corresponding ν(CO) values at somewhat higher energies.

Concluding Remarks. The sodium in liquid ammonia reduction of cyclopentadienyltetracarbonylniobium and -tantalum provides the corresponding $Na_2[C_5H_5M(CO)_3]$ that may be isolated as the very thermally stable dicesium salts $Cs_2[C_5H_5M(CO)_3]$. This is the first isolated compound of satisfactory purity containing the niobium dianion and the first report on the substituted tantalum dianion. These materials have been further characterized by their conversion to monoanionic hydrides and triphenylstannyl derivatives, which have been isolated as pure tetraethylammonium salts for the first time. Further studies on the chemical properties of these highly reactive substances are in progress.

Acknowledgment. We thank the donors of the Petroleum Research Fund, administered by the American Chemical Society, and the National Science Foundation (CHE82-10496) for continuing support of this research.

Registry No. 1, 87801-25-0; 2, 87801-26-1; 3, 87801-28-3; 4, 84950-24-3; 5, 87801-30-7; 6, 87801-32-9; $Na_2[C_5H_5Ta(CO)_3]$, 87801-33-0; $Na_2[C_5H_5Nb(CO)_3]$, 84950-22-1; $C_5H_5Ta(CO)_4$, 32628-95-8; $C_5H_5Nb(CO)_4$, 12108-03-1; CsI, 7789-17-5; $[Et_4N]Br$, 71-91-0; Ph_3SnCl, 639-58-7.

X-ray Crystal and Molecular Structures of the Monomeric 1:1 Adducts of Diphenyltin(IV) Dichloride and Trimethyltin(IV) Chloride with 2,6-Dimethylpyridine (2,6-Lutidine) *N*-Oxide

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Diphenyltin(IV) dichloride–2,6-dimethylpyridine *N*-oxide (I), $C_{19}H_{19}Cl_2NOSn$, forms colorless crystals, mp 159 °C, in the triclinic space group $P\bar{1}$ with $a = 9.359$ (2) Å, $b = 10.534$ (2) Å, $c = 10.996$ (3) Å, $\alpha = 97.46$ (2)°, $\beta = 114.74$ (2)°, $\gamma = 93.26$ (2)°, $V = 968.8$ (3) Å^3, $Z = 2$, and $\rho = 1.60$ g cm^{-3}. The structure was determined from 3609 reflections collected on a Nicolet R3 diffractometer with monochromatic Mo Kα radiation and refined to a final R value of 0.0253 for the 3102 reflections included in the least-squares sums. The monomeric complex contains five-coordinated, trigonal-bipyramidal tin with the phenyl rings forming an angle of 124.1 (1)° in the equatorial plane. The ligand oxygen and one of the chlorine atoms occupy the apical positions at an angle of 172.8 (1)°. The sum of the angles subtended at the tin atom in the trigonal plane is 359.0 (3)°. The tin atom is displaced from this plane by 0.1279 (2) Å in the direction of the chlorine atom. The Sn–Cl$_{eq}$ bond [2.366 (1) Å] is shorter than the Sn–Cl$_{ax}$ bond [2.471 (1) Å]. The Sn–O distance is 2.296 (2) Å, and the two Sn–C bond distances are approximately equal [2.119 (3) and 2.128 (4) Å]. Trimethyltin(IV) chloride–2,6-dimethylpyridine *N*-oxide (II), $C_{10}H_{18}ClNOSn$, mp 106 °C, also crystallizes as colorless crystals in the same space group with $a = 8.174$ (2) Å, $b = 9.599$ (2) Å, $c = 9.895$ (3) Å, $\alpha = 115.33$ (2)°, $\beta = 105.50$ (2)°, $\gamma = 81.45$ (2)°, $V = 675.7$ (2) Å^3, $Z = 2$, and $\rho = 1.58$ g cm^{-3}. This structure was solved from 2437 reflections and refined to a final R value of 0.0283 for the 2207 reflections that were used in the least-squares sums. The molecule has the tin atom in a trigonal-bipyramidal geometry with the methyl carbons in the trigonal plane. The Sn–C bond distances are 2.119 (5), 2.120 (4), and 2.125 (6) Å, and the C–Sn–C angles are 114.1 (2), 121.8 (3), and 123.1 (2)°, giving a sum of 358.9 (3)°. The apical Cl–Sn–O angle is 177.4 (1)°, and the tin atom is displaced from the trigonal plane by 0.2357 (2) Å toward the chlorine atom. Both the Sn–Cl and Sn–O distances are larger than those found in structure I [2.533 (1) and 2.400 (2) Å, respectively)]. The angle at the oxygen atom is 125.7 (2)° in I and 130.6 (2)° in II, and the angle formed by the Sn–O–N system and the pyridine *N*-oxide ring is 86.98 (4)° in I and 84.75 (16)° in II. The N–O distances are 1.346 (4) Å in I and 1.326 (4) Å in II.

We have discovered that the 1:1 adducts formed by diorganotin(IV) dihalides with certain pointed ligands containing the >C=O, >C=S, >S=O, ⋟N→O, or ⋟P=O grouping are in fact six coordinated.[1] The structures of the 1:1 dimethyltin(IV) dichloride complexes with diphenylcyclopropenone[2] and with 2,6-dimethylpyridine (2,6-lutidine) *N*-oxide[3] reveal dimer formation through bridging chlorine atoms, and the structure of the 1:1 salicylaldehyde adduct[4,5] can be reinterpreted in similar terms as an associated solid with bridging phenolic oxygen atoms. Only a single example of a true five-coordinated 1:1 adduct has been reported: diphenyltin(IV) dichloride–benzthiazole.[6]

Di-[7] and trimethyltin(IV)[8] chlorides are themselves associated in the solid state through bridging chlorine atoms,

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Table I. Crystal Data and Data Collection Parameters for $Ph_2SnCl_2 \cdot 2,6\text{-}(CH_3)_2C_5H_3NO$ (I) and $Me_3SnCl \cdot 2,6\text{-}(CH_3)_2C_5H_3NO$ (II) at 23 °C

	I	II
formula	$C_{19}H_{19}Cl_2NOSn$	$C_{10}H_{18}ClNOSn$
space group	$P\bar{1}$	
a, Å	9.359 (2)	8.174 (2)
b, Å	10.534 (2)	9.599 (2)
c, Å	10.996 (3)	9.895 (3)
α, deg	97.46 (2)	115.33 (2)
β, deg	114.74 (2)	105.50 (2)
γ, deg	93.26 (2)	81.45 (2)
V, Å^3	968.8 (3)	675.7 (2)
Z	2	
d(calcd), g cm^{-3}	1.60	1.58
μ, cm^{-1} (Mo Kα)	16.1	20.7
transmission (max/min)	1.23	1.29
2θ scan range, deg	4–50	
diffractometer	Nicolet R3	
data collected	$\pm h,\pm k,+l$	
unique data measd	3414 (3609 collected)	2298 (2437 collected)
unique data used [$I \geqslant 3\sigma(I)$]	3102	2207
std rflns	3/97 (7% decay)	3/97 (8% decay)
R_F, %	2.53	2.83
R_{wF}, %	2.28	3.13
GOF	1.61	1.21
highest peak, final diff map, e Å^{-3}	0.44	0.82
mean shift/max esd, final cycle	0.08	0.04
data/parameters, final cycle	10.6/1	17.2/1

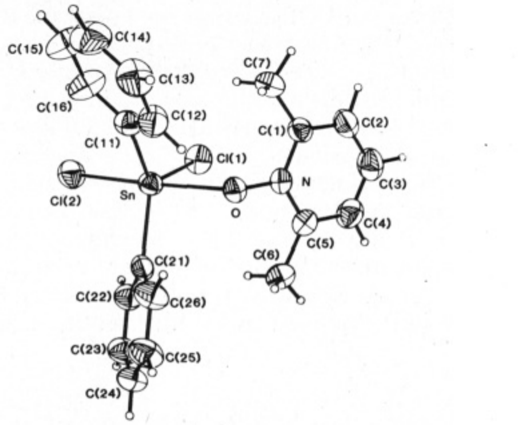

Figure 1. Molecular geometry and labeling diagram for $(C_6H_5)_2SnCl_2 \cdot 2,6\text{-}(CH_3)_2C_5H_3NO$ (I). Thermal ellipsiods are drawn at the 50% probability level.

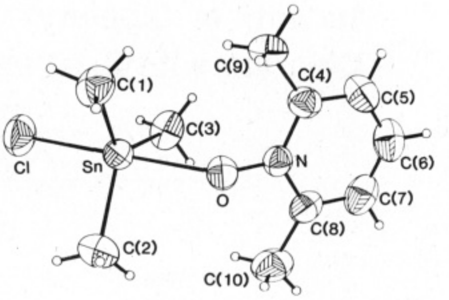

Figure 2. Molecular geometry and labeling diagram for $(CH_3)_3SnCl \cdot 2,6\text{-}(CH_3)_2C_5H_3NO$ (II). Thermal ellipsoids are drawn at the 50% probability level.

but triphenyltin(IV) chloride is a monomer,[9] and the diphenyl analogue crystallizes in a complex structure that contains both four- and six-coordinated tin atoms. The structure was originally interpreted in terms of discrete molecules,[10] and in any case the association must be weak.[11]

We have used tin-119m Mössbauer data to choose a complex that might be 1:1 and monomeric. The quadrupole splitting (QS) values increase with carbon–tin–carbon angles in diorganotin(IV) systems.[12] A treatment based upon a point-charge model has been successful in predicting these angles in diphenyltin(IV) systems[13,14] where structural data are available.[15] Since the *trans*-R_2SnX_2 octahedral configuration is ubiquitous for six-coordinated adducts and axially most electronegative, trigonal-bipyramidal configurations for five-coordinated adducts,[15] the QS values can specify for the 1:1 $R_2SnX_2 \cdot L$ complexes whether they are monomeric (equatorial R_2Sn grouping, angle C–Sn–C ≃ 120°) or bridged (trans R_2Sn grouping, angle C–Sn–C = 180°). For the 2,6-dimethylpyridine (2,6-lutidine) *N*-oxide complex of diphenyltin(IV) dichloride (I), we found the isomer shift (IS) = 1.01 ± 0.03 and QS = 2.94 ± 0.06 mm s^{-1} which corresponds to a C–Sn–C angle of 133°. Thus we carried out the structural determination reported in this paper.

The structure of the analogous 1:1 trimethyltin(IV) chloride adduct (II) was determined to complete the series $(CH_3)_nSnCl_{4-n} \cdot x2,6\text{-}R_2C_5H_3NO$ for which the $n = 2$ and $x = 0$,[7] 1,[3] and 2[16] and the $n = 3$ and $x = 0$[7] members have already been studied.

Experimental Section

Preparation of Diphenyltin(IV) Dichloride–2,6-Dimethylpyridine (2,6-Lutidine) *N*-Oxide [$(C_6H_5)_2SnCl_2 \cdot 2,6\text{-}(CH_3)_2C_5H_3NO$] and Trimethyltin(IV) Chloride–2,6-Dimethylpyridine (2,6-Lutidine) *N*-Oxide [$(CH_3)_3SnCl \cdot 2,6\text{-}(CH_3)_2C_5H_3NO$]. The adducts were prepared by mixing stoichiometric amounts of the organotin(IV) chloride [diphenyltin(IV) dichloride, 3.44 g, 10.0 mmol; trimethyltin(IV) chloride, 1.99 g, 10.0 mmol] and 2,6-dimethylpyridine (2,6-lutidine) *N*-oxide (1.23 g, 10.0 mmol) in chloroform. The organotins were gifts from M&T Chemicals, and the ligand was an Aldrich product. Single crystals were obtained from the slow evaporation of chloroform solutions of the complexes which were isolated in ca. 70% yield: I, mp 159 °C; II, mp 106 °C.

X-ray Data Collection. Crystals of I and II were trimmed with a razor blade to nearly spherical shapes (radius: I, 0.17 mm; II, 0.14 mm) and mounted in nitrogen-flushed, thin-walled capillary tubes. Crystal parameters and some details of data collection are provided in Table I. Final unit-cell dimensions were obtained from the angular settings of 25 well-centered reflections, including Friedel pairs, in the range 30° < 2θ < 36°. Intensity data were corrected for *Lp* effects and decay but not for absorption. Redundant data were averaged with less than 1.5% disagreement.

Solution and Refinement of the Structure. The positions of the tin, chlorine, and oxygen atoms were obtained from a sharpened Patterson map. Subsequent difference Fourier syntheses revealed the locations of all atoms, including hydrogen, for I and all non-hydrogen atoms for II. In the final refinement cycles, all non-hydrogen atoms were refined with anisotropic thermal parameters. In I, the found hydrogen atoms were refined

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Table II. Atomic Coordinates and Isotropic Thermal Parameters for I and II[a]

atom	*x*	*y*	*z*	*U*, Å²
		I		
Sn	0.18251 (2)	0.23379 (2)	0.27358 (2)	0.0380 (1)
Cl(1)	−0.04265 (9)	0.08661 (8)	0.22456 (8)	0.0536 (3)
Cl(2)	0.0034 (1)	0.39252 (8)	0.17062 (9)	0.0585 (4)
O	0.3205 (2)	0.0694 (2)	0.3633 (2)	0.0407 (8)
N	0.2564 (4)	−0.0457 (3)	0.3698 (3)	0.039 (1)
C(1)	0.2116 (3)	−0.1430 (3)	0.2621 (3)	0.043 (1)
C(2)	0.1519 (4)	−0.2614 (4)	0.2728 (4)	0.051 (2)
C(3)	0.1349 (5)	−0.2802 (4)	0.3860 (5)	0.061 (2)
C(4)	0.1805 (5)	−0.1786 (4)	0.4936 (4)	0.057 (2)
C(5)	0.2413 (4)	−0.0597 (3)	0.4852 (3)	0.045 (1)
C(6)	0.2926 (6)	0.0532 (4)	0.5959 (4)	0.062 (2)
C(7)	0.2299 (5)	−0.1166 (3)	0.1405 (3)	0.057 (2)
C(11)	0.2924 (3)	0.2252 (3)	0.1385 (3)	0.041 (1)
C(12)	0.4395 (4)	0.1815 (3)	0.1742 (3)	0.054 (1)
C(13)	0.5135 (5)	0.1830 (4)	0.0879 (5)	0.068 (2)
C(14)	0.4427 (5)	0.2274 (4)	−0.0320 (4)	0.074 (2)
C(15)	0.2979 (6)	0.2708 (6)	−0.0683 (4)	0.080 (2)
C(16)	0.2234 (4)	0.2698 (4)	0.0176 (3)	0.062 (2)
C(21)	0.2845 (3)	0.3434 (3)	0.4722 (3)	0.039 (1)
C(22)	0.1922 (4)	0.4046 (3)	0.5259 (4)	0.049 (2)
C(23)	0.2593 (5)	0.4747 (4)	0.6559 (4)	0.059 (2)
C(24)	0.4189 (5)	0.4853 (3)	0.7326 (4)	0.059 (2)
C(25)	0.5135 (5)	0.4270 (4)	0.6813 (4)	0.064 (2)
C(26)	0.4467 (4)	0.3559 (4)	0.5515 (4)	0.056 (2)
		II		
Sn	0.22184 (3)	0.24047 (3)	0.07284 (2)	0.0470 (1)
Cl	0.0951 (2)	0.1091 (2)	−0.2125 (1)	0.0785 (6)
O	0.3430 (3)	0.3538 (3)	0.3438 (3)	0.052 (1)
N	0.4322 (4)	0.2899 (3)	0.4374 (3)	0.045 (1)
C(1)	0.3854 (6)	0.3744 (5)	0.0455 (5)	0.066 (2)
C(2)	−0.0076 (6)	0.3585 (7)	0.1284 (6)	0.083 (3)
C(3)	0.2991 (6)	0.0255 (4)	0.0875 (5)	0.065 (2)
C(4)	0.6043 (4)	0.2694 (4)	0.4528 (4)	0.051 (2)
C(5)	0.6967 (5)	0.2156 (5)	0.5591 (5)	0.068 (2)
C(6)	0.6208 (6)	0.1819 (5)	0.6478 (5)	0.072 (2)
C(7)	0.4485 (6)	0.2003 (5)	0.6276 (5)	0.070 (2)
C(8)	0.3514 (5)	0.2521 (4)	0.5209 (4)	0.054 (2)
C(9)	0.6794 (5)	0.3075 (5)	0.3537 (5)	0.065 (2)
C(10)	0.1633 (6)	0.2739 (6)	0.4904 (6)	0.076 (2)

[a] Estimated standard deviations for the last digit in parentheses.

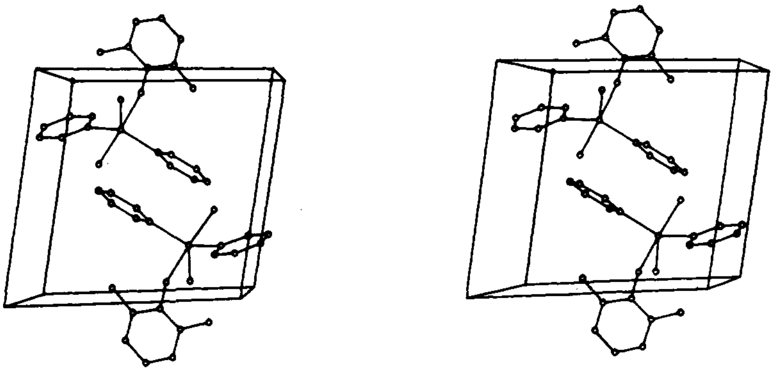

Figure 3. Unit-cell packing diagram for I as viewed along the *a* axis.

isotropically, and in II, the hydrogen atoms were placed in idealized locations [d(C–H) = 0.96 Å] but not refined. In both cases an initial assumption of the centrosymmetric, triclinic space group $P\bar{1}$ was shown to be correct by the successful and chemically reasonable structural solutions ultimately obtained.

All calculations were performed by using a Data General Nova 4 computer with programs contained in the Nicolet libraries P3, XP, and SHELXTL.

Positional and thermal parameters for the non-hydrogen atoms are given in Table II, selected bond distances and angles in Table III, and least-squares planes in Table IV; tables of hydrogen atom data, structure factors, and anisotropic thermal parameters are available as supplementary material.

The ORTEP drawing of diphenyltin(IV) dichloride–2,6-dimethylpyridine *N*-oxide is shown in Figure 1 and that of trimethyltin(IV) chloride–2,6-dimethylpyridine *N*-oxide in Figure 2. Figures 3 and 4 are their respective stereo packing diagrams.

Description of Structures and Discussion

An examination of the unit-cell contents reveals no intermolecular contacts. Thus the title compounds are indeed monomeric, and I is the second authenticated example of a 1:1, five-coordinated, diorganotin(IV) dihalide complex.[6,15]

The Structure of I. In the analogous 1:1 dimethyl-

Table III. Selected Bond Distances and Angles in I and II[a]

I		II	
	(a) Bond Distances (Å)		
Sn-Cl(1)	2.366 (1)	Sn-Cl	2.533 (1)
Sn-Cl(2)	2.471 (1)	Sn-O	2.400 (2)
Sn-O	2.296 (2)	Sn-C(1)	2.125 (6)
Sn-C(11)	2.128 (4)	Sn-C(2)	2.119 (5)
Sn-C(21)	2.119 (3)	Sn-C(3)	2.120 (4)
O-N	1.346 (4)	O-N	1.326 (4)
	(b) Bond Angles (deg)		
Sn-O-N	125.7 (2)	Sn-O-N	130.6 (2)
O-Sn-Cl(2)	172.8 (1)	O-Sn-Cl	177.4 (1)
O-Sn-Cl(1)	83.9 (1)	O-Sn-C(1)	88.1 (1)
O-Sn-C(11)	87.8 (1)	O-Sn-C(2)	86.0 (2)
O-Sn-C(21)	88.1 (1)	O-Sn-C(3)	85.7 (1)
Cl(2)-Sn-Cl(1)	88.92 (4)	Cl-Sn-C(1)	93.7 (1)
Cl(2)-Sn-C(11)	96.0 (1)	Cl-Sn-C(2)	94.9 (1)
Cl(2)-Sn-C(21)	94.9 (1)	Cl-Sn-C(3)	91.7 (1)
Cl(1)-Sn-C(11)	120.2 (1)	C(1)-Sn-C(2)	114.1 (2)
Cl(1)-Sn-C(21)	114.7 (1)	C(1)-Sn-C(3)	123.1 (2)
C(11)-Sn-C(21)	124.1 (1)	C(2)-Sn-C(3)	121.8 (3)

[a] Estimated standard deviations for the last digit in parentheses.

tin(IV) dichloride complex the methyl-tin-methyl angle is opened from 123.5° in $(CH_3)_2SnCl_2$[7] to 145.3° in the trans-trans-trans octahedral chlorine-bridged dimer.[3] With the weaker Lewis acid the angles involving the bulkier phenyl groups change from 123.9° and 127.0° in $(C_6H_5)_2SnCl_2$[10,11] to 124.1 (1)° in I. The first-listed angle corresponds to a diphenyltin(IV) dichloride molecule, one of whose chlorine atoms bridge to an adjacent tin atom at 3.78 Å.[11] Its tin atom is four coordinated in a distorted tetrahedral environment. The tin atom in I, on the other hand, is five coordinated in a trigonal-bipyramidal arrangement. The two phenyl groups and one chlorine atom are equatorial, while the ligand oxygen and the other chlorine atom are axial [angle O–Sn–Cl(2) = 172.8 (1)°]. The sum of the angles in the trigonal girdle of the complex is 359.0 (3)°. The axial tin–chlorine bond [*d*(Sn–Cl(2) = 2.471 (1) Å] is longer than the equatorial [*d*(Sn–Cl(1)) = 2.366 (1) Å] by 0.105 (1) Å. In comparison to the previously reported benzthiazole·Ph_2SnCl_2 complex,[6] I possesses a much more regular trigonal-bipyramidal geometry; e.g., the range of the bond angles in the equitorial plane for 1 is 114.7–124.1° (109.2–132.6° for the benzthiazole complex) and the range of the axial–equitorial angles in 1 is 83.9–96.0° (vs. 82.1–98.2°).

Structure of II. The chlorine atom occupies one apical site while the oxygen atom of the ligand occupies the other at an O–Sn–Cl angle of 177.4 (1)°. The sum of the angles made by the three equatorial methyl groups at the central tin atom is 358.9 (3)°, but the tin atom is displaced out of this plane toward the chlorine atom by 0.2357 (2) Å. That the girdle of this complex is not flat is suggested by the observation of two bands in the ν(Sn–C) region of both the infrared (538, 520 cm^{-1}) and Raman (561, 530 cm^{-1}) in the expected reversed intensity ratios using solid samples in both instances.

Bonding in the Pyridine *N*-Oxide Complexes. A number of molecular structure parameters reflect the bonding situation in pyridine *N*-oxide complexes. These include the N→O and the Sn–O distances, the angle Sn–O–N, and the angle made between the planes containing the Sn–O–N system, and the pyridine ring which intersects at the nitrogen atom.

It would seem obvious that coordination of the oxygen would lengthen the N→O distance over that of the ligand, but our survey of the structures of over 30 complexes of

Table IV. Comparison Molecular Data for Organotin(IV) Chlorides and Their Pyridine *N*-Oxide Complexes

	$(CH_3)_3SnCl\cdot n2,6\text{-}R_2C_5H_3NO$		$(CH_3)_2SnCl_2\cdot n2,6\text{-}R_2C_5H_3NO$			$(C_6H_5)_2SnCl_2\cdot n2,6\text{-}R_2C_5H_3NO$		
	$n = 1$, R = CH_3 (II)[a]	$n = 0$[b]	$n = 2$, R = H[c]	$n = 1$, R = CH_3[d]	$n = 0$[e]	$n = 1$, R = CH_3 (I)[a]	$n = 0$[f]	
d(Sn-Cl), Å	2.533 (1)	2.430 3.269	2.584	2.400 2.528	2.40 3.54	2.366 (1) 2.471 (1)	2.336 2.353	2.336 2.357
d(Sn-O), Å	2.400 (2)		2.251	2.289		2.296 (2)		
d(Sn-C), Å	2.119 (5) 2.120 (4) 2.125 (6)	2.109 2.121 2.126	2.225	2.110 2.112	2.21	2.119 (3) 2.128 (4)	2.105 2.119	2.112 2.118
d(O-N), Å	1.326 (4)		1.37	1.352		1.346 (4)		
d(N-C)$_{av}$, Å	1.367 (4)		1.35	1.364		1.360 (3)		
∠C-Sn-C, deg	114.1 (2) 121.8 (3) 123.1 (2)	115.2 116.3 119.8	180	145.3	123.5	124.1 (1)	123.9	127.0
∠Cl-Sn-Cl, deg		176.85	180	79.46 89.50 168.79	93.0	88.92 (4)	101.7	97.8
∠Cl-Sn-O, deg	177.4 (1)		89.5 90.5	87.95 103.09 177.44		83.9 (1) 172.8 (1)		
∠C-Sn-O, deg	85.7 (1) 86.0 (2) 88.1 (1)		84.4 95.6	85.3 87.4		87.8 (1) 88.8 (1)		
∠Sn-O-N, deg	130.6 (2)		117	123.0		125.7 (2)		
∠Cl-Sn-C, deg	91.7 (1) 93.7 (1) 94.9 (1)	82.6 79.6 78.0 99.1 100.1 100.6	89.5 90.5	72.3 76.4 93.7 95.1 102.9 110.7	109.0	94.9 (1) 96.0 (1) 114.7 (1) 120.2 (1)	106.0 106.5 107.8 108.8	105.0 105.4 107.1 110.1
∠O-Sn-O, deg			180					
∠pyNO ring and Sn-O-N plane, deg	84.75 (16)		83	87.17		86.98 (4)		

[a] This work. [b] Reference 8. [c] Reference 16. [d] Reference 3. [e] Reference 7. [f] Reference 10 and 11. There are two crystallographically independent molecules in the unit cell.

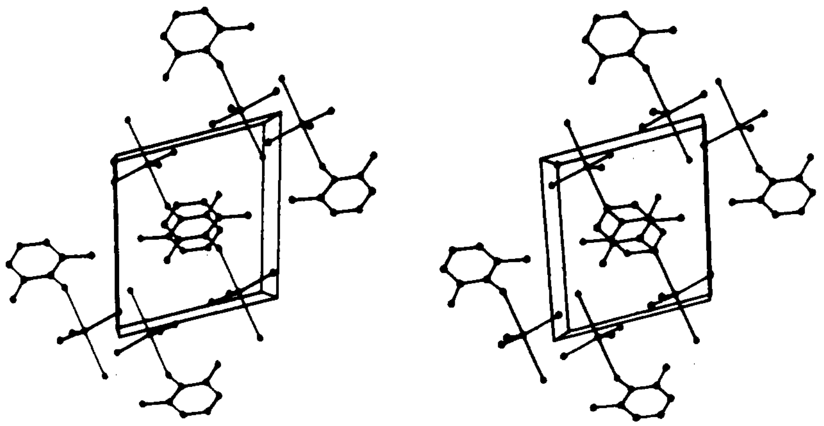

Figure 4. Unit-cell packing diagram for II as viewed along the *b* axis.

Table V. Comparison Infrared Data for the 1:1 Adducts of 2,6-Dimethylpyridine *N*-Oxide with Tin(II) and Tin(IV) Chlorides (in cm^{-1})

	ν(N-O)	ν(Sn-C)
$2,6\text{-}(CH_3)_2C_5H_3NO$	1246[a]	
$SnCl_2\cdot 2,6\text{-}(CH_3)_2C_5H_3NO$	1181[b]	
$[(CH_3)_2SnCl_2\cdot 2,6\text{-}(CH_3)_2C_5H_3NO]_2$	1172	546, 518[c]
$(C_6H_5)_2SnCl_2\cdot 2,6\text{-}(CH_3)_2C_5H_3NO$ (I)	1205[d]	
$(CH_3)_3SnCl\cdot 2,6\text{-}(CH_3)_2C_5H_3NO$ (II)	1200	538[d]
$(C_6H_5)_3SnCl\cdot 2,6\text{-}(CH_3)_2C_5H_3NO$	1210[e]	

[a] Reference 18. [b] Reference 19. [c] Reference 3. [d] This work. [e] Ng, S. W.; Zuckerman, J. J., unpublished results.

variously substituted pyridine *N*-oxides fails to confirm this expectation.[3] The claimed correlations between the frequencies assigned to ν(N→O) modes and complexation must, therefore, arise from other origins. The structure of 2,6-dimethylpyridine (2,6-lutidine) is unknown, so no direct comparison can be made.

The structures of four organotin(IV) complexes with pyridine *N*-oxide ligands are available for comparison at this writing, but any conclusion based on these Sn–O bond distances would involve cross-comparing different tin coordination numbers and different organic substituents at the tin and on the ligand. The *d*(Sn–O) data are listed, inter alia, in Table IV.

The angles M–O–N in pyridine *N*-oxide–metal (M) complexes are generally close to 120° reflecting sp^2 hybridization at the coordinated oxygen atom.[3] These values for the M = Sn complexes are listed in Table IV and are 125.7 (2) and 130.6 (2)° for I and II, respectively. The sp^2 hybridization would leave one lone pair of electrons in a pure p orbital that would be available for π overlap with the aromatic pyridine system. The extent of this interaction would be dependent upon the angle between the Sn–O–N plane and the plane of the aromatic ring and would maximize when the two were coincident. However, our survey reveals that the two planes are in general nearly perpendicular, the smallest interplane angle being 41° in $[(2\text{-}CH_3pyNO)_5Co]^{2+}(ClO_4^-)_2$.[17] The angles in the organotin complexes are listed in Table IV with the values being 86.98 (4)° and 84.75 (16)° for I and II, respectively. Thus a description based upon distortion from sp^3 hybridization at oxygen is to be preferred. Forcing the Sn–O–N and pyridine ring into the same plane would give rise to severe steric repulsions between the 2,6-dimethyl groups and the substituents on the tin atom.

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Supplementary Material Available: Tables of hydrogen atom data, structure factors, and anisotropic thermal parameters (39 pages). Ordering information is given on any current masthead page.

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