Table to be deposited

Table S1. Atomic coordinates and equivalent isotropic displacement parameters (Å^2) for Complex [OsCl{κ^2-*C,N*-=CHCH(Ph)N=CMe$_2$}(CO)(P^{i}Pr$_3$)$_2$][CF$_3$SO$_3$] (**3**).

Atom	X/*a*	Y/*b*	Z/*c*	U_{eq}[a]/U_{iso}[b]
Os	0.30203(2)	0.013791(19)	0.782794(18)	0.03715(12)
Cl	0.45996(16)	-0.02679(16)	0.87025(14)	0.0549(6)
P1	0.39055(17)	-0.02160(15)	0.67477(13)	0.0454(5)
P2	0.25130(16)	0.05837(14)	0.90573(13)	0.0409(5)
S[b]	0.0860(3)	-0.2688(2)	0.99786(16)	0.0803(9)
F1[b]	0.0045(10)	-0.4133(6)	0.9798(9)	0.087(6)
F2[b]	-0.0396(11)	-0.3304(9)	0.8811(6)	0.103(6)
F3[b]	-0.0965(9)	-0.3122(10)	0.9887(11)	0.165(9)
F1B[b]	-0.007(2)	-0.3211(15)	0.8624(10)	0.084(11)
F2B[b]	-0.010(2)	-0.4046(13)	0.9594(16)	0.097(17)
F3B[b]	0.1268(19)	-0.3887(14)	0.9126(15)	0.105(12)
F1C[b]	-0.0739(12)	-0.3065(10)	0.8954(9)	0.073(6)
F2C[b]	0.0213(14)	-0.4082(8)	0.9410(17)	0.177(19)
F3C[b]	-0.0713(15)	-0.3494(17)	1.0145(10)	0.192(18)
O2[b]	0.1042(12)	-0.2852(10)	1.0806(5)	0.085(7)
O3[b]	0.1632(10)	-0.2973(12)	0.9582(11)	0.129(10)
O4[b]	0.0485(13)	-0.1891(6)	0.9753(12)	0.120(9)
O2B[b]	0.1333(14)	-0.2128(10)	0.9540(9)	0.025(5)
O3B[b]	0.1525(18)	-0.3086(12)	1.0614(11)	0.062(17)
O4B[b]	-0.0064(13)	-0.2415(15)	1.0177(16)	0.18(3)
O2C[b]	0.0311(11)	-0.1948(8)	1.0131(13)	0.129(10)
O3C[b]	0.1411(11)	-0.2575(12)	0.9374(8)	0.085(6)
O4C[b]	0.1361(13)	-0.3075(10)	1.0677(8)	0.074(10)
C31[b]	-0.0202(7)	-0.3387(6)	0.9576(5)	0.107(6)
C31B[b]	0.0453(15)	-0.3544(9)	0.9263(10)	0.080(17)
O1	0.3334(6)	0.1895(4)	0.7466(4)	0.071(2)
N	0.2270(5)	-0.1081(4)	0.7786(4)	0.0383(15)
C1	0.2389(7)	-0.1846(5)	0.7951(5)	0.046(2)
C2	0.1723(8)	-0.2498(6)	0.7555(6)	0.063(3)
C3	0.3235(8)	-0.2124(6)	0.8574(5)	0.059(3)
C4	0.1754(7)	0.0099(6)	0.7181(5)	0.044(2)
C5	0.1419(6)	-0.0776(6)	0.7193(5)	0.043(2)
C6	0.0360(6)	-0.0878(5)	0.7351(5)	0.043(2)
C7	0.0140(7)	-0.1213(6)	0.8034(5)	0.051(2)
C8	-0.0846(8)	-0.1237(6)	0.8142(6)	0.061(3)
C9	-0.1599(8)	-0.0955(8)	0.7575(7)	0.079(4)
C10	-0.1365(8)	-0.0633(10)	0.6890(7)	0.096(5)
C11	-0.0406(7)	-0.0590(8)	0.6783(6)	0.069(3)
C12	0.5242(8)	0.0043(7)	0.6970(7)	0.068(3)
C13	0.5504(9)	0.0885(8)	0.7347(8)	0.096(4)
C14	0.5779(9)	-0.0059(10)	0.6267(9)	0.110(5)
C15	0.3858(8)	-0.1320(6)	0.6526(6)	0.059(3)
C16	0.3944(10)	-0.1595(8)	0.5680(7)	0.086(4)
C17	0.4619(10)	-0.1800(7)	0.7123(7)	0.082(4)
C18	0.3431(8)	0.0260(7)	0.5775(6)	0.066(3)
C19	0.2337(9)	0.0017(9)	0.5465(7)	0.092(4)
C20	0.3538(12)	0.1173(8)	0.5718(7)	0.104(5)
C21	0.3529(7)	0.1098(5)	0.9757(5)	0.050(2)
C22	0.4182(8)	0.1699(7)	0.9398(6)	0.068(3)
C23	0.3166(8)	0.1495(7)	1.0463(6)	0.071(3)
C24	0.1425(8)	0.1285(6)	0.8916(6)	0.059(3)
C25	0.0485(7)	0.0918(7)	0.8458(7)	0.069(3)
C26	0.1620(9)	0.2117(7)	0.8568(8)	0.080(3)

C27	0.2147(7)	-0.0288(6)	0.9636(5)	0.048(2)
C28	0.1404(9)	-0.0102(7)	1.0163(8)	0.079(4)
C29	0.3041(8)	-0.0734(7)	1.0108(6)	0.066(3)
C30	0.3238(7)	0.1218(6)	0.7629(6)	0.054(2)

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[a] Equivalent isotropic *U* defined as one third of the trace of the orthogonalized *Uij* tensor.
b The triflate anion was observed severely disordered, and was refined with three sites for O and F atoms, two sites for C and one for the S atom with complementary occupancy factors and restrained geometry.

Table to be deposited

Table S2. Anisotropic displacement coefficients U_{ij} (Å^2) for the non-hydrogen atoms for the complex [OsCl{κ^2-*C,N*-=CHCH(Ph)N=CMe$_2$}(CO)(P^{i}Pr$_3$)$_2$][CF$_3$SO$_3$] (**3**).

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Os	0.03655(18)	0.04016(19)	0.03630(18)	0.00111(15)	0.01082(12)	0.00172(15)
Cl	0.0390(11)	0.0720(16)	0.0513(13)	0.0011(11)	0.0025(10)	0.0032(11)
P1	0.0447(12)	0.0513(13)	0.0445(12)	0.0066(10)	0.0194(10)	0.0083(11)
P2	0.0412(12)	0.0435(13)	0.0391(11)	-0.0064(9)	0.0102(9)	-0.0011(10)
S	0.096(2)	0.084(2)	0.0566(16)	0.0111(15)	0.0034(16)	-0.0282(18)
O1	0.085(5)	0.049(4)	0.085(5)	0.006(4)	0.032(4)	-0.015(4)
N	0.040(4)	0.043(4)	0.034(3)	-0.002(3)	0.012(3)	0.002(3)
C1	0.051(5)	0.047(5)	0.043(5)	-0.007(4)	0.015(4)	0.004(4)
C2	0.078(7)	0.049(6)	0.063(6)	-0.008(5)	0.017(6)	-0.004(5)
C3	0.069(7)	0.052(6)	0.055(6)	0.011(5)	0.009(5)	0.013(5)
C4	0.047(5)	0.056(5)	0.034(4)	0.004(4)	0.020(4)	0.012(4)
C5	0.047(5)	0.056(5)	0.027(4)	-0.003(4)	0.006(4)	-0.002(4)
C6	0.039(5)	0.054(5)	0.037(4)	-0.005(4)	0.010(4)	-0.003(4)
C7	0.052(6)	0.057(6)	0.045(5)	-0.003(4)	0.009(4)	0.002(4)
C8	0.056(6)	0.075(7)	0.058(6)	-0.001(5)	0.026(5)	-0.005(5)
C9	0.036(6)	0.127(11)	0.078(8)	0.001(7)	0.019(6)	-0.001(6)
C10	0.042(6)	0.170(14)	0.075(8)	0.028(9)	0.004(6)	0.023(8)
C11	0.049(6)	0.107(9)	0.048(6)	0.015(6)	0.002(5)	0.004(6)
C12	0.050(6)	0.088(9)	0.071(7)	0.010(6)	0.023(5)	0.006(5)
C13	0.076(9)	0.108(11)	0.112(11)	0.008(9)	0.041(8)	-0.037(8)
C14	0.057(7)	0.184(16)	0.101(10)	0.001(10)	0.051(8)	0.006(9)
C15	0.065(7)	0.052(6)	0.069(7)	0.000(5)	0.035(5)	0.007(5)
C16	0.105(10)	0.081(8)	0.083(8)	-0.021(7)	0.049(7)	0.022(7)
C17	0.109(10)	0.066(7)	0.075(8)	0.021(6)	0.030(7)	0.035(7)
C18	0.077(7)	0.075(7)	0.052(6)	0.014(5)	0.029(5)	0.027(6)
C19	0.079(8)	0.152(13)	0.047(6)	0.022(7)	0.017(6)	0.023(8)
C20	0.172(15)	0.076(9)	0.070(8)	0.034(7)	0.041(9)	0.031(9)
C21	0.047(5)	0.044(5)	0.055(5)	-0.012(4)	0.004(4)	-0.001(4)
C22	0.074(7)	0.065(7)	0.065(7)	-0.008(5)	0.010(6)	-0.030(6)
C23	0.079(8)	0.073(7)	0.061(7)	-0.029(6)	0.016(6)	-0.015(6)
C24	0.063(7)	0.064(6)	0.055(6)	-0.010(5)	0.018(5)	0.015(5)
C25	0.045(6)	0.085(8)	0.076(7)	-0.020(6)	0.006(5)	0.024(5)
C26	0.081(8)	0.060(7)	0.104(9)	0.011(6)	0.027(7)	0.024(6)
C27	0.052(5)	0.057(6)	0.039(4)	-0.007(4)	0.015(4)	-0.007(4)
C28	0.070(7)	0.083(8)	0.100(9)	0.005(7)	0.057(7)	-0.006(6)
C29	0.068(7)	0.064(7)	0.068(7)	0.012(5)	0.015(6)	-0.001(5)
C30	0.056(6)	0.056(6)	0.055(6)	-0.001(5)	0.019(5)	-0.005(5)

* The anisotropic displacement factor exponent takes the form:

$-2\pi^2 (h^2 a^{*2} U_{11} + ... + 2\,h\,k\,a^*\,b^*\,U_{12})$

Table to be deposited

Table S3. Hydrogen atom coordinates* and isotropic displacement coefficient ($Å^2$) for the compound $[OsCl\{\kappa^2\text{-}C,N\text{-}=CHCH(Ph)N=CMe_2\}(CO)(P^iPr_3)_2][CF_3SO_3]$ (**3**).

Atom	X/*a*	Y/*b*	Z/*c*	*U*
H2A	0.1065	-0.2286	0.7395	0.094
H2B	0.1709	-0.2939	0.7910	0.094
H2C	0.1966	-0.2685	0.7105	0.094
H3A	0.3687	-0.1684	0.8720	0.088
H3B	0.3574	-0.2563	0.8377	0.088
H3C	0.2983	-0.2303	0.9023	0.088
H4	0.130(6)	0.041(5)	0.690(5)	0.04(2)
H5	0.153(5)	-0.099(4)	0.664(4)	0.020(17)
H7	0.0644	-0.1418	0.8415	0.062
H8	-0.0991	-0.1447	0.8603	0.074
H9	-0.2253	-0.0977	0.7644	0.095
H10	-0.1871	-0.0445	0.6501	0.115
H11	-0.0267	-0.0365	0.6327	0.083
H12	0.5548	-0.0353	0.7361	0.082
H13A	0.6210	0.0940	0.7485	0.144
H13B	0.5219	0.0935	0.7807	0.144
H13C	0.5243	0.1300	0.6981	0.144
H14A	0.5427	0.0236	0.5827	0.164
H14B	0.5800	-0.0621	0.6135	0.164
H14C	0.6442	0.0147	0.6405	0.164
H15	0.3205	-0.1503	0.6602	0.071
H16A	0.4551	-0.1393	0.5555	0.128
H16B	0.3394	-0.1386	0.5312	0.128
H16C	0.3942	-0.2175	0.5654	0.128
H17A	0.4448	-0.2363	0.7093	0.123
H17B	0.4612	-0.1603	0.7640	0.123
H17C	0.5269	-0.1730	0.7005	0.123
H18	0.3818	0.0026	0.5411	0.079
H19A	0.1913	0.0311	0.5744	0.138
H19B	0.2259	-0.0552	0.5540	0.138
H19C	0.2165	0.0142	0.4919	0.138
H20A	0.3332	0.1340	0.5185	0.156
H20B	0.4217	0.1322	0.5898	0.156
H20C	0.3131	0.1433	0.6036	0.156
H21	0.3978	0.0664	0.9982	0.059
H22A	0.3787	0.2151	0.9179	0.102
H22B	0.4452	0.1433	0.8996	0.102
H22C	0.4712	0.1887	0.9798	0.102
H23A	0.3724	0.1704	1.0823	0.106
H23B	0.2834	0.1097	1.0721	0.106
H23C	0.2717	0.1928	1.0281	0.106
H24	0.1292	0.1395	0.9439	0.071
H25A	-0.0074	0.1232	0.8539	0.104
H25B	0.0418	0.0373	0.8631	0.104
H25C	0.0516	0.0915	0.7912	0.104
H26A	0.1808	0.2040	0.8070	0.121
H26B	0.2143	0.2388	0.8914	0.121
H26C	0.1028	0.2438	0.8503	0.121
H27	0.1819	-0.0679	0.9250	0.058
H28A	0.1210	-0.0596	1.0381	0.118
H28B	0.0831	0.0155	0.9863	0.118
H28C	0.1704	0.0252	1.0577	0.118
H29A	0.3314	-0.0421	1.0560	0.100

H29B	0.3534	-0.0809	0.9790	0.100
H29C	0.2833	-0.1251	1.0268	0.100

* The hydrogen atoms were included in observed or calculated positions and refined riding on their respective carbon atoms.

Table to be deposited

Table S4. Full experimental details for the X-Ray analysis of the [OsCl{κ^2-*C,N*-=CHCH(Ph)N=CMe_2}(CO)(P^iPr_3)$_2$][CF_3SO_3] (**3**).

Crystal data:

Formula	$C_{31}H_{55}ClF_3NO_4OsP_2S$
Molecular weight	882.41
Crystal habit	irregular block
Size(mm)	0.44 x 0.36 x 0.29
Symmetry	monoclinic, P21/c
Unit cell dimensions	13.748(2), 16.540(3), 17.438(3) Å 90, 100.795(6), 90 °
Packing: V (Å^3), Z	3895.1(11), 4
D_{calc}(g cm^{-3}), F (000)	1.505, 1784

Experimental data:

Radiation and technique	Bruker Siemens-P4 Mo-K_α (λ = 0.71073 Å)
Monochromator	Graphite oriented
Range	($5 < 2\theta < 50°$)
Number of reflections:	
measured	7644 (h: -16, 1; k: -19, 1; l: -20, 20)
unique	6803 (R_{int} = 0.0374)
Absorption correction	psi scan*
Max. and min. trans. fact.	0.259, 0.225
μ (mm^{-1})	3.525
Temperature (K)	296.0(2)

Table to be deposited

Table S4. Full experimental details for the X-Ray analysis of the [OsCl{κ^2-*C,N*-=CHCH(Ph)N=CMe_2}(CO)(P^iPr_3)$_2$][CF_3SO_3] (**3**).

Solution and refinement:	SHELX97 dos/win95/nt version
Solution mode	Patterson
Refinement	Full-matrix least-squares on F'^2s.
	All reflections.
Hydrogen atoms	From calculated positions. Refined riding on C
	atoms with thermal parameters related to bonded atoms.
No. parameters/restrains	439/59
Weighting scheme	$w^{-1}=[\sigma^2(F_o^2)+(0.0541P)^2+0P]$
	where $P=((\text{Max } F_o^2,0)+2F_c^2)/3$
ΔF final (max)	1.157 e/Å^3 (close to Os atom)
ΔF final (min)	-1.084 e/Å^3
Max/Mean shift/esd	0.436, 0.016
$\omega R2(F^2, \text{ all data})^a$	0.1321
$R1(F, F_o > 4.0\ \sigma F)^b$	0.0645
Goodness-of-Fit[c]	1.021
Data-to-Parameter Ratio	15.5:1

Atomic scattering factors from the International Tables for X-Ray Crystallography; Vol C (1992). Anomalous dispersion is implemented by the program.

[a] $wR2(F^2) = \{\Sigma[w(F_o^2-F_c^2)^2]/\Sigma[w(F_o^2)^2]\}^{1/2}$ [b] $R1(F) = \Sigma||F_o|-|F_c||/\Sigma|F_o|$. [c] $S = \{\Sigma[w(F_o^2-F_c^2)^2]/(n-p)\}^{1/2}$, where n is the number of observed reflections, and p is the number of parameters refined.

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* Shelxtl 5.0., Bruker-Siemens Analytical X-ray Instruments, Inc: Madison, WI.

Table to be deposited

Table S5. Bond Lenghts (Å) and angles (deg) for the complex [OsCl{κ^2-*C,N*-=CHCH(Ph)N=CMe_2}(CO)(P^iPr_3)$_2$][CF_3SO_3] (**3**).

Os C30 1.854(10)	O4 O4B 1.44(3)
Os C4 1.890(10)	O2B O3C 0.81(2)
Os N 2.260(7)	O4B O2C 0.94(3)
Os P2 2.487(2)	O4B C31 1.91(2)
Os P1 2.496(2)	C31 C31B 1.17(2)
Os Cl 2.500(2)	O1 C30 1.169(11)
P1 C12 1.856(11)	N C1 1.301(11)
P1 C15 1.865(10)	N C5 1.497(10)
P1 C18 1.874(10)	C1 C2 1.497(13)
P2 C24 1.873(10)	C1 C3 1.507(12)
P2 C21 1.878(9)	C4 C5 1.520(13)
P2 C27 1.882(9)	C5 C6 1.542(12)
S O3C 1.421(9)	C6 C11 1.387(12)
S O2B 1.432(10)	C6 C7 1.398(12)
S O4C 1.433(9)	C7 C8 1.404(13)
S O2 1.443(9)	C8 C9 1.373(15)
S O4 1.443(10)	C9 C10 1.399(16)
S O3 1.451(10)	C10 C11 1.367(14)
S O4B 1.450(10)	C12 C13 1.553(16)
S O3B 1.456(10)	C12 C14 1.555(15)
S O2C 1.488(10)	C15 C17 1.549(14)
S C31 1.891(9)	C15 C16 1.570(14)
S C31B 1.900(12)	C18 C20 1.521(16)
F1 F2C 0.76(3)	C18 C19 1.552(17)
F1 C31 1.319(9)	C21 C22 1.547(13)
F1 C31B 1.53(2)	C21 C23 1.557(13)
F1 F3C 1.67(3)	C24 C25 1.513(14)
F2 F1B 0.63(4)	C24 C26 1.547(15)
F2 F1C 0.69(2)	C27 C28 1.529(12)
F2 C31 1.317(9)	C27 C29 1.533(13)
F2 C31B 1.34(3)	
F2 F2C 1.77(2)	C30 Os C4 94.8(4)
F3 F3C 0.80(3)	C30 Os N 160.4(4)
F3 C31 1.342(9)	C4 Os N 65.6(3)
F3 O4B 1.71(3)	C30 Os P2 87.5(3)
F3 F1C 1.72(2)	C4 Os P2 98.6(2)
F1B F1C 1.20(4)	N Os P2 95.33(16)
F1B C31B 1.326(10)	C30 Os P1 88.1(3)
F1B C31 1.73(3)	C4 Os P1 93.8(2)
F2B F2C 0.58(4)	N Os P1 92.86(17)
F2B C31 1.10(3)	P2 Os P1 167.17(8)
F2B C31B 1.324(10)	C30 Os Cl 102.7(3)
F2B F3C 1.67(4)	C4 Os Cl 162.3(3)
F3B C31B 1.318(10)	N Os Cl 96.84(18)
F3B F2C 1.65(4)	P2 Os Cl 84.58(8)
F3B O3 1.74(2)	P1 Os Cl 84.66(8)
F1C C31 1.305(9)	C12 P1 C15 105.3(5)
F1C C31B 1.81(3)	C12 P1 C18 104.8(5)
F2C C31B 1.00(3)	C15 P1 C18 103.4(5)
F2C C31 1.340(10)	C12 P1 Os 113.1(4)
F3C C31 1.330(10)	C15 P1 Os 112.5(3)
O2 O4C 0.65(3)	C18 P1 Os 116.6(3)
O2 O3B 0.89(3)	C24 P2 C21 106.5(4)
O3 O3C 0.78(2)	C24 P2 C27 105.0(4)
O3 O2B 1.46(3)	C21 P2 C27 103.9(4)
O3 C31B 1.87(2)	C24 P2 Os 114.6(3)
O4 O2C 0.75(3)	C21 P2 Os 113.6(3)
O4 O2B 1.35(3)	C27 P2 Os 112.4(3)

O3C S O2B 32.9(9)
O3C S O4C 116.8(7)
O2B S O4C 123.3(11)
O3C S O2 138.6(9)
O2B S O2 130.1(10)
O4C S O2 25.9(11)
O3C S O4 83.7(10)
O2B S O4 55.8(11)
O4C S O4 136.9(10)
O2 S O4 115.5(7)
O3C S O3 31.7(10)
O2B S O3 60.6(11)
O4C S O3 89.0(11)
O2 S O3 114.3(7)
O4 S O3 115.0(7)
O3C S O4B 138.5(15)
O2B S O4B 115.6(9)
O4C S O4B 103.9(16)
O2 S O4B 79.4(14)
O4 S O4B 59.8(13)
O3 S O4B 165.4(14)
O3C S O3B 106.2(15)
O2B S O3B 114.6(8)
O4C S O3B 10.7(16)
O2 S O3B 35.6(14)
O4 S O3B 139.6(12)
O3 S O3B 78.7(14)
O4B S O3B 114.4(8)
O3C S O2C 112.6(6)
O2B S O2C 81.9(11)
O4C S O2C 113.3(6)
O2 S O2C 88.5(10)
O4 S O2C 29.5(10)
O3 S O2C 142.6(10)
O4B S O2C 37.3(11)
O3B S O2C 120.2(15)
O3C S C31 106.6(6)
O2B S C31 126.4(8)
O4C S C31 104.9(6)
O2 S C31 103.5(6)
O4 S C31 104.2(6)
O3 S C31 102.0(6)
O4B S C31 68.2(10)
O3B S C31 109.8(10)
O2C S C31 100.8(6)
O3C S C31B 75.1(8)
O2B S C31B 103.8(7)
O4C S C31B 105.1(10)
O2 S C31B 119.4(9)
O4 S C31B 117.1(9)
O3 S C31B 66.2(9)
O4B S C31B 103.0(8)
O3B S C31B 103.3(7)
O2C S C31B 129.3(10)
C31 S C31B 35.8(7)
F2C F1 C31 74.9(10)
F2C F1 C31B 34.7(10)
C31 F1 C31B 47.7(9)
F2C F1 F3C 124.5(13)
C31 F1 F3C 51.1(7)
C31B F1 F3C 98.2(10)
F1B F2 F1C 131(4)
F1B F2 C31 121.7(18)
F1C F2 C31 73.7(10)
F1B F2 C31B 75(2)
F1C F2 C31B 122.1(13)
C31 F2 C31B 52.0(10)
F1B F2 F2C 100(3)
F1C F2 F2C 118.7(15)
C31 F2 F2C 48.9(7)
C31B F2 F2C 34.2(10)
F3C F3 C31 71.8(11)
F3C F3 O4B 98.4(18)
C31 F3 O4B 76.4(8)
F3C F3 F1C 115.9(14)
C31 F3 F1C 48.7(6)
O4B F3 F1C 89.6(12)
F2 F1B F1C 26(2)
F2 F1B C31B 78(2)
F1C F1B C31B 91.2(16)
F2 F1B C31 40.4(13)
F1C F1B C31 48.9(10)
C31B F1B C31 42.3(10)
F2C F2B C31 101(3)
F2C F2B C31B 45(2)
C31 F2B C31B 56.6(12)
F2C F2B F3C 151.4(19)
C31 F2B F3C 52.7(15)
C31B F2B F3C 107.4(13)
C31B F3B F2C 37.3(10)
C31B F3B O3 74.0(9)
F2C F3B O3 103.2(9)
F2 F1C F1B 23.3(19)
F2 F1C C31 75.6(10)
F1B F1C C31 87.2(14)
F2 F1C F3 124.3(14)
F1B F1C F3 137.4(12)
C31 F1C F3 50.5(7)
F2 F1C C31B 38.9(9)
F1B F1C C31B 47.2(10)
C31 F1C C31B 40.0(8)
F3 F1C C31B 90.4(9)
F2B F2C F1 31(4)
F2B F2C C31B 111(3)
F1 F2C C31B 119.7(17)
F2B F2C C31 54(2)
F1 F2C C31 71.9(11)
C31B F2C C31 57.6(14)
F2B F2C F3B 157(2)
F1 F2C F3B 136(2)
C31B F2C F3B 53.0(17)
C31 F2C F3B 109.3(13)
F2B F2C F2 86(4)
F1 F2C F2 114.7(13)
C31B F2C F2 48.9(16)
C31 F2C F2 47.8(6)
F3B F2C F2 91.5(18)
F3 F3C C31 73.3(10)
F3 F3C F2B 108.1(18)
C31 F3C F2B 41.1(10)
F3 F3C F1 121.0(13)
C31 F3C F1 50.5(8)
F2B F3C F1 13.7(14)
O4C O2 O3B 9(2)
O4C O2 S 76.2(11)
O3B O2 S 73.0(9)
O3C O3 O2B 24.5(13)
O3C O3 S 72.1(9)
O2B O3 S 59.1(7)
O3C O3 F3B 118(2)
O2B O3 F3B 139.3(17)
S O3 F3B 109.1(12)

O3C O3 C31B 92.7(16)
O2B O3 C31B 104.4(12)
S O3 C31B 68.5(7)
F3B O3 C31B 42.7(6)
O2C O4 O2B 130.1(18)
O2C O4 O4B 35.7(14)
O2B O4 O4B 122.0(9)
O2C O4 S 78.6(10)
O2B O4 S 61.7(7)
O4B O4 S 60.4(8)
O3C O2B O4 123.0(16)
O3C O2B S 72.8(9)
O4 O2B S 62.5(7)
O3C O2B O3 23.7(15)
O4 O2B O3 121.2(8)
S O2B O3 60.3(6)
O2 O3B S 71.4(10)
O2C O4B O4 27.6(14)
O2C O4B S 73.5(9)
O4 O4B S 59.9(7)
O2C O4B F3 156(3)
O4 O4B F3 133(2)
S O4B F3 109.6(13)
O2C O4B C31 130.1(18)
O4 O4B C31 103.4(13)
S O4B C31 66.9(8)
F3 O4B C31 43.1(7)
O4 O2C O4B 117(2)
O4 O2C S 71.9(9)
O4B O2C S 69.1(8)
O3 O3C O2B 132(3)
O3 O3C S 76.2(10)
O2B O3C S 74.3(9)
O2 O4C S 77.9(11)
F2B C31 C31B 71.5(16)
F2B C31 F1C 119(2)
C31B C31 F1C 93.9(15)
F2B C31 F2 97.6(18)
C31B C31 F2 65.0(13)
F1C C31 F2 30.7(10)
F2B C31 F1 15.9(17)
C31B C31 F1 75.5(12)
F1C C31 F1 134.4(11)
F2 C31 F1 112.8(8)
F2B C31 F3C 86.2(19)
C31B C31 F3C 151.7(17)
F1C C31 F3C 112.3(9)
F2 C31 F3C 137.2(11)
F1 C31 F3C 78.4(14)
F2B C31 F3 115.0(18)
C31B C31 F3 173.1(14)
F1C C31 F3 80.8(11)
F2 C31 F3 110.5(8)
F1 C31 F3 111.4(8)
F3C C31 F3 34.9(13)
F2B C31 F2C 25.2(18)
C31B C31 F2C 46.4(13)
F1C C31 F2C 111.5(9)
F2 C31 F2C 83.3(12)
F1 C31 F2C 33.2(13)
F3C C31 F2C 110.6(9)
F3 C31 F2C 139.9(13)
F2B C31 F1B 99.0(14)
C31B C31 F1B 50.0(10)
F1C C31 F1B 43.9(14)
F2 C31 F1B 18.0(10)
F1 C31 F1B 111.8(12)
F3C C31 F1B 154.9(14)
F3 C31 F1B 124.6(14)
F2C C31 F1B 79.0(16)
F2B C31 S 120.4(19)
C31B C31 S 72.5(6)
F1C C31 S 109.7(7)
F2 C31 S 108.0(7)
F1 C31 S 108.9(7)
F3C C31 S 106.4(8)
F3 C31 S 104.9(7)
F2C C31 S 106.0(8)
F1B C31 S 92.2(9)
F2B C31 O4B 145.1(19)
C31B C31 O4B 116.0(8)
F1C C31 O4B 95.5(13)
F2 C31 O4B 116.7(12)
F1 C31 O4B 129.3(12)
F3C C31 O4B 73.4(12)
F3 C31 O4B 60.5(8)
F2C C31 O4B 147.1(11)
F1B C31 O4B 111.6(12)
S C31 O4B 44.9(4)
F2C C31B C31 76.0(17)
F2C C31B F3B 90(2)
C31 C31B F3B 159.8(18)
F2C C31B F2 97.0(18)
C31 C31B F2 63.0(13)
F3B C31B F2 134.2(16)
F2C C31B F2B 24(2)
C31 C31B F2B 52.0(13)
F3B C31B F2B 112.4(10)
F2 C31B F2B 86.4(17)
F2C C31B F1B 116(2)
C31 C31B F1B 87.7(16)
F3B C31B F1B 111.6(10)
F2 C31B F1B 27.2(15)
F2B C31B F1B 111.5(10)
F2C C31B F1 25.6(15)
C31 C31B F1 56.8(10)
F3B C31B F1 105.0(18)
F2 C31B F1 99.7(14)
F2B C31B F1 13.9(16)
F1B C31B F1 125.3(17)
F2C C31B F1C 98.2(17)
C31 C31B F1C 46.1(10)
F3B C31B F1C 152.6(15)
F2 C31B F1C 19.0(8)
F2B C31B F1C 81.2(16)
F1B C31B F1C 41.7(14)
F1 C31B F1C 92.6(13)
F2C C31B O3 133(2)
C31 C31B O3 116.9(8)
F3B C31B O3 63.3(9)
F2 C31B O3 129.6(15)
F2B C31B O3 136.1(13)
F1B C31B O3 109.9(13)
F1 C31B O3 122.6(14)
F1C C31B O3 123.7(11)
F2C C31B S 125(2)
C31 C31B S 71.7(6)
F3B C31B S 106.5(8)
F2 C31B S 106.4(13)
F2B C31B S 107.7(9)
F1B C31B S 106.8(8)
F1 C31B S 99.9(10)

F1C C31B S 90.5(10)
O3 C31B S 45.3(4)
C1 N C5 122.1(7)
C1 N Os 145.4(6)
C5 N Os 90.8(5)
N C1 C2 123.9(8)
N C1 C3 120.3(8)
C2 C1 C3 115.8(8)
C5 C4 Os 105.7(6)
N C5 C4 97.6(7)
N C5 C6 118.8(7)
C4 C5 C6 113.9(7)
C11 C6 C7 119.2(9)
C11 C6 C5 116.8(8)
C7 C6 C5 123.9(8)
C6 C7 C8 119.6(9)
C9 C8 C7 120.7(10)
C8 C9 C10 118.8(10)
C11 C10 C9 121.2(10)
C10 C11 C6 120.5(10)
C13 C12 C14 108.9(10)
C13 C12 P1 115.9(8)
C14 C12 P1 114.1(9)
C17 C15 C16 108.8(8)
C17 C15 P1 111.6(8)
C16 C15 P1 118.2(7)
C20 C18 C19 109.5(10)
C20 C18 P1 117.0(9)
C19 C18 P1 110.9(7)
C22 C21 C23 110.2(8)
C22 C21 P2 116.5(7)
C23 C21 P2 113.3(7)
C25 C24 C26 110.1(9)
C25 C24 P2 113.7(7)
C26 C24 P2 114.2(7)
C28 C27 C29 109.4(8)
C28 C27 P2 116.4(7)
C29 C27 P2 112.9(6)
O1 C30 Os 176.1(9)

Table to be deposited

Table S6. Atomic coordinates and equivalent isotropic displacement parameters (Å2) for [Os{(Z)-CH=C(Ph)NH=CR_2}Cl$(CO)_2(P^iPr_3)_2$][BF_4] (**10**).

Atom	X/*a*	Y/*b*	Z/*c*	U_{eq}[a]/U_{iso}[b]
Os	0.28594(3)	0.889734(11)	0.05574(3)	0.04128(14)
Cl	0.3393(2)	0.96387(7)	0.0965(2)	0.0502(7)
P1	0.1561(2)	0.89265(9)	0.1855(2)	0.0502(7)
P2	0.4194(2)	0.89728(8)	-0.0697(2)	0.0431(7)
F1	0.7154(9)	0.7214(3)	0.0917(8)	0.160(4)
F2	0.6586(7)	0.7824(2)	0.0273(6)	0.102(3)
F3	0.5789(10)	0.7230(4)	-0.0311(8)	0.172(5)
F4	0.7692(11)	0.7365(4)	-0.0461(9)	0.179(5)
O1	0.0563(6)	0.9093(2)	-0.0884(6)	0.058(2)
O2	0.2232(6)	0.7980(2)	0.0094(6)	0.061(2)
N	0.4742(7)	0.7989(2)	0.1484(6)	0.043(2)
B	0.6868(14)	0.7408(5)	0.0049(14)	0.071(5)
C1	0.4435(8)	0.8750(3)	0.1533(8)	0.045(3)
C2	0.5086(8)	0.8413(3)	0.1897(7)	0.042(2)
C3	0.4310(8)	0.7681(3)	0.1907(8)	0.048(3)
C4	0.3959(9)	0.7281(3)	0.1325(9)	0.063(4)
C5	0.4495(12)	0.6888(4)	0.1829(10)	0.079(4)
C6	0.4255(13)	0.6879(4)	0.2844(12)	0.095(5)
C7	0.4747(11)	0.7274(4)	0.3377(10)	0.084(4)
C8	0.4166(10)	0.7677(4)	0.2923(8)	0.063(3)
C9	0.6217(9)	0.8400(3)	0.2611(8)	0.049(3)
C10	0.7077(9)	0.8072(3)	0.2654(8)	0.056(3)
C11	0.8099(9)	0.8088(4)	0.3337(9)	0.067(4)
C12	0.8283(11)	0.8417(4)	0.3981(9)	0.072(4)
C13	0.7477(11)	0.8746(4)	0.3933(10)	0.081(4)
C14	0.6444(10)	0.8743(3)	0.3257(9)	0.066(4)
C15	0.1437(9)	0.9046(3)	-0.0345(8)	0.044(3)
C16	0.2471(8)	0.8317(3)	0.0278(8)	0.049(3)
C17	0.2477(9)	0.8887(4)	0.3045(7)	0.056(3)
C18	0.3246(10)	0.9288(4)	0.3321(8)	0.070(4)
C19	0.1748(11)	0.8769(4)	0.3876(9)	0.082(4)
C20	0.0393(9)	0.8501(3)	0.1839(9)	0.058(3)
C21	-0.0470(9)	0.8484(3)	0.0935(9)	0.072(4)
C22	0.0974(10)	0.8060(3)	0.2035(9)	0.073(4)
C23	0.0643(10)	0.9437(3)	0.1927(9)	0.060(3)
C24	0.0208(10)	0.9655(3)	0.0987(8)	0.069(4)
C25	-0.0426(11)	0.9397(4)	0.2535(9)	0.084(4)
C26	0.5864(9)	0.8892(3)	-0.0258(7)	0.051(3)
C27	0.6748(10)	0.8799(4)	-0.0964(9)	0.076(4)
C28	0.6372(9)	0.9262(3)	0.0364(8)	0.057(3)
C29	0.3919(9)	0.8596(3)	-0.1693(8)	0.054(3)
C30	0.2617(10)	0.8618(4)	-0.2230(9)	0.072(4)
C31	0.4286(11)	0.8137(3)	-0.1396(9)	0.078(4)
C32	0.4127(9)	0.9513(3)	-0.1268(8)	0.046(3)
C33	0.2857(9)	0.9708(3)	-0.1459(8)	0.057(3)
C34	0.4751(10)	0.9536(3)	-0.2135(9)	0.066(3)
C50[b]	0.4812(14)	0.8279(3)	-0.4528(10)	0.157(8)
C13A[b]	0.441(2)	0.8808(4)	-0.4297(15)	0.114(9)
C14A[b]	0.607(2)	0.8208(9)	-0.3551(15)	0.140(12)
C13B[b]	0.528(2)	0.8807(5)	-0.422(2)	0.29(2)
C14B[b]	0.561(2)	0.7876(7)	-0.3864(19)	0.118(8)
C14C[b]	0.5882(15)	0.7894(5)	-0.4737(15)	0.096(6)

Cl3D[b]	0.5804(16)	0.8084(7)	-0.3468(11)	0.081(6)
Cl4D[b]	0.494(2)	0.8834(3)	-0.4299(11)	0.062(4)
O60A[b]	0.820(2)	0.9690(8)	0.5577(16)	0.093(8)
C60A[b]	0.893(2)	0.9893(16)	0.496(3)	0.193(19)
C61A[b]	0.812(4)	1.0183(11)	0.430(2)	0.131(14)
C62A[b]	0.687(3)	1.0003(13)	0.431(2)	0.128(14)
C63A[b]	0.695(3)	0.9743(17)	0.520(3)	0.161(14)
C64[b]	0.617(4)	0.9935(16)	0.454(3)	0.078(14)
C65[b]	0.638(3)	0.970(2)	0.547(4)	0.140(19)
O66[b]	0.765(3)	0.9652(11)	0.574(2)	0.077(11)
C67[b]	0.793(3)	0.9416(12)	0.660(2)	0.060(12)
C68[b]	0.928(2)	0.9358(10)	0.671(2)	0.037(9)

[a] Equivalent isotropic *U* defined as one third of the trace of the orthogonalized *Uij* tensor.
b **10** crystallizes with several molecules of disordered solvent, modeled as 0.9 molecules of disordered dichromethane (C(50)-Cl(4D)), 0.4 molecules of THF (O(60A)-C(63A)) and 0.25 molecules of diethyl ether (C(64)-C(68)).

Table to be deposited

Table S7. Anisotropic displacement coefficients U_{ij} (Å^2) for the non-hydrogen atoms for the complex [Os{(Z)-CH=C(Ph)NH=CR$_2$}Cl(CO)$_2$(P^{i}Pr$_3$)$_2$][BF$_4$].

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Os	0.0373(2)	0.02902(19)	0.0546(3)	0.0000(2)	-0.00578(16)	0.0020(2)
Cl	0.0531(16)	0.0308(13)	0.064(2)	-0.0040(12)	-0.0043(13)	0.0010(11)
P1	0.0427(15)	0.0432(15)	0.062(2)	0.0024(16)	-0.0029(13)	0.0095(13)
P2	0.0436(15)	0.0318(15)	0.0513(19)	0.0001(12)	-0.0034(13)	0.0013(11)
F1	0.179(10)	0.144(9)	0.157(11)	0.063(8)	0.021(8)	0.064(7)
F2	0.130(7)	0.082(5)	0.095(7)	-0.008(5)	0.018(5)	0.033(5)
F3	0.164(9)	0.176(10)	0.169(12)	-0.053(8)	-0.006(8)	-0.051(8)
F4	0.190(11)	0.177(10)	0.204(12)	-0.011(9)	0.160(10)	0.009(8)
O1	0.043(4)	0.050(4)	0.076(6)	0.003(4)	-0.014(4)	0.006(3)
O2	0.070(5)	0.025(4)	0.083(6)	-0.008(4)	-0.013(4)	-0.012(3)
N	0.043(5)	0.033(5)	0.049(6)	0.006(4)	-0.005(4)	0.007(4)
B	0.056(10)	0.058(10)	0.098(15)	0.017(9)	0.001(9)	-0.007(8)
C1	0.030(5)	0.036(5)	0.067(8)	-0.008(5)	-0.005(5)	-0.001(4)
C2	0.040(6)	0.044(6)	0.045(7)	-0.003(5)	0.010(5)	0.003(5)
C3	0.035(6)	0.038(6)	0.070(9)	0.009(6)	0.002(5)	0.013(5)
C4	0.049(7)	0.041(6)	0.097(11)	0.014(6)	0.001(6)	-0.002(5)
C5	0.082(9)	0.054(8)	0.102(13)	0.020(8)	0.014(8)	0.015(7)
C6	0.091(11)	0.065(9)	0.131(16)	0.043(10)	0.018(10)	0.016(8)
C7	0.059(8)	0.106(11)	0.087(11)	0.048(9)	0.010(7)	0.023(8)
C8	0.063(8)	0.066(8)	0.062(9)	0.037(7)	0.013(6)	0.009(6)
C9	0.043(6)	0.050(6)	0.052(8)	0.013(5)	-0.007(5)	0.002(5)
C10	0.042(6)	0.061(7)	0.061(8)	0.007(6)	-0.007(5)	0.008(5)
C11	0.037(7)	0.084(9)	0.079(10)	0.032(8)	0.000(6)	0.001(6)
C12	0.072(9)	0.077(9)	0.058(9)	0.010(7)	-0.025(7)	-0.001(7)
C13	0.078(9)	0.072(9)	0.085(11)	-0.029(7)	-0.025(8)	0.014(7)
C14	0.063(8)	0.050(7)	0.081(10)	-0.013(6)	-0.007(7)	0.011(6)
C15	0.048(6)	0.029(5)	0.053(7)	-0.002(5)	0.000(5)	0.000(4)
C16	0.032(6)	0.052(7)	0.058(8)	0.005(6)	-0.016(5)	-0.002(5)
C17	0.047(6)	0.072(7)	0.046(7)	0.008(7)	-0.008(5)	0.008(6)
C18	0.061(8)	0.088(9)	0.059(9)	-0.003(7)	-0.004(6)	0.006(7)
C19	0.078(9)	0.106(11)	0.061(10)	0.019(8)	0.005(7)	0.020(7)
C20	0.041(6)	0.044(6)	0.085(10)	0.013(6)	-0.007(6)	-0.005(5)
C21	0.039(7)	0.061(8)	0.115(12)	0.005(7)	0.009(7)	-0.005(5)
C22	0.064(8)	0.039(6)	0.119(12)	0.021(7)	0.027(8)	-0.003(6)
C23	0.058(7)	0.036(6)	0.086(10)	-0.005(6)	0.010(7)	0.022(5)
C24	0.065(8)	0.057(7)	0.083(10)	0.009(7)	0.003(7)	0.025(6)
C25	0.096(10)	0.097(10)	0.063(10)	-0.001(8)	0.029(8)	0.042(8)
C26	0.052(6)	0.055(6)	0.047(7)	0.014(6)	0.008(5)	0.011(6)
C27	0.047(7)	0.094(10)	0.083(10)	0.011(8)	0.002(6)	0.012(6)
C28	0.057(7)	0.051(7)	0.060(8)	-0.012(6)	-0.003(6)	-0.006(5)
C29	0.055(7)	0.039(6)	0.068(9)	-0.011(6)	0.010(6)	-0.014(5)
C30	0.067(8)	0.064(8)	0.084(11)	-0.028(7)	0.005(7)	-0.013(6)
C31	0.104(10)	0.047(7)	0.082(11)	-0.011(7)	0.010(8)	0.012(7)
C32	0.045(6)	0.034(5)	0.059(8)	0.011(5)	0.003(5)	0.002(4)
C33	0.069(8)	0.041(6)	0.058(8)	0.005(5)	-0.005(6)	-0.002(5)
C34	0.060(8)	0.052(7)	0.085(10)	0.007(7)	0.003(7)	0.004(6)

* The anisotropic displacement factor exponent takes the form:

$-2\pi^2 (h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12})$

Table to be deposited

Table S8. Hydrogen atom coordinates* and isotropic displacement coefficient ($Å^2$) for the compound $[Os\{(Z)\text{-}CH{=}C(Ph)NH{=}CR_2\}Cl(CO)_2(P^iPr_3)_2][BF_4]$.

Atom	X/*a*	Y/*b*	Z/*c*	*U*
H0	0.4849	0.7949	0.0885	0.051
H1	0.4799	0.9005	0.1799	0.054
H4A	0.3046	0.7256	0.1213	0.076
H4B	0.4268	0.7305	0.0700	0.076
H5A	0.4125	0.6632	0.1498	0.095
H5B	0.5399	0.6880	0.1802	0.095
H6A	0.4654	0.6624	0.3159	0.114
H6B	0.3352	0.6856	0.2869	0.114
H7A	0.4570	0.7257	0.4044	0.100
H7B	0.5656	0.7288	0.3383	0.100
H8A	0.3275	0.7686	0.3003	0.076
H8B	0.4572	0.7930	0.3239	0.076
H10	0.6964	0.7840	0.2221	0.067
H11	0.8693	0.7865	0.3363	0.081
H12	0.8977	0.8412	0.4460	0.087
H13	0.7619	0.8979	0.4362	0.098
H14	0.5879	0.8975	0.3225	0.079
H17	0.3085	0.8651	0.3001	0.067
H18A	0.2708	0.9510	0.3529	0.106
H18B	0.3623	0.9389	0.2771	0.106
H18C	0.3897	0.9219	0.3841	0.106
H19A	0.2325	0.8669	0.4417	0.123
H19B	0.1152	0.8543	0.3673	0.123
H19C	0.1305	0.9021	0.4065	0.123
H20	-0.0116	0.8564	0.2363	0.069
H21A	-0.0023	0.8382	0.0421	0.108
H21B	-0.0799	0.8770	0.0780	0.108
H21C	-0.1154	0.8290	0.1012	0.108
H22A	0.0362	0.7840	0.1825	0.109
H22B	0.1238	0.8028	0.2718	0.109
H22C	0.1692	0.8029	0.1687	0.109
H23	0.1236	0.9642	0.2280	0.072
H24A	-0.0390	0.9472	0.0604	0.103
H24B	0.0920	0.9707	0.0642	0.103
H24C	-0.0186	0.9927	0.1108	0.103
H25A	-0.0682	0.9681	0.2716	0.126
H25B	-0.0149	0.9231	0.3108	0.126
H25C	-0.1126	0.9252	0.2167	0.126
H26	0.5899	0.8639	0.0171	0.061
H27A	0.6931	0.9063	-0.1288	0.113
H27B	0.6373	0.8592	-0.1432	0.113
H27C	0.7516	0.8681	-0.0631	0.113
H28A	0.7178	0.9184	0.0706	0.085
H28B	0.5797	0.9330	0.0822	0.085
H28C	0.6469	0.9512	-0.0035	0.085
H29	0.4490	0.8683	-0.2158	0.065
H30A	0.2520	0.8396	-0.2721	0.108
H30B	0.2486	0.8898	-0.2529	0.108
H30C	0.2009	0.8573	-0.1786	0.108
H31A	0.3682	0.8020	-0.1008	0.116
H31B	0.5109	0.8138	-0.1025	0.116
H31C	0.4302	0.7961	-0.1965	0.116
H32	0.4610	0.9706	-0.0797	0.055

H33A	0.2911	0.9983	-0.1777	0.086
H33B	0.2521	0.9750	-0.0856	0.086
H33C	0.2311	0.9517	-0.1868	0.086
H34A	0.4259	0.9382	-0.2653	0.099
H34B	0.5575	0.9407	-0.2009	0.099
H34C	0.4833	0.9835	-0.2319	0.099

* The hydrogen atoms were included in observed or calculated positions and refined riding on their respective carbon atoms.

Table to be deposited

Table S9. Full experimental details for the X-Ray analysis of the [Os{(Z)-CH=C(Ph)NH=CR_2}Cl$(CO)_2(P^iPr_3)_2$][BF_4].

Crystal data:

Formula	C34H59BClF4NO2OsP2 $0.9CH_2Cl_2$ 0.4THF $0.25C_4H_{10}O$
Molecular weight	884.19
Crystal habit	irregular block
Size(mm)	0.26 x 0.20 x 0.20
Symmetry	monoclinic, P21/n
Unit cell dimensions	10.887(3), 31.389(8), 14.117(4) Å 90, 97.210(7), 90 °
Packing: V (Å^3), Z	4806(2), 4
D_{calc}(g cm^{-3}), F (000)	1.391, 2034

Experimental data:	Bruker Siemens-SMART APEX
Radiation and technique	Mo-$K\alpha$ (λ = 0.71073 Å)
Monochromator	Graphite oriented
Range	($5 < 2\theta < 55°$)
Number of reflections:	
measured	31470 (h: -14, 10; k: -40, 40; l: -11, 18)
unique	10932 (R_{int} = 0.1345)
Absorption correction	sababs*
Max. and min. trans. fact.	1.000, 0.712
μ (mm^{-1})	2.922
Temperature (K)	173.0(2)

Table to be deposited

Table S9. Full experimental details for the X-Ray analysis of the [Os{(Z)-CH=C(Ph)NH=CR_2}Cl$(CO)_2(P^iPr_3)_2$][BF_4].

Solution and refinement:	SHELX97 dos/win95/nt version
Solution mode	Patterson
Refinement	Full-matrix least-squares on F'^2s.
	All reflections.
Hydrogen atoms	From calculated positions. Refined riding on C
	atoms with thermal parameters related to bonded atoms.
No. parameters/restrains	501/37
Weighting scheme	$w^{-1}=[\sigma^2(F_o^2)+(0.0541P)^2+0P]$
	where $P=((\text{Max } F_o^2,0)+2F_c^2)/3$
ΔF final (max)	1.362 e/Å^3 (close to Os atom)
ΔF final (min)	-0.909 e/Å^3
Max/Mean shift/esd	0.182, 0.011
$\omega R2(F^2, \text{all data})^a$	0.1562
$R1(F, F_o > 4.0\ \sigma F)^b$	0.0645
Goodness-of-Fit[c]	0.794
Data-to-Parameter Ratio	20.9:1

Atomic scattering factors from the International Tables for X-Ray Crystallography; Vol C (1992). Anomalous dispersion is implemented by the program.

[a] $wR2(F^2) = \{\Sigma[w(F_o^2-F_c^2)^2]/\Sigma[w(F_o^2)^2]\}^{1/2}$ [b] $R1(F) = \Sigma||F_o|-|F_c||/\Sigma|F_o|$. [c] $S = \{\Sigma[w(F_o^2-F_c^2)^2]/(n-p)\}^{1/2}$, where n is the number of observed reflections, and p is the number of parameters refined.

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* Smart Apex 5.0., Bruker-Siemens Analytical X-ray Instruments, Inc: Madison, WI 2000.

Table to be deposited

Table S10. Bond Lenghts (Å) and angles (deg) for the complex [Os{(Z)-CH=C(Ph)NH=CR_2}Cl$(CO)_2(P^iPr_3)_2$][BF_4].

Os C16 1.901(11)	C61A C62A 1.479(18)
Os C15 1.938(10)	C62A C64 0.89(6)
Os C1 2.115(9)	C62A C63A 1.500(17)
Os P2 2.445(3)	C62A C65 2.03(5)
Os Cl 2.450(2)	C63A C65 0.78(5)
Os P1 2.458(3)	C63A O66 1.04(4)
P1 C20 1.841(10)	C63A C64 1.33(4)
P1 C17 1.853(10)	C64 C65 1.50(2)
P1 C23 1.898(9)	C65 O66 1.397(19)
P2 C29 1.838(10)	O66 C67 1.435(19)
P2 C26 1.863(10)	C67 C68 1.464(19)
P2 C32 1.877(9)	
F1 B 1.373(18)	C16 Os C15 87.4(4)
F2 B 1.387(17)	C16 Os C1 93.9(4)
F3 B 1.343(16)	C15 Os C1 178.5(4)
F4 B 1.228(18)	C16 Os P2 94.5(3)
O1 C15 1.152(10)	C15 Os P2 89.7(3)
O2 C16 1.113(10)	C1 Os P2 89.4(3)
N C3 1.260(12)	C16 Os Cl 178.2(3)
N C2 1.482(11)	C15 Os Cl 94.2(3)
C1 C2 1.340(12)	C1 Os Cl 84.5(3)
C2 C9 1.494(12)	P2 Os Cl 86.25(9)
C3 C8 1.469(15)	C16 Os P1 93.3(3)
C3 C4 1.525(14)	C15 Os P1 89.9(3)
C4 C5 1.507(14)	C1 Os P1 90.8(3)
C5 C6 1.495(17)	P2 Os P1 172.16(8)
C6 C7 1.517(18)	Cl Os P1 85.96(10)
C7 C8 1.518(14)	C20 P1 C17 104.7(5)
C9 C10 1.388(13)	C20 P1 C23 104.2(5)
C9 C14 1.415(14)	C17 P1 C23 103.6(5)
C10 C11 1.382(13)	C20 P1 Os 115.1(4)
C11 C12 1.376(15)	C17 P1 Os 112.6(4)
C12 C13 1.354(15)	C23 P1 Os 115.3(4)
C13 C14 1.383(14)	C29 P2 C26 103.1(5)
C17 C18 1.534(14)	C29 P2 C32 104.9(5)
C17 C19 1.547(15)	C26 P2 C32 104.5(5)
C20 C21 1.492(14)	C29 P2 Os 116.2(4)
C20 C22 1.534(12)	C26 P2 Os 112.8(3)
C23 C24 1.521(14)	C32 P2 Os 114.0(4)
C23 C25 1.537(15)	C3 N C2 126.2(10)
C26 C27 1.501(14)	F4 B F3 113.5(18)
C26 C28 1.520(13)	F4 B F1 112.3(14)
C29 C30 1.524(13)	F3 B F1 104.2(13)
C29 C31 1.539(13)	F4 B F2 115.9(14)
C32 C34 1.479(14)	F3 B F2 105.7(12)
C32 C33 1.505(12)	F1 B F2 104.1(15)
C50 Cl4C 1.729(9)	C2 C1 Os 140.4(7)
C50 Cl4B 1.744(10)	C1 C2 N 117.4(8)
C50 Cl3A 1.761(10)	C1 C2 C9 129.3(9)
C50 Cl4D 1.777(10)	N C2 C9 113.0(8)
C50 Cl3B 1.774(10)	N C3 C8 124.6(10)
C50 Cl4A 1.835(10)	N C3 C4 117.2(11)
C50 Cl3D 1.841(10)	C8 C3 C4 118.2(10)
Cl4B Cl4C 1.31(3)	C5 C4 C3 111.3(10)
O60A O66 0.68(4)	C6 C5 C4 111.6(11)
O60A C63A 1.404(19)	C5 C6 C7 111.7(12)
O60A C60A 1.411(19)	C8 C7 C6 111.6(11)
O60A C67 1.75(4)	C3 C8 C7 109.5(10)
C60A C61A 1.512(18)	C10 C9 C14 118.3(9)

C10 C9 C2 122.9(10)
C14 C9 C2 118.7(9)
C11 C10 C9 118.9(11)
C12 C11 C10 121.9(11)
C13 C12 C11 120.0(11)
C12 C13 C14 119.7(12)
C13 C14 C9 120.9(10)
O1 C15 Os 173.2(8)
O2 C16 Os 178.4(9)
C18 C17 C19 108.4(10)
C18 C17 P1 113.1(8)
C19 C17 P1 116.2(7)
C21 C20 C22 108.9(10)
C21 C20 P1 113.3(8)
C22 C20 P1 112.6(7)
C24 C23 C25 110.7(9)
C24 C23 P1 116.1(8)
C25 C23 P1 113.7(8)
C27 C26 C28 108.6(9)
C27 C26 P2 119.0(8)
C28 C26 P2 111.0(7)
C30 C29 C31 111.9(9)
C30 C29 P2 114.0(7)
C31 C29 P2 112.4(8)
C34 C32 C33 109.8(9)
C34 C32 P2 113.9(7)
C33 C32 P2 115.4(7)
Cl4C C50 Cl4B 44.3(10)
Cl4C C50 Cl3A 151.8(15)
Cl4B C50 Cl3A 134.8(14)
Cl4C C50 Cl4D 132.5(15)
Cl4B C50 Cl4D 126.1(13)
Cl3A C50 Cl4D 19.2(8)
Cl4C C50 Cl3B 121.0(15)
Cl4B C50 Cl3B 115.7(14)
Cl3A C50 Cl3B 31.1(12)
Cl4D C50 Cl3B 12.4(13)
Cl4C C50 Cl4A 65.8(12)
Cl4B C50 Cl4A 39.5(11)
Cl3A C50 Cl4A 98.9(12)
Cl4D C50 Cl4A 86.9(12)
Cl3B C50 Cl4A 76.2(13)
Cl4C C50 Cl3D 64.5(10)
Cl4B C50 Cl3D 27.9(9)
Cl3A C50 Cl3D 107.2(11)
Cl4D C50 Cl3D 98.7(11)
Cl3B C50 Cl3D 89.2(13)
Cl4A C50 Cl3D 15.8(11)
Cl4C Cl4B C50 67.3(7)
Cl4B Cl4C C50 68.5(7)
O66 O60A C63A 45(3)
O66 O60A C60A 152(4)
C63A O60A C60A 108.1(13)
O66 O60A C67 52(3)
C63A O60A C67 97(2)
C60A O60A C67 155(2)
O60A C60A C61A 108.8(15)
C62A C61A C60A 103.5(13)
C64 C62A C61A 158(6)
C64 C62A C63A 61(3)
C61A C62A C63A 105.0(13)
C64 C62A C65 43(2)
C61A C62A C65 121.6(19)
C63A C62A C65 19.0(17)
C65 C63A O66 99(5)
C65 C63A C64 87(4)
O66 C63A C64 169(6)
C65 C63A O60A 126(5)
O66 C63A O60A 27(2)
C64 C63A O60A 145(3)
C65 C63A C62A 123(5)
O66 C63A C62A 136(3)
C64 C63A C62A 36(3)
O60A C63A C62A 109.7(14)
C62A C64 C63A 83(3)
C62A C64 C65 114(3)
C63A C64 C65 31(2)
C63A C65 O66 48(3)
C63A C65 C64 62(3)
O66 C65 C64 109(2)
C63A C65 C62A 38(3)
O66 C65 C62A 85.2(18)
C64 C65 C62A 23.7(16)
O60A O66 C63A 107(4)
O60A O66 C65 141(3)
C63A O66 C65 34(3)
O60A O66 C67 106(3)
C63A O66 C67 146(3)
C65 O66 C67 112.9(19)
O66 C67 C68 105.0(17)
O66 C67 O60A 21.9(13)
C68 C67 O60A 83.1(17)