14)
N(2)-C(36)	1.358(14)
C(1)-C(2)	1.402(17)
C(1)-C(6)	1.488(16)
C(2)-C(3)	1.35(2)
C(3)-C(4)	1.39(2)
C(4)-C(5)	1.387(18)
C(5)-C(19)	1.516(16)
C(7)-C(8)	1.36(2)
C(7)-C(12)	1.39(2)
C(8)-C(9)	1.39(3)
C(9)-C(10)	1.36(3)
C(10)-C(11)	1.36(3)
C(11)-C(12)	1.37(3)
C(13)-C(14)	1.385(18)

C(13)-C(18)	1.387(19)
C(14)-C(15)	1.39(2)
C(15)-C(16)	1.39(3)
C(16)-C(17)	1.36(2)
C(17)-C(18)	1.36(2)
C(20)-C(25)	1.34(2)
C(20)-C(21)	1.35(3)
C(21)-C(22)	1.42(3)
C(22)-C(23)	1.33(3)
C(23)-C(24)	1.29(3)
C(24)-C(25)	1.39(3)
C(26)-C(31)	1.394(18)
C(26)-C(27)	1.396(18)
C(27)-C(28)	1.39(2)
C(28)-C(29)	1.33(2)
C(29)-C(30)	1.34(2)
C(30)-C(31)	1.38(2)
C(32)-C(33)	1.386(18)
C(32)-C(37)	1.502(16)
C(33)-C(34)	1.37(2)
C(34)-C(35)	1.35(2)
C(35)-C(36)	1.377(17)
C(36)-C(50)	1.503(15)
C(38)-C(43)	1.37(2)
C(38)-C(39)	1.38(2)
C(39)-C(40)	1.40(2)
C(40)-C(41)	1.35(2)
C(41)-C(42)	1.36(3)
C(42)-C(43)	1.40(2)
C(44)-C(45)	1.353(18)
C(44)-C(49)	1.407(18)
C(45)-C(46)	1.41(2)
C(46)-C(47)	1.39(3)
C(47)-C(48)	1.31(3)
C(48)-C(49)	1.38(2)
C(51)-C(52)	1.36(2)

C(51)-C(56)	1.369(18)
C(52)-C(53)	1.40(3)
C(53)-C(54)	1.36(3)
C(54)-C(55)	1.34(2)
C(55)-C(56)	1.39(2)
C(57)-C(58)	1.38(2)
C(57)-C(62)	1.390(19)
C(58)-C(59)	1.40(2)
C(59)-C(60)	1.36(3)
C(60)-C(61)	1.33(2)
C(61)-C(62)	1.40(2)
N(3)-O(9)	1.214(15)
N(3)-O(11)	1.217(17)
N(3)-O(10)	1.238(18)
N(4)-O(12)	1.211(17)
N(4)-O(13)	1.229(17)
N(4)-O(14)	1.239(16)
N(5)-O(15)	1.097(15)
N(5)-O(17)	1.119(16)
N(5)-O(16)	1.325(19)

O(6)-Eu(1)-O(5)	95.3(3)
O(6)-Eu(1)-O(3)	92.5(3)
O(5)-Eu(1)-O(3)	144.1(3)
O(6)-Eu(1)-O(2)	146.0(3)
O(5)-Eu(1)-O(2)	92.8(3)
O(3)-Eu(1)-O(2)	99.9(3)
O(6)-Eu(1)-O(7)	75.8(3)
O(5)-Eu(1)-O(7)	73.4(3)
O(3)-Eu(1)-O(7)	142.3(3)
O(2)-Eu(1)-O(7)	75.1(3)
O(6)-Eu(1)-O(8)	77.9(3)
O(5)-Eu(1)-O(8)	144.1(3)
O(3)-Eu(1)-O(8)	71.7(3)
O(2)-Eu(1)-O(8)	76.2(3)
O(7)-Eu(1)-O(8)	70.8(3)

O(6)-Eu(1)-O(4)	72.4(3)
O(5)-Eu(1)-O(4)	75.4(3)
O(3)-Eu(1)-O(4)	73.8(3)
O(2)-Eu(1)-O(4)	141.4(3)
O(7)-Eu(1)-O(4)	132.4(3)
O(8)-Eu(1)-O(4)	132.9(3)
O(6)-Eu(1)-O(1)	142.0(3)
O(5)-Eu(1)-O(1)	78.5(3)
O(3)-Eu(1)-O(1)	74.0(3)
O(2)-Eu(1)-O(1)	72.0(3)
O(7)-Eu(1)-O(1)	134.9(3)
O(8)-Eu(1)-O(1)	127.5(3)
O(4)-Eu(1)-O(1)	69.8(2)
O(2)-P(1)-C(7)	109.5(6)
O(2)-P(1)-C(13)	112.4(6)
C(7)-P(1)-C(13)	108.0(6)
O(2)-P(1)-C(6)	113.2(5)
C(7)-P(1)-C(6)	108.6(6)
C(13)-P(1)-C(6)	104.9(6)
O(3)-P(2)-C(20)	108.8(6)
O(3)-P(2)-C(26)	113.9(5)
C(20)-P(2)-C(26)	107.0(6)
O(3)-P(2)-C(19)	111.5(5)
C(20)-P(2)-C(19)	108.7(6)
C(26)-P(2)-C(19)	106.7(6)
O(5)-P(3)-C(44)	110.7(5)
O(5)-P(3)-C(38)	112.5(6)
C(44)-P(3)-C(38)	107.3(6)
O(5)-P(3)-C(37)	110.4(5)
C(44)-P(3)-C(37)	106.5(6)
C(38)-P(3)-C(37)	109.3(6)
O(6)-P(4)-C(51)	109.0(5)
O(6)-P(4)-C(57)	110.7(6)
C(51)-P(4)-C(57)	114.2(6)
O(6)-P(4)-C(50)	112.7(5)
C(51)-P(4)-C(50)	106.9(6)

C(57)-P(4)-C(50)	103.3(6)
O(1)-N(1)-C(1)	119.1(9)
O(1)-N(1)-C(5)	119.4(9)
C(1)-N(1)-C(5)	121.5(10)
O(4)-N(2)-C(32)	118.9(9)
O(4)-N(2)-C(36)	119.9(9)
C(32)-N(2)-C(36)	121.2(10)
N(1)-O(1)-Eu(1)	131.0(6)
P(1)-O(2)-Eu(1)	150.5(5)
P(2)-O(3)-Eu(1)	143.7(5)
N(2)-O(4)-Eu(1)	130.8(6)
P(3)-O(5)-Eu(1)	140.4(5)
P(4)-O(6)-Eu(1)	150.3(5)
N(1)-C(1)-C(2)	118.6(11)
N(1)-C(1)-C(6)	119.4(10)
C(2)-C(1)-C(6)	122.0(10)
C(3)-C(2)-C(1)	121.6(12)
C(2)-C(3)-C(4)	118.6(12)
C(5)-C(4)-C(3)	120.3(12)
N(1)-C(5)-C(4)	119.3(11)
N(1)-C(5)-C(19)	117.6(10)
C(4)-C(5)-C(19)	123.1(11)
C(1)-C(6)-P(1)	112.7(8)
C(8)-C(7)-C(12)	118.9(14)
C(8)-C(7)-P(1)	118.5(11)
C(12)-C(7)-P(1)	122.5(11)
C(7)-C(8)-C(9)	121.6(18)
C(10)-C(9)-C(8)	119(2)
C(9)-C(10)-C(11)	120(2)
C(10)-C(11)-C(12)	122(2)
C(11)-C(12)-C(7)	118.7(17)
C(14)-C(13)-C(18)	119.5(13)
C(14)-C(13)-P(1)	118.1(10)
C(18)-C(13)-P(1)	122.3(10)
C(13)-C(14)-C(15)	119.7(15)
C(14)-C(15)-C(16)	118.3(18)

C(17)-C(16)-C(15)	121.4(18)
C(18)-C(17)-C(16)	119.8(17)
C(17)-C(18)-C(13)	120.8(15)
C(5)-C(19)-P(2)	109.1(8)
C(25)-C(20)-C(21)	118.5(15)
C(25)-C(20)-P(2)	121.9(13)
C(21)-C(20)-P(2)	119.4(12)
C(20)-C(21)-C(22)	120(2)
C(23)-C(22)-C(21)	118(3)
C(24)-C(23)-C(22)	122(3)
C(23)-C(24)-C(25)	121(2)
C(20)-C(25)-C(24)	120.2(19)
C(31)-C(26)-C(27)	119.4(12)
C(31)-C(26)-P(2)	123.4(10)
C(27)-C(26)-P(2)	117.0(10)
C(28)-C(27)-C(26)	118.4(14)
C(29)-C(28)-C(27)	121.8(17)
C(28)-C(29)-C(30)	119.8(17)
C(29)-C(30)-C(31)	122.4(16)
C(30)-C(31)-C(26)	118.2(13)
N(2)-C(32)-C(33)	119.2(11)
N(2)-C(32)-C(37)	117.3(10)
C(33)-C(32)-C(37)	123.4(11)
C(34)-C(33)-C(32)	120.4(12)
C(35)-C(34)-C(33)	118.9(13)
C(34)-C(35)-C(36)	121.5(13)
N(2)-C(36)-C(35)	118.9(11)
N(2)-C(36)-C(50)	117.9(10)
C(35)-C(36)-C(50)	123.2(11)
C(32)-C(37)-P(3)	108.2(8)
C(43)-C(38)-C(39)	122.3(14)
C(43)-C(38)-P(3)	122.7(12)
C(39)-C(38)-P(3)	115.0(11)
C(38)-C(39)-C(40)	118.4(15)
C(41)-C(40)-C(39)	119.0(18)
C(40)-C(41)-C(42)	122.9(19)

C(41)-C(42)-C(43)	119.6(19)
C(38)-C(43)-C(42)	117.6(17)
C(45)-C(44)-C(49)	121.9(13)
C(45)-C(44)-P(3)	118.0(10)
C(49)-C(44)-P(3)	120.0(10)
C(44)-C(45)-C(46)	118.4(14)
C(47)-C(46)-C(45)	118.2(18)
C(48)-C(47)-C(46)	123(2)
C(47)-C(48)-C(49)	121(2)
C(48)-C(49)-C(44)	117.6(15)
C(36)-C(50)-P(4)	111.6(8)
C(52)-C(51)-C(56)	118.9(14)
C(52)-C(51)-P(4)	123.3(12)
C(56)-C(51)-P(4)	117.7(9)
C(51)-C(52)-C(53)	121(2)
C(54)-C(53)-C(52)	119(2)
C(55)-C(54)-C(53)	122(2)
C(54)-C(55)-C(56)	118.3(16)
C(51)-C(56)-C(55)	121.2(14)
C(58)-C(57)-C(62)	121.0(13)
C(58)-C(57)-P(4)	116.5(10)
C(62)-C(57)-P(4)	122.1(11)
C(57)-C(58)-C(59)	117.8(16)
C(60)-C(59)-C(58)	120.8(19)
C(61)-C(60)-C(59)	121(2)
C(60)-C(61)-C(62)	120.4(18)
C(57)-C(62)-C(61)	118.5(15)
O(9)-N(3)-O(11)	124.2(14)
O(9)-N(3)-O(10)	117.4(14)
O(11)-N(3)-O(10)	118.4(14)
O(12)-N(4)-O(13)	119.1(14)
O(12)-N(4)-O(14)	119.0(13)
O(13)-N(4)-O(14)	121.9(14)
O(15)-N(5)-O(17)	115.89(15)
O(15)-N(5)-O(16)	115.46(11)
O(17)-N(5)-O(16)	128.0

Table S32. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Eu}(\text{L})_2(\text{H}_2\text{O})_2][3(\text{NO}_3)]$. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^{*2} U^{11} + \dots + 2 h k a^{*} b^{*} U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Eu(1)	19(1)	22(1)	21(1)	0(1)	7(1)	0(1)
P(1)	29(2)	33(1)	23(1)	1(1)	3(1)	-8(1)
P(2)	27(1)	30(1)	36(2)	4(1)	15(1)	-2(1)
P(3)	24(1)	37(2)	39(2)	-3(1)	16(1)	1(1)
P(4)	25(1)	23(2)	32(1)	1(1)	6(1)	2(1)
N(1)	22(4)	30(5)	36(5)	7(4)	11(4)	7(4)
N(2)	30(4)	23(4)	29(5)	-4(4)	13(4)	-2(4)
O(1)	26(4)	40(4)	26(4)	1(3)	7(3)	-3(3)
O(2)	39(5)	37(5)	42(5)	-1(4)	7(4)	1(4)
O(3)	33(4)	37(5)	40(5)	-3(4)	18(4)	-6(4)
O(4)	27(4)	34(4)	29(4)	1(3)	12(3)	2(3)
O(5)	34(4)	35(4)	47(5)	-1(3)	22(4)	1(3)
O(6)	32(4)	29(4)	39(5)	1(3)	6(4)	3(3)
C(1)	24(5)	35(6)	32(6)	10(5)	-3(5)	3(5)
C(2)	36(7)	58(9)	33(7)	10(6)	-8(5)	-3(6)
C(3)	56(9)	65(9)	42(8)	28(7)	12(7)	-1(7)
C(4)	43(7)	39(7)	62(9)	21(6)	20(7)	-3(6)
C(5)	23(5)	41(7)	41(7)	6(5)	10(5)	1(5)
C(6)	15(5)	39(7)	35(6)	6(5)	-2(5)	3(5)
C(7)	46(7)	38(7)	27(6)	-2(5)	11(5)	-4(5)
C(13)	43(7)	40(7)	25(6)	-1(5)	7(5)	-14(5)
C(19)	24(5)	30(6)	48(7)	9(5)	17(5)	-1(4)
C(20)	54(8)	47(7)	39(7)	16(6)	34(6)	12(6)
C(26)	24(5)	44(7)	34(6)	8(5)	7(5)	-5(5)
C(32)	23(5)	34(6)	49(7)	-8(5)	14(5)	1(4)
C(33)	48(8)	56(8)	55(9)	-25(7)	23(7)	7(6)
C(34)	63(10)	92(13)	49(9)	-35(9)	22(8)	6(9)
C(35)	51(8)	57(9)	38(7)	-16(6)	4(6)	3(7)
C(36)	30(5)	25(5)	39(6)	-7(5)	12(5)	-3(4)
C(37)	27(5)	36(6)	44(7)	4(5)	14(5)	12(5)

C(38)	31(6)	57(8)	47(8)	-10(6)	17(6)	4(6)
C(44)	30(6)	41(7)	45(7)	-10(5)	20(5)	-4(5)
C(50)	21(5)	26(5)	37(6)	-1(5)	4(4)	-2(4)
C(51)	38(6)	35(6)	28(6)	1(5)	5(5)	-2(5)
C(57)	33(6)	37(7)	52(8)	4(6)	18(6)	8(5)
O(7)	36(4)	37(4)	39(5)	1(4)	20(4)	-3(4)
O(8)	48(5)	32(5)	51(5)	-1(4)	30(4)	1(4)
N(3)	39(6)	62(8)	54(7)	3(6)	27(6)	-7(5)
N(4)	61(7)	33(6)	45(6)	-2(5)	18(6)	-5(5)

X-ray Data Collection, Structure Solution and Refinement for $[\text{Nd}(\text{L})_2(\text{H}_2\text{O})_2]\text{[3(NO}_3\text{)]}$.

A colorless crystal of approximate dimensions 1.0 x 0.2 x 0.2 mm was mounted in a quartz capillary tube containing a small amount of its original mother liquor which was sealed using sealing wax to provide a solvent saturated environment. The crystal-containing capillary was then secured with mounting putty in a MiTeGen Reusable Base™ before being transferred to a Bruker D8 Venture diffractometer. The APEX3¹ program package was used to determine the unit-cell parameters and for data collection which was performed at room temperature (30 sec/frame scan time for a sphere of diffraction data). The raw frame data was processed using SAINT² and SADABS³ to yield the reflection data file. Subsequent calculations were carried out using the SHELXL⁴ program. The centrosymmetric monoclinic space group $C2/c$ was assigned and later determined to be correct.

The structure was solved by direct methods and refined on F^2 by full-matrix least-squares techniques. There was one disordered diethyl ether molecule of solvation present included using

multiple components with partial site-occupancy-factors. Additionally, all nitrate anions were disordered and included using multiple components with partial site-occupancy-factors. Hydrogen atoms were included using a riding model except in the case of the water ligands for which they could not be located. There were several high residuals present in the final difference-Fourier map. It was not possible to determine the nature of the residuals, although it was probable that another diethyl ether solvent molecule was present. The SQUEEZE routine in the PLATON⁵ program package was used to account for the electrons in the solvent accessible voids.

Table S33. Crystal data and structure refinement for [Nd(L)₂(H₂O)₂][3(NO₃)].

Empirical formula	[C ₆₂ H ₅₄ N ₂ NdO ₆ P ₄ (H ₂ O) ₂][3(NO ₃)](OC ₄ H ₁₀)	
Formula weight	1487.37	
Temperature	293(2) K	
Wavelength	0.56086 Å	
Crystal system	Monoclinic	
Space group	C2/c	
Unit cell dimensions	a = 17.3974(10) Å	α= 90°.
	b = 20.1764(14) Å	β= 95.506(2)°.
	c = 21.3801(13) Å	γ = 90°.
Volume	7470.2(8) Å ³	
Z	4	
Density (calculated)	1.323 mg/m ³	
Absorption coefficient	0.454 mm ⁻¹	
F(000)	3052	
Crystal size	1.0 x 0.2 x 0.2 mm ³	
Theta range for data collection	2.195 to 22.024°.	
Index ranges	-23 ≤ h ≤ 23, -26 ≤ k ≤ 26, -28 ≤ l ≤ 28	
Reflections collected	122935	
Independent reflections	9328 [R(int) = 0.0469]	
Completeness to theta = 19.665°	99.7 %	
Absorption correction	Semi-empirical from equivalents	

Max. and min. transmission	0.7447 and 0.6118
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	9328 / 0 / 427
Goodness-of-fit on F^2	1.083
Final R indices [$I > 2\text{sigma}(I)$]	$R_1 = 0.0344, wR_2 = 0.0966$
R indices (all data)	$R_1 = 0.0401, wR_2 = 0.1013$
Largest diff. peak and hole	0.755 and -0.981 e. \AA^{-3}

Table S34. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Nd}(\text{L})_2(\text{H}_2\text{O})_2][3(\text{NO}_3)]$. $U(\text{eq})$ is defined as one third of the trace of the orthogonalized U_{ij}^{eq} tensor.

	x	y	z	$U(\text{eq})$
Nd(1)	5000	8229(1)	7500	23(1)
P(1)	5460(1)	7310(1)	8986(1)	33(1)
P(2)	7119(1)	8496(1)	7402(1)	29(1)
O(1)	5378(1)	7889(1)	8546(1)	40(1)
O(2)	6272(1)	8619(1)	7399(1)	37(1)
O(3)	5840(1)	7250(1)	7318(1)	33(1)
O(4)	4868(1)	9237(1)	6808(1)	51(1)
N(1)	3736(1)	7088(1)	8144(1)	32(1)
C(1)	2968(1)	7244(1)	8096(1)	35(1)
C(2)	2527(2)	7060(2)	8565(2)	52(1)
C(3)	2852(2)	6714(2)	9083(2)	64(1)
C(4)	3623(2)	6556(2)	9118(2)	54(1)
C(5)	4069(2)	6742(1)	8650(1)	38(1)
C(6)	4915(2)	6600(1)	8676(1)	39(1)
C(7)	5108(2)	7538(2)	9719(1)	39(1)
C(8)	4742(3)	8137(2)	9767(2)	67(1)
C(9)	4497(4)	8324(3)	10340(3)	93(2)
C(10)	4616(3)	7938(3)	10854(2)	76(1)
C(11)	4967(2)	7332(2)	10806(2)	65(1)
C(12)	5217(2)	7127(2)	10244(1)	52(1)

C(13)	6447(2)	7062(2)	9138(1)	39(1)
C(14)	6672(2)	6426(2)	9311(2)	64(1)
C(15)	7457(2)	6285(2)	9433(2)	81(1)
C(16)	7999(2)	6771(2)	9390(2)	74(1)
C(17)	7778(2)	7398(2)	9220(2)	65(1)
C(18)	6996(2)	7552(2)	9102(1)	51(1)
C(19)	7357(1)	7622(1)	7467(1)	33(1)
C(20)	7451(1)	8790(1)	6685(1)	33(1)
C(21)	8216(2)	8730(2)	6552(1)	47(1)
C(22)	8444(2)	8989(2)	6005(2)	58(1)
C(23)	7921(2)	9307(2)	5589(2)	60(1)
C(24)	7164(2)	9367(2)	5710(2)	62(1)
C(25)	6925(2)	9107(2)	6262(2)	48(1)
C(26)	7639(1)	8901(1)	8056(1)	34(1)
C(27)	7226(2)	9308(2)	8421(2)	59(1)
C(28)	7602(2)	9653(3)	8915(2)	83(2)
C(29)	8378(2)	9586(2)	9054(2)	72(1)
C(30)	8796(2)	9182(2)	8701(2)	58(1)
C(31)	8428(2)	8835(2)	8200(1)	45(1)
N(2)	0	7788(2)	7500	50(1)
O(5)	0	8392(5)	7500	56(3)
O(5A)	-65(7)	8371(8)	7291(7)	60(3)
O(6)	621(2)	7479(2)	7566(2)	78(1)
N(3)	10000	10000	10000	50(1)
O(7)	10392(9)	10280(8)	9563(8)	85(4)
O(9)	9556(6)	10326(6)	10233(6)	77(3)
O(7A)	10026(8)	10497(7)	9799(7)	54(3)
O(9A)	9657(11)	9886(10)	10571(9)	68(5)
O(8)	9782(4)	10585(3)	10018(3)	64(1)
N(4)	4564(6)	9714(5)	5304(5)	91(2)
O(10)	5215(6)	9766(6)	5557(5)	103(3)
O(11)	4137(8)	9335(7)	5548(6)	43(3)
O(10A)	5428(7)	9953(7)	5265(6)	112(3)
O(11A)	4118(9)	9541(9)	5603(7)	44(4)
O(10B)	5604(10)	10324(10)	5331(9)	84(5)
O(11B)	4219(13)	9726(12)	5708(10)	34(5)

O(12)	5000	5515(3)	7500	98(2)
C(32)	4315(5)	5169(4)	7191(4)	82(2)
C(33)	3559(7)	5552(6)	7301(6)	101(4)
C(32A)	4428(9)	5236(8)	7609(8)	106(4)
C(33A)	3782(11)	5435(9)	7175(9)	96(5)

Table S35. Bond lengths [Å] and angles [°] for $[\text{Nd}(\text{L})_2(\text{H}_2\text{O})_2][\text{3}(\text{NO}_3)]$.

Nd(1)-O(1)	2.3704(17)
Nd(1)-O(1)#1	2.3704(17)
Nd(1)-O(2)#1	2.3783(17)
Nd(1)-O(2)	2.3783(17)
Nd(1)-O(3)	2.5104(17)
Nd(1)-O(3)#1	2.5105(18)
Nd(1)-O(4)	2.512(2)
Nd(1)-O(4)#1	2.512(2)
P(1)-O(1)	1.4990(19)
P(1)-C(13)	1.789(3)
P(1)-C(7)	1.796(3)
P(1)-C(6)	1.809(3)
P(2)-O(2)	1.4926(17)
P(2)-C(26)	1.789(3)
P(2)-C(20)	1.789(3)
P(2)-C(19)	1.814(3)
O(3)-N(1)#1	1.328(3)
N(1)-C(1)	1.367(3)
N(1)-C(5)	1.369(3)
C(1)-C(2)	1.371(4)
C(1)-C(19)#1	1.489(3)
C(2)-C(3)	1.384(5)
C(3)-C(4)	1.373(5)
C(4)-C(5)	1.376(4)
C(5)-C(6)	1.497(4)

C(7)-C(8)	1.374(5)
C(7)-C(12)	1.394(4)
C(8)-C(9)	1.388(6)
C(9)-C(10)	1.347(7)
C(10)-C(11)	1.374(7)
C(11)-C(12)	1.378(5)
C(13)-C(14)	1.381(5)
C(13)-C(18)	1.383(4)
C(14)-C(15)	1.395(5)
C(15)-C(16)	1.370(7)
C(16)-C(17)	1.362(6)
C(17)-C(18)	1.394(5)
C(20)-C(25)	1.380(4)
C(20)-C(21)	1.392(4)
C(21)-C(22)	1.376(4)
C(22)-C(23)	1.369(5)
C(23)-C(24)	1.372(5)
C(24)-C(25)	1.390(5)
C(26)-C(27)	1.380(4)
C(26)-C(31)	1.385(4)
C(27)-C(28)	1.378(5)
C(28)-C(29)	1.361(5)
C(29)-C(30)	1.367(5)
C(30)-C(31)	1.383(4)
N(2)-O(5)	1.218(12)
N(2)-O(6)#2	1.244(4)
N(2)-O(6)	1.244(4)
N(2)-O(5A)#2	1.258(16)
N(2)-O(5A)	1.259(15)
O(5A)-O(5A)#2	0.90(3)
N(3)-O(7A)	1.093(13)
N(3)-O(7A)#3	1.093(13)
N(3)-O(9)	1.162(11)
N(3)-O(9)#3	1.162(11)
N(3)-O(8)#3	1.242(6)
N(3)-O(8)	1.242(6)

N(3)-O(7)	1.333(16)
N(3)-O(7)#3	1.333(16)
N(3)-O(9A)#3	1.428(19)
N(3)-O(9A)	1.428(19)
O(7)-O(9)#3	1.297(18)
O(7)-O(8)	1.627(17)
O(9)-O(8)	0.822(11)
O(7A)-O(8)	0.686(14)
O(7A)-O(9A)#3	1.27(3)
N(4)-O(11B)	1.10(2)
N(4)-O(11A)	1.108(17)
N(4)-O(10)	1.212(12)
N(4)-O(11)	1.217(16)
N(4)-O(10B)#4	1.36(2)
N(4)-O(10A)#4	1.392(15)
N(4)-O(10A)	1.589(16)
O(10A)-O(10A)#4	1.79(2)
O(12)-C(32A)#1	1.186(16)
O(12)-C(32A)	1.186(16)
O(12)-C(32)	1.481(9)
O(12)-C(32)#1	1.481(9)
C(32)-C(33)	1.563(14)
C(32A)-C(33A)	1.44(2)
O(1)-Nd(1)-O(1)#1	146.35(9)
O(1)-Nd(1)-O(2)#1	100.75(7)
O(1)#1-Nd(1)-O(2)#1	90.27(7)
O(1)-Nd(1)-O(2)	90.27(7)
O(1)#1-Nd(1)-O(2)	100.75(7)
O(2)#1-Nd(1)-O(2)	141.42(9)
O(1)-Nd(1)-O(3)	78.78(6)
O(1)#1-Nd(1)-O(3)	74.87(6)
O(2)#1-Nd(1)-O(3)	146.63(6)
O(2)-Nd(1)-O(3)	71.63(6)
O(1)-Nd(1)-O(3)#1	74.87(6)
O(1)#1-Nd(1)-O(3)#1	78.78(6)

O(2)#1-Nd(1)-O(3)#1	71.63(6)
O(2)-Nd(1)-O(3)#1	146.63(6)
O(3)-Nd(1)-O(3)#1	76.18(8)
O(1)-Nd(1)-O(4)	142.28(7)
O(1)#1-Nd(1)-O(4)	71.19(7)
O(2)#1-Nd(1)-O(4)	75.69(7)
O(2)-Nd(1)-O(4)	73.29(7)
O(3)-Nd(1)-O(4)	124.56(7)
O(3)#1-Nd(1)-O(4)	134.97(7)
O(1)-Nd(1)-O(4)#1	71.19(7)
O(1)#1-Nd(1)-O(4)#1	142.28(7)
O(2)#1-Nd(1)-O(4)#1	73.28(7)
O(2)-Nd(1)-O(4)#1	75.69(7)
O(3)-Nd(1)-O(4)#1	134.97(7)
O(3)#1-Nd(1)-O(4)#1	124.56(7)
O(4)-Nd(1)-O(4)#1	71.91(11)
O(1)-P(1)-C(13)	111.30(13)
O(1)-P(1)-C(7)	109.25(13)
C(13)-P(1)-C(7)	108.38(13)
O(1)-P(1)-C(6)	111.90(12)
C(13)-P(1)-C(6)	107.71(13)
C(7)-P(1)-C(6)	108.19(14)
O(2)-P(2)-C(26)	110.60(12)
O(2)-P(2)-C(20)	109.97(12)
C(26)-P(2)-C(20)	109.78(12)
O(2)-P(2)-C(19)	112.24(11)
C(26)-P(2)-C(19)	106.78(12)
C(20)-P(2)-C(19)	107.35(12)
P(1)-O(1)-Nd(1)	145.02(12)
P(2)-O(2)-Nd(1)	150.65(12)
N(1)#1-O(3)-Nd(1)	133.04(14)
O(3)#1-N(1)-C(1)	119.7(2)
O(3)#1-N(1)-C(5)	119.3(2)
C(1)-N(1)-C(5)	120.9(2)
N(1)-C(1)-C(2)	119.6(2)
N(1)-C(1)-C(19)#1	118.1(2)

C(2)-C(1)-C(19)#1	122.3(2)
C(1)-C(2)-C(3)	120.5(3)
C(4)-C(3)-C(2)	118.9(3)
C(3)-C(4)-C(5)	120.8(3)
N(1)-C(5)-C(4)	119.3(3)
N(1)-C(5)-C(6)	117.9(2)
C(4)-C(5)-C(6)	122.8(3)
C(5)-C(6)-P(1)	109.87(19)
C(8)-C(7)-C(12)	119.4(3)
C(8)-C(7)-P(1)	119.3(2)
C(12)-C(7)-P(1)	121.3(2)
C(7)-C(8)-C(9)	119.3(4)
C(10)-C(9)-C(8)	121.8(4)
C(9)-C(10)-C(11)	119.1(4)
C(10)-C(11)-C(12)	120.8(4)
C(11)-C(12)-C(7)	119.6(3)
C(14)-C(13)-C(18)	120.0(3)
C(14)-C(13)-P(1)	123.4(2)
C(18)-C(13)-P(1)	116.5(2)
C(13)-C(14)-C(15)	119.2(4)
C(16)-C(15)-C(14)	120.6(4)
C(17)-C(16)-C(15)	120.3(4)
C(16)-C(17)-C(18)	120.1(4)
C(13)-C(18)-C(17)	119.8(4)
C(1)#1-C(19)-P(2)	111.73(18)
C(25)-C(20)-C(21)	119.5(3)
C(25)-C(20)-P(2)	117.8(2)
C(21)-C(20)-P(2)	122.6(2)
C(22)-C(21)-C(20)	120.0(3)
C(23)-C(22)-C(21)	120.2(3)
C(22)-C(23)-C(24)	120.6(3)
C(23)-C(24)-C(25)	119.8(3)
C(20)-C(25)-C(24)	119.9(3)
C(27)-C(26)-C(31)	119.3(3)
C(27)-C(26)-P(2)	117.5(2)
C(31)-C(26)-P(2)	123.1(2)

C(28)-C(27)-C(26)	120.0(3)
C(29)-C(28)-C(27)	120.4(4)
C(28)-C(29)-C(30)	120.5(3)
C(29)-C(30)-C(31)	119.9(3)
C(30)-C(31)-C(26)	119.9(3)
O(5)-N(2)-O(6)#2	120.1(2)
O(5)-N(2)-O(6)	120.1(2)
O(6)#2-N(2)-O(6)	119.9(4)
O(5)-N(2)-O(5A)#2	20.9(7)
O(6)#2-N(2)-O(5A)#2	123.7(6)
O(6)-N(2)-O(5A)#2	112.4(6)
O(6)#2-N(2)-O(5A)	112.4(6)
O(6)-N(2)-O(5A)	123.7(6)
O(5A)#2-N(2)-O(5A)	41.8(13)
O(5A)#2-O(5A)-N(2)	69.1(7)
O(9)-N(3)-O(9)#3	180.0(10)
O(9)-N(3)-O(8)#3	140.2(6)
O(9)#3-N(3)-O(8)#3	39.8(6)
O(7A)-N(3)-O(8)	33.4(8)
O(7A)#3-N(3)-O(8)	146.6(8)
O(9)-N(3)-O(8)	39.8(6)
O(9)#3-N(3)-O(8)	140.2(6)
O(8)#3-N(3)-O(8)	180.0
O(9)-N(3)-O(7)	117.8(9)
O(9)#3-N(3)-O(7)	62.2(9)
O(8)#3-N(3)-O(7)	101.7(7)
O(8)-N(3)-O(7)	78.3(7)
O(9)-N(3)-O(7)#3	62.2(9)
O(9)#3-N(3)-O(7)#3	117.8(9)
O(8)#3-N(3)-O(7)#3	78.3(7)
O(8)-N(3)-O(7)#3	101.7(7)
O(7)-N(3)-O(7)#3	180.0
O(9)-N(3)-O(9A)#3	128.7(9)
O(9)#3-N(3)-O(9A)#3	51.3(9)
O(8)#3-N(3)-O(9A)#3	88.4(8)
O(8)-N(3)-O(9A)#3	91.6(9)

O(7)-N(3)-O(9A)#3	18.1(10)
O(7)#3-N(3)-O(9A)#3	161.9(10)
O(7A)-N(3)-O(9A)	121.3(11)
O(7A)#3-N(3)-O(9A)	58.7(11)
O(8)#3-N(3)-O(9A)	91.6(9)
O(8)-N(3)-O(9A)	88.4(8)
O(9A)#3-N(3)-O(9A)	180.0
O(9)#3-O(7)-N(3)	52.4(8)
O(9)#3-O(7)-O(8)	100.7(11)
N(3)-O(7)-O(8)	48.4(6)
O(8)-O(9)-N(3)	75.3(10)
O(8)-O(9)-O(7)#3	140.0(15)
N(3)-O(9)-O(7)#3	65.4(9)
O(8)-O(7A)-N(3)	85.3(15)
O(8)-O(7A)-O(9A)#3	157(2)
N(3)-O(7A)-O(9A)#3	74.0(12)
O(7A)#3-O(9A)-N(3)	47.4(9)
O(7A)-O(8)-N(3)	61.3(13)
O(9)-O(8)-N(3)	64.9(9)
O(9)-O(8)-O(7)	117.9(11)
N(3)-O(8)-O(7)	53.3(6)
O(10)-N(4)-O(11)	116.5(12)
O(10)-N(4)-O(10B)#4	123.4(12)
O(11)-N(4)-O(10B)#4	108.1(13)
O(10)-N(4)-O(10A)#4	105.1(10)
O(11)-N(4)-O(10A)#4	137.4(12)
O(11A)-N(4)-O(10A)	147.5(14)
O(10A)#4-N(4)-O(10A)	73.6(9)
N(4)#4-O(10A)-N(4)	106.4(9)
N(4)#4-O(10A)-O(10A)#4	58.2(8)
N(4)-O(10A)-O(10A)#4	48.1(7)
C(32A)#1-O(12)-C(32A)	123.4(18)
C(32)-O(12)-C(32)#1	123.8(8)
O(12)-C(32)-C(33)	110.7(7)
O(12)-C(32A)-C(33A)	111.0(14)

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,y,-z+3/2 #2 -x,y,-z+3/2 #3 -x+2,-y+2,-z+2

#4 -x+1,-y+2,-z+1

Table S36. Anisotropic displacement parameters ($\text{\AA}^2 \times 10^3$) for $[\text{Nd}(\mathbf{L})_2(\text{H}_2\text{O})_2][3(\text{NO}_3)]$. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^{*2} U^{11} + \dots + 2 h k a^{*} b^{*} U^{12}]$

	U ¹¹	U ²²	U ³³	U ²³	U ¹³	U ¹²
Nd(1)	20(1)	26(1)	23(1)	0	2(1)	0
P(1)	38(1)	33(1)	28(1)	5(1)	-4(1)	0(1)
P(2)	22(1)	33(1)	32(1)	-2(1)	3(1)	-4(1)
O(1)	55(1)	35(1)	29(1)	6(1)	-6(1)	-3(1)
O(2)	23(1)	37(1)	51(1)	-1(1)	6(1)	-3(1)
O(3)	31(1)	35(1)	34(1)	-2(1)	6(1)	1(1)
O(4)	54(1)	40(1)	56(1)	16(1)	-1(1)	-3(1)
N(1)	32(1)	27(1)	35(1)	4(1)	-1(1)	-2(1)
C(1)	31(1)	34(1)	40(1)	4(1)	2(1)	-2(1)
C(2)	38(1)	60(2)	61(2)	18(2)	12(1)	-4(1)
C(3)	54(2)	79(3)	62(2)	31(2)	15(2)	-9(2)
C(4)	53(2)	56(2)	53(2)	24(2)	-3(1)	-11(2)
C(5)	40(1)	30(1)	42(1)	8(1)	-5(1)	-5(1)
C(6)	41(1)	30(1)	44(1)	6(1)	-9(1)	1(1)
C(7)	38(1)	46(2)	32(1)	4(1)	0(1)	-5(1)
C(8)	81(3)	70(2)	54(2)	13(2)	19(2)	28(2)
C(9)	117(4)	85(3)	81(3)	-2(3)	38(3)	41(3)
C(10)	79(3)	102(3)	53(2)	-4(2)	27(2)	5(3)
C(11)	71(2)	85(3)	39(2)	12(2)	10(2)	-9(2)
C(12)	61(2)	55(2)	39(2)	9(1)	2(1)	-4(2)
C(13)	38(1)	48(2)	32(1)	1(1)	-2(1)	-1(1)
C(14)	48(2)	52(2)	88(3)	1(2)	-11(2)	6(2)
C(15)	58(2)	69(3)	110(4)	-3(2)	-16(2)	22(2)
C(16)	40(2)	107(4)	72(3)	-7(2)	-5(2)	9(2)
C(17)	45(2)	98(3)	50(2)	5(2)	-1(1)	-16(2)
C(18)	47(2)	65(2)	40(2)	8(1)	-1(1)	-10(1)
C(19)	28(1)	34(1)	37(1)	-1(1)	-2(1)	0(1)

C(20)	32(1)	35(1)	33(1)	1(1)	4(1)	-4(1)
C(21)	34(1)	62(2)	46(2)	10(1)	9(1)	3(1)
C(22)	53(2)	70(2)	56(2)	12(2)	27(2)	4(2)
C(23)	77(2)	62(2)	43(2)	13(2)	18(2)	-3(2)
C(24)	64(2)	70(2)	49(2)	20(2)	-3(2)	1(2)
C(25)	39(1)	55(2)	48(2)	10(1)	1(1)	2(1)
C(26)	32(1)	38(1)	33(1)	-2(1)	3(1)	-6(1)
C(27)	36(1)	76(2)	64(2)	-32(2)	3(1)	-1(1)
C(28)	57(2)	110(4)	81(3)	-60(3)	0(2)	5(2)
C(29)	58(2)	87(3)	67(2)	-36(2)	-15(2)	-3(2)
C(30)	41(2)	73(2)	57(2)	-10(2)	-14(1)	-4(2)
C(31)	34(1)	54(2)	48(2)	-9(1)	-2(1)	2(1)

References and Definitions

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Definitions:

$$wR2 = [\sum[w(F_o^2 - F_c^2)^2] / \sum[w(F_o^2)^2]]^{1/2}$$

$$R1 = \Sigma |F_o - F_c| / \Sigma |F_o|$$

Goof = S = $[\sum w(F_o^2 - F_c^2)^2] / (n-p)]^{1/2}$ where n is the number of reflections and p is the total number of parameters refined.