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Crystal Structures of 2,2-Dimethyltriazanium Chloride and 1,1,1-Trimethylhydrazinium Chloride

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The crystal structures of 1,1,1-trimethylhydrazinium chloride, $[(CH_3)_3NNH_2]^+Cl^-$, I, and 2,2-dimethyltriazanium chloride, $[(CH_3)_2N(NH_2)_2]^+Cl^-$, II, have been determined from three-dimensional single-crystal x-ray diffraction data collected on a four-circle automated diffractometer. Both compounds crystallize in the monoclinic space group $P2_1/c$ with $a = 7.316$ (3) Å, $b = 5.502$ (2) Å, $c = 15.194$ (6) Å, and $\beta = 93.08$ (3)° for I and $a = 7.181$ (4) Å, $b = 5.444$ (2) Å, $c = 14.618$ (4) Å, and $\beta = 91.03$ (3)° for II. Both compounds contain 4 formula units/unit cell. Structures I and II were solved by the heavy-atom method and refined by full-matrix least-squares techniques to respective R factors of 3.7% for 1005 reflections and 5.3% for 541 reflections. The chloride ions are hydrogen bonded to the cations through the amino hydrogens. The cations consist of central nitrogens surrounded tetrahedrally by one amino and three methyl groups (I) and by two amino and two methyl groups (II). The respective average C–N distances, 1.493 (4) and 1.490 (7) Å, are normal as are the various N–H and C–H distances. The N–N distances of 1.463 (3) Å in I and 1.439 (6) and 1.462 (6) Å in II are reasonable but appear to be affected by differences in hydrogen bonding. The suggestion that the stability of II results from the quaternization of the central nitrogen is discussed.

The 2,2-dialkyltriazanium salts formed by chloramination reactions[1–4] are the first stable alkyl compounds known which contain a chain of more than two nitrogen atoms bonded by single bonds. They are stable in air and in neutral aqueous solutions[5] and can be recrystallized from boiling solvents such as acetone, ethanol, and acetonitrile.

The crystal structure determination of 2,2-dimethyltriazanium chloride, II, was undertaken to confirm the structural formula $[(CH_3)_2N(NH_2)_2]^+Cl^-$ postulated on the basis of ir and NMR spectra. In addition, structural features might explain the stability of 2,2-dialkyltriazanium salts. We also determined the crystal structure of 1,1,1-trimethylhydrazinium[6] chloride (I) for comparison with II. The compounds discussed here are the first 2,2-dialkyltriazanium and 1,1,1-trialkylhydrazinium salts whose structures have been determined by x-ray crystallography.

Experimental Section

Materials. I and II were prepared by the respective chloraminations of trimethylamine[6] and 1,1-dimethylhydrazine[2] using the chloramine–ammonia effluent from a Sisler–Mattair generator.[7] Suitable single crystals were obtained by dissolving the recrystallized compounds in boiling acetone–ethanol mixtures (4:1 v/v) and allowing the solutions to stand overnight. The colorless crystals which had formed were collected by filtration, washed with ether, and dried under vacuum. Subsequent manipulations were carried out in a drybox to prevent deliquescence Anal. Calcd for $C_3H_{11}N_2Cl$: C, 32.58; H, 9.96; N, 25.34. Found for I: C, 32.41; H, 10.03; N, 25.30. Calcd for $C_2H_{10}N_3Cl$: C, 21.52; H, 8.97; N, 37.62. Found for II: C, 21.62; H, 8.96; N, 37.50. Elemental analyses were carried out by Galbraith Laboratories, Inc., Knoxville, Tenn.

Unit Cell and Space Group Determination. Precession and Weissenberg photographs of crystals of I and II showed both compounds to be monoclinic. Systematic absences of $0k0$, $k = 2n + 1$, and of $h0l$, $l = 2n + 1$, indicated the space group $P2_1/c$. The unit cell parameters were determined by a least-squares fit of 2θ, ω, ϕ, and χ for 15 reflections. Densities were measured by flotation in chloroform–hexane solutions.

Table I. Crystal Data

	$[(CH_3)_3NNH_2]^+Cl^-$	$[(CH_3)_2N(NH_2)_2]^+Cl^-$
a, Å	7.316 (3)	7.181 (4)
b, Å	5.502 (2)	5.444 (2)
c, Å	15.194 (6)	14.618 (4)
β, deg	93.08 (3)	91.03 (3)
Vol., Å^3	610.7 (4)	571.1 (4)
Formula wt	110.5	111.5
Z, formula units/unit cell	4	4
ρ(calcd), g/cm^3	1.20	1.29
ρ(measd), g/cm^3	1.20	1.29
Crystal dimensions, mm	0.20 × 0.05 × 0.15	0.11 × 0.06 × 0.05
Radiation	Mo Kα	Cu Kα
μ, cm^{-1}	5.0	48.5
2θ range, deg	0–50	0–100
No. of unique reflections	1087	584
No. of obsd reflections	1005	541

Pertinent crystal data are listed in Table I.

Collection of Intensity Data. Crystals of I and II mounted in thin-walled glass capillary tubes were used for intensity data collection. Data were gathered on a Syntex $P\bar{1}$ diffractometer using a θ–2θ scan with graphite-monochromated Mo Kα radiation (λ 0.71069 Å) for I and Ni-filtered Cu Kα radiation (λ 1.5418 Å) for II. Background was measured on either side of the peaks for a time equal to half the scan time. The scan rates were 1.0°/min for reflections of $I < 150$ s^{-1}, and 24.0°/min for reflections of $I > 1500$ s^{-1} with a linear interpolation between these limits.

Four standard reflections, whose intensities were measured after every 96 reflections, were used to correct for variations of intensities with time. Standard reflections of crystal I showed no significant change during data collection. However, in the case of II, the standard reflections remained essentially constant at first and then decreased sharply near the end of data collection. The intensities of the standards, measured after the last 50 reflections, were ca. 0.4 times their initial values. The crystal of II, initially colorless, had become opaque by

Table II. Scheme of Refinement[a]

	$[(CH_3)_3\text{-}NNH_2]^+Cl^-$	$[(CH_3)_2\text{-}N(NH_2)_2]^+Cl^-$
R after location of non-H atoms	24.4	21.1
R after isotropic refinement	9.0	10.6
R after anisotropic ref on non-H atoms	7.0	8.2
R final	3.7	5.3
F(low)	2.4	3.4

[a] The *R* factor is $[\Sigma\|F_o| - |F_c\|/\Sigma|F_o|] \times 100$.

the end of data collection, indicating that some decomposition had occurred.

Reflections of $I < 2\sigma(I)$ were classified as unreliable and were not employed in the solution or refinement of the structures. No corrections were made for absorption because of the small value of μr (0.03–0.10 for I and 0.13–0.26 for II).

Structure Determination and Refinement

Both structures were solved by the heavy-atom method and refined by full-matrix least-squares techniques. Scattering factors for Cl^- were obtained from Doyle and Turner,[8] those for C and N from Hanson et al.,[9] and those for H from Stewart, Davidson, and Simpson.[10]

The chloride ion in each case was located in the Patterson function and the "light" atoms were found in the Fourier synthesis phased on the chloride ions. For the first three least-squares cycles the four terminal atoms of the cations of I and II were assigned carbon scattering factors. Those atoms which were found to have "abnormally low" isotropic thermal parameters were treated as N in all subsequent least-square cycles. After three more least-squares cycles using isotropic thermal parameters and three cycles using anisotropic thermal parameters, difference Fourier syntheses were calculated. In the case of I, 11 peaks of 0.58–0.72 e/Å^3 were located and for II 10 peaks of 0.40–0.67 e/Å^3 were found. In both cases these were the highest peaks in the maps. Bond distances and angles were found to be reasonable for methyl and amino hydrogens. During the next three least-squares cycles only nonhydrogen atoms were refined. For the last three least-squares cycles, the hydrogens were refined with isotropic thermal parameters and the other atoms with anisotropic thermal parameters. During the last cycle shifts of all parameters were less than one-tenth an esd. Final difference Fourier syntheses were relatively featureless showing no peaks greater than 0.26 e/Å^3.

The quantity minimized during least-squares refinement was $\sum w(|F_o| - |F_c|)^2$ where $w^{1/2} = F_o/F(\text{low})$ if $F_o < F(\text{low})$, $w^{1/2} = 1$ if $F(\text{low}) \leq F_o \leq 4F(\text{low})$, and $w^{1/2} = 4F(\text{low})/F_o$ if $F_o > 4F(\text{low})$. The value of *F*(low) was 2.7 times the minimum observable *F* for I and 2.4 times the minimum observable *F* for II. Values of *F*(low) and *R* factors at various stages of refinement are listed in Table II. Final positional and anisotropic thermal parameters of nonhydrogen atoms for both compounds are listed in Table III; hydrogen positional and isotropic thermal parameters are listed in Table IV. Structure factors are available on request.[11]

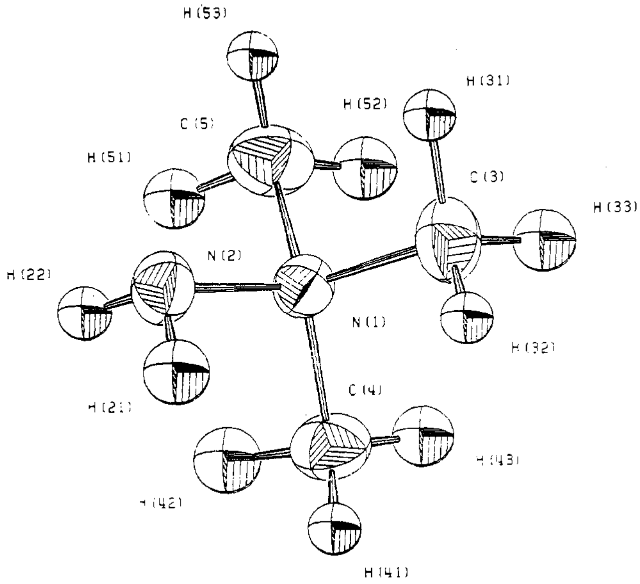

Figure 1. Geometry and atomic numbering of the $[(CH_3)_3\text{-}NNH_2]^+$ cation. Hydrogen isotropic thermal parameters used to calculate this drawing are half the values listed in Table IV (for clarity).

Description and Discussion of the Structures

The crystals of each salt consist of approximately tetrahedral cations, bonded by hydrogen bonds to chloride ions. The central nitrogen in the cation of I is surrounded by one amino and three methyl groups (Figure 1); the central nitrogen in the cation of II is surrounded by two amino and two methyl groups (Figure 2). The average angle between the central nitrogen and the terminal atoms in each cation equals the tetrahedral angle. The average C–N distances in the cations of I and II, 1.493 (4) and 1.490 (7) Å, respectively, are not significantly different and are similar to the values of 1.470 (14) Å in $(CH_3)_4NClO_4$[12] and 1.51 (4) Å in $(CH_3)_4NCl\cdot 5SeOCl$[13]. The N–N distance of 1.463 (4) Å in I and the average N–N distance of 1.450 (6) Å in II are similar to the N–N distance of 1.45 (3) Å found for 1,1-dimethylhydrazine[14] and are in the range 1.44 (4) Å reported for N–N single bonds.[15] The N–H and C–H distances[16] and H–N–H and H–C–H angles are reasonable for both compounds. Internuclear distances and angles for I are listed in Table V and for II in Table VI.

The distances between the chloride ions and the amino hydrogens in I, 2.57 (4) and 2.52 (3) Å, are shorter than the minimum H···Cl nonbonded distance, 3.0 Å (the sum of the chloride ionic radius and the hydrogen van der Waals radius[17]),

Table III. Positional and Anisotropic Thermal Parameters for Nonhydrogen Atoms in $[(CH_3H_3)NNH_2]^+Cl^-$ and $[(CH_3)_2N(NH_2)_2]^+Cl^-$ [a]

Atom	*x*	*y*	*z*	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
				$[(CH_3)_3NNH_2]^+Cl^-$					
Cl	7933 (1)	6275 (1)	1160 (1)	146 (1)	270 (2)	33 (1)	−37 (2)	12 (1)	−23 (1)
N(1)	7411 (2)	4788 (3)	3669 (1)	124 (3)	205 (6)	26 (1)	−16 (6)	0 (2)	−1 (3)
N(2)	7758 (3)	2975 (4)	2994 (1)	172 (4)	268 (7)	28 (1)	−30 (8)	6 (3)	−27 (4)
C(3)	7266 (4)	3636 (5)	4512 (2)	231 (6)	304 (9)	28 (1)	−24 (12)	11 (4)	22 (5)
C(4)	8903 (3)	6651 (4)	3750 (2)	155 (5)	259 (8)	46 (1)	−88 (10)	7 (4)	−28 (5)
C(5)	5628 (3)	5976 (5)	3410 (2)	135 (4)	347 (9)	50 (1)	60 (11)	7 (4)	30 (6)
				$[(CH_3)_2N(NH_2)_2]^+Cl^-$					
Cl	7891 (2)	6592 (2)	4058 (1)	224 (3)	336 (6)	39 (1)	3 (7)	23 (2)	−17 (3)
N(1)	7574 (5)	4760 (7)	1397 (2)	192 (10)	254 (16)	37 (22)	−53 (23)	16 (7)	−10 (10)
N(2)	7847 (7)	3121 (10)	2160 (3)	280 (14)	371 (20)	40 (27)	−37 (28)	0 (9)	72 (13)
N(3)	9215 (7)	6319 (9)	1292 (3)	218 (12)	349 (20)	55 (29)	−151 (27)	24 (9)	−23 (13)
C(4)	7170 (9)	3199 (12)	0577 (4)	235 (15)	337 (23)	44 (31)	−25 (35)	−4 (11)	−62 (16)
C(5)	5970 (9)	6399 (12)	1587 (5)	236 (16)	349 (25)	62 (40)	110 (35)	33 (12)	−36 (17)

[a] All parameters have been multiplied by 10^4. The form of the anisotropic thermal ellipsoid is $\exp[-(\beta_{11}h^2 + \beta_{22}k^2 + \beta_{33}l^2 + \beta_{12}hk + \beta_{13}hl + \beta_{23}kl)]$. Standard deviations are given in parentheses in this and in all subsequent tables.

Table IV. Hydrogen Parameters for $[(CH_3)_3NNH_2]^+Cl^-$ and $[(CH_3)_2N(NH_2)_2]^+Cl^-$ [a]

Atom	x	y	z	B_{iso}, A^2
		$[(CH_3)_3NNH_2]^+Cl^-$		
H(21)	812 (5)	234 (7)	312 (2)	5.4 (7)
H(22)	783 (4)	374 (5)	254 (2)	3.7 (6)
H(31)	604 (4)	232 (5)	444 (2)	3.4 (5)
H(32)	840 (4)	258 (5)	467 (2)	3.4 (5)
H(33)	764 (4)	468 (6)	497 (2)	4.9 (6)
H(41)	1011 (4)	567 (5)	384 (2)	3.5 (5)
H(42)	879 (4)	755 (7)	315 (2)	5.7 (7)
H(43)	859 (4)	770 (6)	420 (2)	4.7 (6)
H(51)	583 (4)	679 (5)	285 (2)	4.6 (6)
H(52)	551 (4)	716 (6)	386 (2)	5.1 (7)
H(53)	466 (4)	474 (5)	335 (2)	3.2 (5)
		$[(CH_3)_2N(NH_2)_2]^+Cl^-$		
H(21)	905 (11)	231 (16)	211 (5)	8 (2)
H(22)	793 (9)	414 (13)	266 (5)	7 (2)
H(31)	1033 (9)	530 (12)	126 (4)	5 (2)
H(32)	895 (10)	727 (15)	72 (6)	8 (2)
H(41)	839 (8)	209 (11)	56 (4)	5 (1)
H(42)	590 (10)	223 (13)	66 (4)	7 (2)
H(43)	707 (7)	440 (11)	5 (4)	5 (1)
H(51)	476 (8)	534 (10)	163 (3)	4 (1)
H(52)	634 (8)	735 (14)	217 (5)	6 (2)
H(53)	576 (9)	757 (14)	100 (5)	7 (2)

[a] Positional parameters are × 10^3.

Table V. Internuclear Distances (A) and Angles (deg) for $[(CH_3)_3NNH_2]^+Cl^-$

Distances

N(1)-N(2)	1.463 (3)	C(4)-H(42)	1.04 (3)
N(1)-C(3)	1.490 (3)	C(4)-H(43)	0.93 (3)
N(1)-C(4)	1.497 (3)	C(5)-H(51)	0.98 (3)
N(1)-C(5)	1.493 (3)	C(5)-H(52)	0.96 (3)
N(2)-H(21)	0.92 (4)	C(5)-H(53)	0.98 (3)
N(2)-H(22)	0.82 (3)	Cl-H(21′)	2.57 (4)
C(3)-H(31)	1.09 (3)	Cl-H(22)	2.52 (3)
C(3)-H(32)	0.97 (3)	Cl-N(2)	3.334 (2)
C(3)-H(33)	0.99 (3)	Cl-N(2′)	3.468 (2)
C(4)-H(41)	1.04 (3)		

Angles

N(2)-N(1)-C(3)	106.6 (2)	H(32)-C(3)-H(33)	109 (2)
N(2)-N(1)-C(4)	111.8 (2)	H(21′)-Cl-H(22)	80 (1)
N(2)-N(1)-C(5)	107.2 (2)	H(41)-C(4)-H(42)	112 (2)
C(3)-N(1)-C(4)	110.9 (2)	H(41)-C(4)-H(43)	118 (3)
C(3)-N(1)-C(5)	110.1 (2)	H(42)-C(4)-H(43)	110 (3)
C(4)-N(1)-C(5)	110.2 (2)	H(51)-C(5)-H(52)	109 (3)
H(21)-N(2)-H(22)	105 (3)	H(51)-C(5)-H(53)	113 (3)
H(31)-C(3)-H(32)	116 (2)	H(52)-C(5)-H(53)	116 (3)
H(31)-C(3)-H(33)	107 (2)		

Table VI. Internuclear Distances (A) and Angles (deg) for $[(CH_3)_2N(NH_2)_2]^+Cl^-$

Distances

N(1)-N(2)	1.439 (6)	C(4)-H(43)	1.01 (6)
N(1)-N(3)	1.462 (6)	C(5)-H(51)	1.05 (6)
N(1)-C(4)	1.494 (7)	C(5)-H(52)	1.03 (7)
N(1)-C(5)	1.487 (8)	C(5)-H(53)	1.07 (7)
N(2)-H(21)	0.97 (8)	Cl-H(21′)	2.84 (8)
N(2)-H(22)	0.91 (7)	Cl-H(22)	2.45 (7)
N(3)-H(31)	0.98 (6)	Cl-H(31′)	2.44 (6)
N(3)-H(32)	1.00 (8)	Cl-H(32′)	2.60 (8)
C(4)-H(41)	1.07 (6)	Cl-N(2)	3.356 (5)
C(4)-H(42)	1.06 (7)	Cl-N(3′)	3.353 (5)

Angles

N(2)-N(1)-N(3)	110.0 (4)	H(41)-C(4)-H(43)	114 (4)
N(2)-N(1)-C(4)	106.9 (4)	H(42)-C(4)-H(43)	111 (5)
N(2)-N(1)-C(5)	108.8 (4)	H(51)-C(5)-H(52)	116 (5)
N(3)-N(1)-C(4)	113.0 (4)	H(51)-C(5)-H(53)	105 (5)
N(3)-N(1)-C(5)	107.4 (4)	H(52)-C(5)-H(53)	113 (6)
C(4)-N(1)-C(5)	110.6 (4)	H(22)-Cl-H(31′)	106 (2)
H(21)-N(2)-H(22)	106 (6)	H(22)-Cl-H(32′)	154 (2)
H(31)-N(3)-H(32)	113 (6)	H(31′)-Cl-H(32′)	80 (2)
H(41)-C(4)-H(42)	115 (5)		

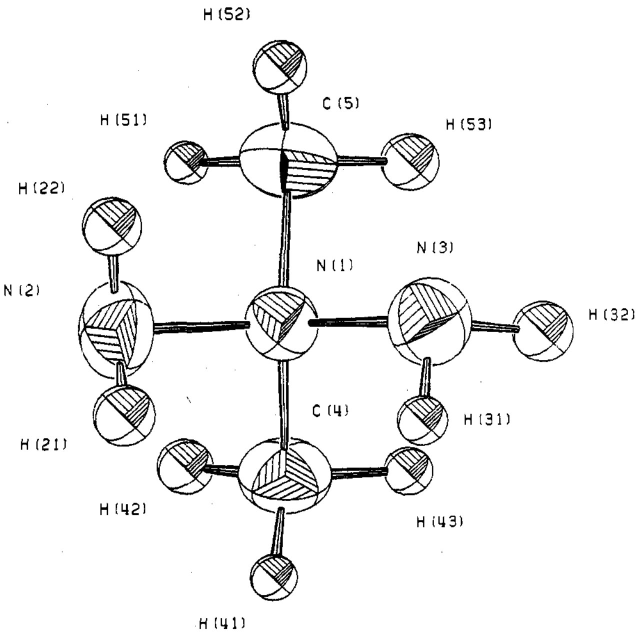

Figure 2. Geometry and atomic numbering of the $[(CH_3)_2N(NH_2)_2]^+$ cation. Hydrogen isotropic thermal parameters used to calculate this drawing are one-fourth the values listed in Table IV (for clarity).

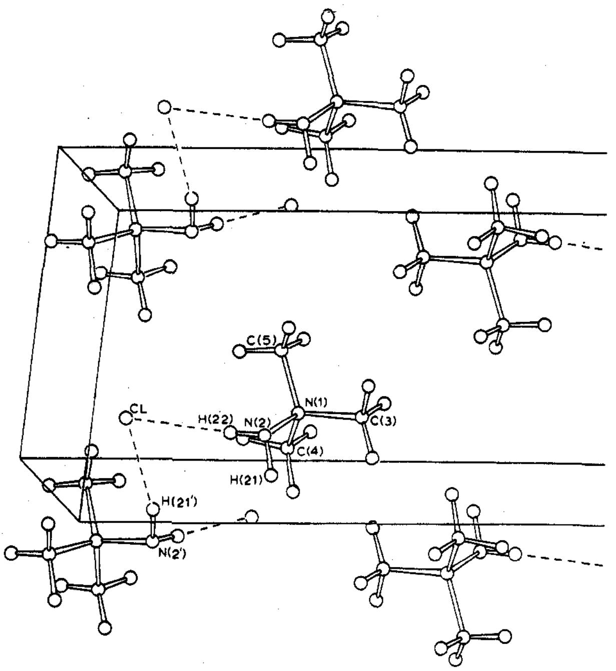

Figure 3. Portion of the unit cell of $[(CH_3)_3NNH_2]^+Cl^-$ showing packing and hydrogen bonding. The a axis runs from top to bottom, b from front to back, and c from left to right.

indicating that both amino hydrogens form hydrogen bonds to chloride ions. The two hydrogen bonds H(22)···Cl and Cl···H(21′) link the cation at x, y, z with the cation at $\bar{x}$, $^1/_2 + y$, $^1/_2 - z$. The result is a spiral with amino groups on the inside and methyl groups on the outside (Figure 3). Hydrogen bonds have also been reported in the structures of the salts of hydrazinium cations. For example, the short N···Cl distances of 3.10 (4) Å in $N_2H_6Cl_2$ indicate that hydrogen bonds are present.[18]

Three amino hydrogens in II, H(22), H(31), and H(32) (whose H···Cl distances are 2.45 (7), 2.44 (6), and 2.60 (8) Å) form hydrogen bonds. The H(21″)···Cl distance of 2.84 (8) Å may not be significantly shorter than the minimum H···Cl nonbonded distance (t_0 = 2.0).[19] Therefore, it is

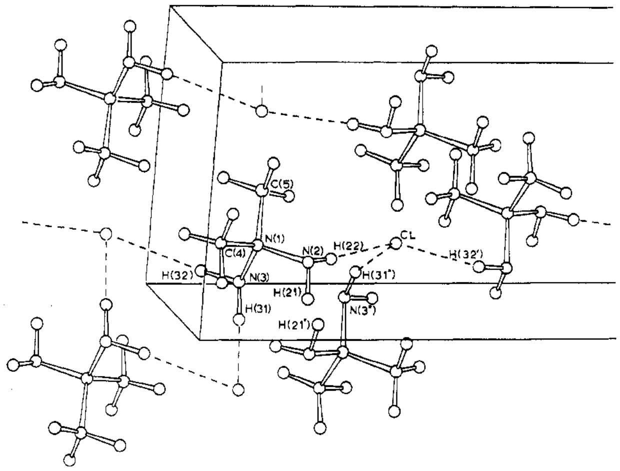

Figure 4. Portion of the unit cell of $[(CH_3)_2N(NH_2)_2]^+Cl^-$ showing packing and hydrogen bonding. The *a* axis runs from top to bottom, *b* from front to back, and *c* from left to right.

questionable whether H(21) forms a hydrogen bond.

The two hydrogen bonds H(22)···Cl and Cl···H(32') link a cation at x, y, z with a cation at $x, 1 + (^1/_2 - y), ^1/_2 + z$. The result is a chain along *c*. Each chain forms a pair with another chain through Cl···H(31'') bonds. The amino groups in each chain point toward the opposite chain of the pair and the methyl groups point away (Figure 4).

It has been reported that covalent bond distances can be affected by the presence of nearby hydrogen bonds.[20,21] The unusually long N–N distance of 1.474 (7) Å in $(N_2H_6)(TiF_6)$ is believed to result from unusually strong hydrogen bonding (N–H···F).[20] One of the chlorine atoms in dichloro(acetone thiosemicarbazone)zinc(II) which is involved in hydrogen bonding is 0.049 (2) Å further from the zinc than the other chlorine which is not.[21]

In the structures discussed here N–N bond distances may also be affected by hydrogen bonding. The N(1)–N(3) distance in II (virtually identical with the N–N distance in I) is 0.023 Å longer than the N(1)–N(2) distance. According to Cruickshank's test[19] this difference is probably significant ($t_0 = 2.54$, t_0 for $p = 0.01$ is 2.58). Both hydrogens bonded to N(3) in II as well as those bonded to the terminal nitrogen in I are bonded to chloride ions by hydrogen bonds; however, whereas one of the hydrogens bonded to N(2) in II forms a hydrogen bond, the other forms either a weak hydrogen bond or none at all.

Conclusions

The structures of the two cations are quite similar to that of the tetramethylammonium cation. There are no significant differences in C–N bond lengths and R–N–R angles as a result of replacing terminal methyl groups with amino groups. The N–N bond distances are reasonable for N–N single bonds. Small differences in the two N–N bond distances in II can be explained by differences in hydrogen bonding. Therefore, I and II can be thought of as amino-substituted quaternary ammonium salts.

We believe that the stability of II and other 2,2-dialkyltriazanium salts is a result of the quaternization of the central nitrogen. Triazanium chloride $[(NH_2NH_2NH_2)Cl]$, formed at −50 °C by the chloramination of hydrazine, decomposes upon being warmed[2]

$$2N_3H_6Cl \rightarrow 2NH_4Cl + N_2H_4 + N_2$$

This decomposition appears to involve disproportionation as both oxidation and reduction products are present. If we assume that the first step of the decomposition of triazanium chloride is the loss of hydrogen atoms and the formation of a nitrogen–nitrogen double bond (which is the first step in the oxidation of compounds with nitrogen–nitrogen single bonds[22]), then the reason for the stability of 2,2-dialkyltriazanium salts becomes evident. The central nitrogen in I cannot form a nitrogen–nitrogen double bond without first breaking an existing C–N or N–N bond. Therefore, decomposition through disproportionation cannot readily occur. We believe that this explanation is reasonable, although somewhat speculative.

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Registry No. I, 5675-48-9; II, 13166-44-4.

Supplementary Material Available: Listings of structure factor amplitudes (8 pages). Ordering information is given on any current masthead page.

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