Table 3. *Hydrogen bonds (e.s.d.'s are in parentheses)*

D—H···*A*	*D*···*A* (Å)	*D*—H (Å)	H···*A* (Å)	*D*—H···*A* (°)
N—H(101)^{i}···O(1)^{ii}	2·782 (4)	0·89 (5)	2·02 (5)	143 (3)
···O(23)^{ii}	3·259 (4)		2·63 (5)	129 (3)
N—H(102)^{i}···O(11)^{iii}	2·929 (4)	0·81 (8)	2·23 (8)	144 (5)
···O(23)^{iv}	3·112 (4)		2·59 (8)	123 (5)
N—H(103)^{i}···O(11)^{v}	2·871 (4)	0·92 (5)	2·01 (5)	155 (3)
···O(22)^{v}	3·132 (4)		2·57 (5)	120 (3)
O(1)—H(1)^{i}···O(21)^{iv}	2·720 (3)	0·82 (4)	1·90 (4)	174 (3)
O(22)—H(221)^{i}···O(11)^{vi}	2·632 (3)	0·83 (5)	1·81 (5)	175 (3)
O(12)—H(12)^{i}···O(21)^{iv}	2·602 (3)	0·75 (6)	1·86 (6)	170 (4)
O(13)—H(13)^{i}···O(23)^{vii}	2·511 (3)	0·78 (6)	1·73 (6)	173 (4)

Symmetry code: (i) x, y, z; (ii) $1-x, \frac{1}{2}+y, \frac{1}{2}-z$; (iii) $-x, \frac{1}{2}+y, \frac{1}{2}-z$; (iv) $x, \frac{1}{2}-y, \frac{1}{2}+z$; (v) $1+x, \frac{1}{2}-y, \frac{1}{2}+z$; (vi) $-x, -y, -z$; (vii) $-1+x, y, z$.

The torsion angles for O(12)—P(1)—C(1)—P(2) and P(1)—C(1)—P(2)—O(23) are −177·7 (1) and −148·9 (1)° respectively. Overall the molecular geometry of the diphosphonate moiety compares well with those found in analogous compounds (Nardelli, Pelizzi, Staibano & Zucchi, 1983, and references therein).

The alkylamino side chain adopts a *trans–trans–gauche*$^-$*–gauche*$^-$ backbone conformation, with the torsion angles around C(2)—C(3), C(3)—C(4), C(4)—C(5) and C(5)—C(6) bonds being −171·3 (2), −179·5 (3), −63·0 (4) and −73·5 (4)° respectively. The O(1) hydroxyl group is (+)synclinal with respect to the C(3) methylene group, the torsional angle O(1)—C(1)—C(2)—C(3) being 52·3 (3)°.

The crystal structure is stabilized by seven hydrogen bonds involving the N amino, the O(1) hydroxyl and both phosphonyl groups (Fig. 2), three (bifurcated) of type N—H···O and four of type O—H···O. Details of these are given in Table 3.

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Structure of 1-(4-Chlorophenacyl)-2-methyl-5-morpholino-4-nitroimidazole

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Abstract. $C_{16}H_{17}ClN_4O_4$, $M_r = 364·5$, monoclinic, $P2_1/n$, $a = 14·333$ (3), $b = 11·398$ (2), $c = 10·663$ (2) Å, $\beta = 102·90$ (2)°, $V = 1698·1$ (6) Å^3, $Z = 4$, $D_m = 1·40$, $D_x = 1·43$ g cm^{-3}, λ(Cu $K\alpha$) = 1·54178 Å, $\mu = 21·47$ cm^{-1}, $F(000) = 760$, room temperature, $R = 0·049$ for 1883 unique observed reflections. The molecule is highly overcrowded, the imidazole ring is planar. The presence of the NO_2 group in position 4 of the imidazole makes the protonation of N(3) impossible. The lone pair of the morpholine N atom takes part, to some extent, in the p–π overlap.

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Introduction. *N*-Alkylnitroimidazoles are chemotherapeutically important as antiprotozoal and antibacterial agents (Hoffer & Rachlin, 1972), *e.g.* metronidazole, 1-(2-hydroxyethyl)-2-methyl-5-nitroimidazole, is both an amoebicide and a trichomonacide (Powell, McLeod & Wilmot, 1966), while azomycine, 2-nitroimidazole, exhibits antibiotic properties (Maeda, Osato & Umezawa, 1953). Moreover, metronidazole has been reported to sensitize tumour hypoxic cells both *in vitro* and *in vivo* (Chapman, Reuvers & Borsa, 1973; Asquith, Foster & Willson, 1973). 1-(2-Hydroxy-3-methoxypropyl)-2-nitroimidazole (misonidazole) is the most effective radiosensitizing agent, but its toxicity is

very high. From a toxicological point of view 4-nitroimidazole derivatives are promising because of their lower neurotoxicity in comparison with 2- and 5-nitro derivatives. Surprisingly, 4-nitro-5-substituted imidazole derivatives were found to be effective as radiosensitizers (Adams, Ahmed, Fieldon, O'Neill & Stratford, 1980); their activity is similar to that of metronidazole and higher than those of 4-nitro-5-unsubstituted derivatives.

There are some criteria which should be fulfilled in searching for new radiosensitizers, *e.g.* water solubility, lipophilicity and the presence of electron-affinitive groups. These criteria have been taken into consideration in our chemical synthesis.

Unfortunately, nitroimidazole derivatives do not possess good water solubility, so we decided to synthesize some 4-nitro-5-aminoimidazole derivatives which, in principle, should possibly give hydrochlorides or other salts. In this work we present the X-ray structure of 1-(4-chlorophenacyl)-2-methyl-5-morpholino-4-nitroimidazole. The attempts to obtain a salt of this compound failed and we expect to discover the reasons for this failure from the structural data. On the other hand, the knowledge of the structure can help to understand the biological activity of nitroimidazoles.

Experimental. Crystals grown from ethanol solution by slow evaporation. Space group and approximate cell dimensions determined from Weissenberg and oscillation photographs; density measured by flotation in aqueous KI solution; crystal size 0·2 × 0·2 × 0·4 mm; Syntex $P2_1$ diffractometer, graphite-monochromated Cu $K\alpha$ radiation, θ–2θ scan mode, background and intensity of reflections calculated by peak-profile analysis (Lehmann & Larsen, 1974; Jaskólski, 1982); accurate cell parameters refined from setting angles of 15 reflections with $10 \le 2\theta \le 25°$, max. $(\sin\theta)/\lambda = 0{\cdot}5878$ Å^{-1}, $0 \le h \le 16$, $0 \le k \le 14$, $-13 \le l \le 13$, two reference reflections monitored every 100 reflections showed no significant variation in intensity during data collection, 2637 reflections measured, 2249 unique reflections, 361 unobserved reflections [$I \le 2\sigma(I)$]; no absorption correction. Structure solved by direct methods using *SHELX*76 (Sheldrick, 1976). Best set of phases allowed location of 24 of 25 non-H atoms, methyl C atom found in the difference Fourier map. Structure refined by full-matrix least squares with *SHELX*76, function minimized $\sum w(|F_o| - |F_c|)^2$, $w^{-1} = \sigma^2(F_o) + 0{\cdot}0001F_o^2$, $\sigma(F_o)$ based on counting statistics. Non-H atoms refined anisotropically, all H atoms located from difference synthesis, at the early stage refined isotropically, then kept nonrefined with fixed isotropic displacement parameters. Empirical isotropic extinction parameter x used to correct F_c according to $F_c' = F_c(1-xF_c^2/\sin\theta)$, x converged at 0·013 (2). Five reflections: 301, 202, 002, $\bar{1}22$ and $\bar{2}32$ removed. Final $R = 0{\cdot}049$, $wR = 0{\cdot}062$, $S = 3{\cdot}51$, $(\Delta/\sigma)_{max} = 0{\cdot}013$ in final cycle; largest peak in final ΔF map 0·30 e Å^{-3}, largest hole −0·23 e Å^{-3}; atomic scattering factors those incorporated in *SHELX*76. Other computer programs used: *ORTEP* (Johnson, 1976).

Discussion. Final positional parameters and U_{eq} for non-H atoms are given in Table 1.* The molecule is shown in Fig. 1. The bond lengths and bond angles are listed in Table 2.

* Lists of structure factors, anisotropic thermal parameters, H-atom coordinates, bond lengths and angles involving H atoms, torsion angles, least-squares-planes data and short intramolecular contacts have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 51435 (20 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Final fractional coordinates and equivalent isotropic thermal parameters* (Å^2)

$U_{eq} = (U_{11}U_{22}U_{33})^{1/3}$.

	x	y	z	U_{eq}
N(1)	0·1883 (2)	0·3713 (2)	0·0024 (2)	0·0384 (9)
C(2)	0·1721 (2)	0·4712 (3)	−0·0756 (3)	0·039 (1)
N(3)	0·0814 (2)	0·4822 (2)	−0·1263 (2)	0·0434 (9)
C(4)	0·0380 (2)	0·3881 (3)	−0·0834 (3)	0·037 (1)
C(5)	0·1015 (2)	0·3164 (3)	−0·0018 (3)	0·036 (1)
C(6)	0·2821 (2)	0·3272 (3)	0·0676 (3)	0·039 (1)
C(7)	0·3261 (2)	0·2550 (3)	−0·0245 (3)	0·040 (1)
O(7)	0·2752 (2)	0·2220 (2)	−0·1261 (2)	0·0569 (9)
C(8)	0·4286 (2)	0·2243 (2)	0·0089 (3)	0·036 (1)
C(9)	0·4919 (2)	0·2672 (3)	0·1180 (3)	0·041 (1)
C(10)	0·5875 (2)	0·2347 (3)	0·1443 (3)	0·042 (1)
C(11)	0·6188 (2)	0·1593 (3)	0·0614 (3)	0·046 (1)
C(12)	0·5581 (2)	0·1157 (3)	−0·0485 (3)	0·049 (1)
C(13)	0·4628 (2)	0·1483 (3)	−0·0733 (3)	0·045 (1)
C(14)	0·2506 (3)	0·5487 (3)	−0·0922 (4)	0·061 (1)
N(15)	−0·0625 (2)	0·3702 (3)	−0·1310 (2)	0·0442 (9)
O(151)	−0·1092 (2)	0·4480 (2)	−0·1962 (2)	0·063 (1)
O(152)	−0·0974 (2)	0·2762 (2)	−0·1080 (2)	0·060 (1)
N(16)	0·0976 (2)	0·2203 (2)	0·0765 (2)	0·0394 (8)
C(17)	0·0230 (3)	0·2183 (3)	0·1505 (3)	0·061 (1)
C(18)	0·0571 (3)	0·1358 (3)	0·2630 (4)	0·067 (2)
O(19)	0·0747 (2)	0·0212 (2)	0·2223 (2)	0·0596 (9)
C(20)	0·1444 (2)	0·0238 (3)	0·1452 (4)	0·056 (1)
C(21)	0·1152 (2)	0·1040 (3)	0·0314 (3)	0·049 (1)
Cl	0·73840 (6)	0·1170 (1)	0·0937 (1)	0·0713 (4)

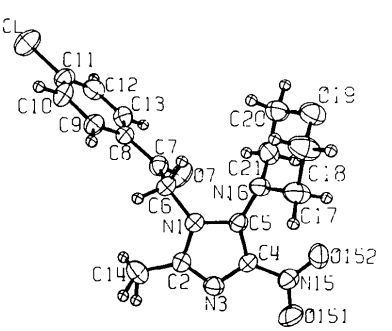


Fig. 1. *ORTEP* drawing of the molecule showing the conformation and the numbering scheme used.

Table 2. *Bond lengths* (Å) *and bond angles* (°) *with e.s.d.'s in parentheses*

N(1)–C(2)	1·398 (4)	C(21)–N(16)	1·451 (4)
C(2)–N(3)	1·299 (4)	N(1)–C(6)	1·457 (4)
N(3)–C(4)	1·367 (4)	C(6)–C(7)	1·520 (4)
C(4)–C(5)	1·376 (4)	C(7)–O(7)	1·222 (4)
C(5)–N(1)	1·384 (4)	C(7)–C(8)	1·475 (4)
C(4)–N(15)	1·432 (4)	C(8)–C(9)	1·392 (4)
N(15)–O(151)	1·228 (4)	C(9)–C(10)	1·387 (4)
N(15)–O(152)	1·230 (4)	C(10)–C(11)	1·376 (5)
C(5)–N(16)	1·385 (4)	C(11)–C(12)	1·386 (5)
N(16)–C(17)	1·462 (5)	C(12)–C(13)	1·385 (5)
C(17)–C(18)	1·513 (5)	C(13)–C(8)	1·395 (4)
C(18)–O(19)	1·416 (4)	C(11)–Cl	1·740 (3)
O(19)–C(20)	1·428 (5)	C(2)–C(14)	1·472 (5)
C(20)–C(21)	1·502 (5)		

N(1)–C(2)–N(3)	110·0 (2)	C(17)–N(16)–C(21)	111·4 (2)
C(2)–N(3)–C(4)	106·0 (2)	N(16)–C(17)–C(18)	106·8 (3)
N(3)–C(4)–C(5)	112·9 (2)	C(17)–C(18)–O(19)	112·3 (3)
C(4)–C(5)–N(1)	102·5 (2)	C(18)–O(19)–C(20)	110·5 (3)
C(5)–N(1)–C(2)	108·7 (2)	O(19)–C(20)–C(21)	111·7 (3)
C(4)–N(15)–O(151)	118·4 (2)	C(20)–C(21)–N(16)	109·2 (3)
C(4)–N(15)–O(152)	118·5 (2)	C(5)–N(1)–C(6)	126·0 (2)
O(151)–N(15)–O(152)	123·1 (3)	C(2)–N(1)–C(6)	125·1 (1)
N(15)–C(4)–C(5)	127·6 (2)	N(1)–C(6)–C(7)	110·7 (2)
N(15)–C(4)–N(3)	119·4 (2)	C(6)–C(7)–O(7)	118·9 (3)
C(4)–C(5)–N(16)	137·6 (2)	C(6)–C(7)–C(8)	120·6 (2)
N(1)–C(5)–N(16)	119·6 (2)	O(7)–C(7)–C(8)	120·5 (3)
C(5)–N(16)–C(17)	117·9 (2)	C(7)–C(8)–C(9)	123·2 (2)
C(5)–N(16)–C(21)	119·4 (2)	C(12)–C(13)–C(8)	121·2 (3)
C(7)–C(8)–C(13)	117·8 (2)	C(13)–C(8)–C(9)	119·0 (3)
C(8)–C(9)–C(10)	120·4 (3)	C(10)–C(11)–Cl	119·6 (2)
C(9)–C(10)–C(11)	119·1 (3)	C(12)–C(11)–Cl	118·3 (2)
C(10)–C(11)–C(12)	122·1 (3)	N(1)–C(2)–C(14)	122·0 (2)
C(11)–C(12)–C(13)	118·2 (3)	N(3)–C(2)–C(14)	128·0 (3)

The imidazole ring is planar with no atomic deviation greater than 0·006 (3) Å from its least-squares plane. The molecule as a whole is highly overcrowded, so the atoms in the nearest neighbourhood of the imidazole are displaced from its plane, N(15) by +0·102 (3), N(16) −0·123 (3), C(6) +0·098 (3) Å; C(14) has the smallest displacement of +0·017 (4) Å.

The C(4)–NO_2 group is planar and makes a dihedral angle with the imidazole plane of 9·7 (1)°. The C(6),-C(7),O(7),C(8) fragment of the chlorophenacyl substituent is flat and perpendicular to the imidazole ring [dihedral angle 90·4 (2)°]; however, the N(1) atom is displaced by 0·340 (2) Å from this fragment's plane. The phenyl ring is twisted by 5·8 (1)° from the C(6),C(7),O(7),C(8) plane and makes a dihedral angle of 84·7 (2)° with the imidazole ring. The orientation of morpholine substituent is described by the torsion angles C(4)–C(5)–N(16)–C(17) −39·8 (4), N(1)–C(5)–N(16)–C(21) −86·9 (3), C(4)–C(5)–N(16)–C(21) +100·9 (3), N(1)–C(5)–N(16)–C(17) +132·4 (3)°. The morpholine ring adopts a chair conformation. The methyl group deviates only slightly from the imidazole plane.

The bond lengths in the heterocyclic five-membered ring differ from those found in the X-ray study of imidazole at 123 K (Martinez-Carrera, 1966) and in the neutron study at 123 and 293 K (Craven, McMullan, Bell & Freeman, 1977) and at 103 K (McMullan, Epstein, Ruble & Craven, 1979); the corresponding bond angles also differ significantly. A common feature of the imidazole ring is that the C(2)–N(3) bond is the shortest in most cases, indicating a more or less localized double bond between these two atoms. The same has been observed in this structure; moreover, the C(2)–N(3) bond, 1·299 (4) Å, has been shortened in comparison with that in imidazole itself probably due to hyperconjugation with the methyl group. Also, the N(3)–C(4) bond, 1·367 (4) Å, has been shortened in comparison with that in imidazole. The C–N bond of the nitro group, 1·432 (4) Å, is shorter than the C–NO_2 bond in dinitrobenzene, 1·478 (2) Å (Di Rienzo, Domenicano & Riva di Sanseverino, 1980). The shortening of these three bonds is possibly due to some delocalization in which the lone pair of N(3) takes part; this can explain the inactivity of N(3) toward protonation. This observation correlates with the lowering of the basicity of nitroimidazoles, which is especially dramatic in the case of 4-nitroimidazoles, *e.g.* the pK_a of imidazole is 7·31, that of 4-amino-1,2-dimethyl-5-nitroimidazole 2·50 and that of 5-amino-1,2-dimethyl-4-nitroimidazole 0·33 (Schofield, Grimmett & Keene, 1976).

Formulation of the nitro group as $-C{=}NO_2^-$ is supported by the N(16)–C(5) and C(5)–C(4) bond lengths. The N(16)–C(5) distance, 1·385 (4) Å, is the same as in the aniline molecule (Fukuyo, Hirotsu & Higuchi, 1982). The sum of the bond angles around the N(16) atom is 348·7° which is an intermediate value between that for sp^2(360°)- and that for sp^3(328·4°)-

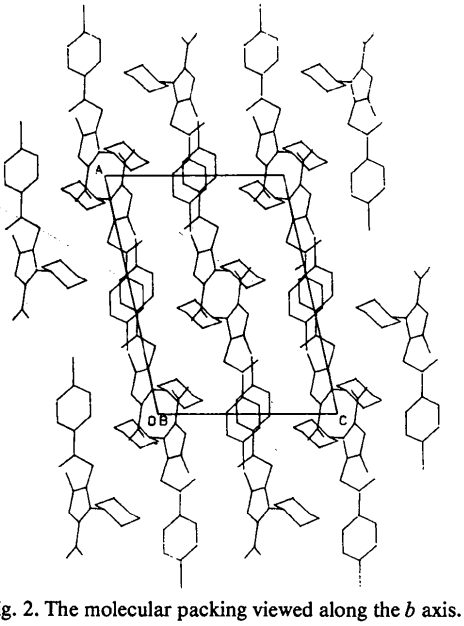
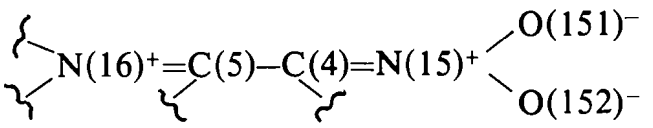


Fig. 2. The molecular packing viewed along the *b* axis.

hybridized N atoms. Judging by these parameters, at least some p–π overlap between N(16) and the imidazole ring exists. The C(5)–C(4) bond length, 1·376 (4) Å, is by 5σ longer than that in imidazole (Craven *et al.*, 1977). According to these bond distances, the inactivity of N(16) toward protonation can result from the structure of the resonance hybrid:



This is further confirmed by the planarity of this fragment.

In spite of the fact that molecular overcrowding has been lowered by noncoplanarity of the substituents, several short intramolecular contacts have been retained. In particular, very short contacts between N(3) and the nitro group and the methyl group exist: O(151)···N(3) 2·696 (4), N(15)···N(3) 2·417 (4), N(3)···C(14) 2·491 (5) Å. On the basis of the structure determination, a conclusion can be drawn: the protonation of 1-(4-chlorophenacyl)-2-methyl-5-morpholino-4-nitroimidazole is impossible for two reasons:

(1) electronic effects,

(2) steric effects due to molecular overcrowding.

The intermolecular contact distances almost coincide with the sum of the van der Waals radii of the corresponding atoms. The packing of molecules is shown in Fig. 2.

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N-Salicylidène Triméthyl-2,4,6 Aniline

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(*Reçu le 5 mai* 1988, *accepté le 22 août* 1988)

Abstract. $C_{16}H_{17}NO$, $M_r = 239{\cdot}3$, monoclinic, $P2_1$, $a = 7{\cdot}729$ (3), $b = 7{\cdot}306$ (2), $c = 12{\cdot}602$ (3) Å, $\beta = 93{\cdot}89$ (8)°, $V = 709{\cdot}9$ (4) Å^3, $Z = 2$, $D_x = 1{\cdot}12$ Mg m^{-3}, λ(Cu $K\alpha$) = 1·5418 Å, $\mu = 0{\cdot}552$ mm^{-1}, $F(000) = 256$, $T = 298$ K, final $R = 0{\cdot}063$ for 917 unique reflections. The two aromatic rings are perpendicular which accounts for the relative weakness of the O—H···N hydrogen bond as previously reported from the γ(OH) vibrational frequency.

Introduction. Les *N*-salicylidène amines C_6H_4(OH-2)–CH=N–*R* ont, à l'état cristallisé, des propriétés

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