^{35}Cl NQR and Structural Studies on Substituted Amides, $X_yC_6H_{5-y}$NHCOR (X = H or Cl; y = 0, 1 or 2 and R = $C(CH_3)_3$, $CHClCH_3$, C_6H_5 or 2-ClC_6H_4)

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^{35}Cl NQR frequencies of some N-(substitutedphenyl)-amides represented by the general formula, $X_yC_6H_{5-y}$NHCOR (where X = H or Cl; $y = 0$, 1 or 2 and R = H, CH_3, CH_2CH_3, $CH(CH_3)_2$, $C(CH_3)_3$, CH_2Cl, $CHCl_2$ or CCl_3) have been measured and compared with those of other compounds in the family to analyse the effect of substitution in the side chain on the frequencies. Comparison of ^{35}Cl NQR frequencies of all the N-(2-chlorophenyl)- and N-(2,6-dichlorophenyl)-amides reveals that the presence of alkyl groups in the side chain lowers the frequency, while that of aryl or chloro-substituted alkyl groups enhance the frequencies to some extent, when compared to the frequencies of either N-(2-chlorophenyl)-acetamide or N-(2,6-dichlorophenyl)-acetamide. In addition, the crystal structures of N-(phenyl)-2-chloro-2-methylacetamide ($C_6H_5NHCOCHClCH_3$) and N-(phenyl)-2-chloro-benzamide (C_6H_5NHCO-(2-ClC_6H_4)) have been determined and the data analysed along with the crystal structures of related compounds. The data (lattice constants in Å) of the new structures are: C_6H_5NHCO- $CHClCH_3$: monoclinic, $P2_1/c$, $Z = 4$, $a = 10.879(2)$, $b = 9.561(2)$, $c = 10.067(2)$, $\beta = 116.080(10)°$; C_6H_5NHCO-(2-ClC_6H_4): tetragonal, P4(3), $Z = 4$, $a = 8.795(4)$, $b = 8.795(4)$, $c = 15.115(6)$, $\beta = 90.0°$. It is evident from a comparison, that the side chain substitution influences the C(S)-C(O) bond length, while the effect on the other bond lengths is not significant except for benzanilide. Similarly, only the side chain angles are affected to some extent. The variations do not show definite trends, probably due to the differences in the crystallisations.

Key words: Substituent Effect; ^{35}Cl NQR and Crystal Structures; Substituted Amides.

1. Introduction

The combined tool of nuclear quadrupole resonance (NQR) spectroscopy and crystal structure studies has been extensively used to investigate the structural aspects of a variety of compounds [1 – 3]. Amides are of fundamental chemical interest as conjugation between the nitrogen lone pair electrons and the carbonyl π-bond results in distinct physical and chemical properties [4]. The amide moiety is an important constituent of many biologically significant compounds [4, 5].

The ^{35}Cl NQR and infrared spectra of a number of N-(chlorophenyl)-chloroacetamides have been studied [6 – 10]. We have recently prepared several substituted amides of the configuration: $X_yC_6H_{5-y}$- NHCO-$CH_{3-y}Cl_y$ (where X = CH_3, NO_2 or Br and $y = 1$, 2 or 3) and measured their ^{35}Cl NQR and IR spectra [9 – 10]. The crystal structures of some of the N-phenyl acetamides have also been determined [11 – 19]. As part of the work aimed at correlating ^{35}Cl NQR frequencies of the substituted N-phenylamides represented by the general formula, $X_yC_6H_{5-y}$NHCOR (where X = H or Cl; $y = 0$, 1 or 2 and R = H, CH_3, CH_2CH_3, $CH(CH_3)_2$, $C(CH_3)_3$, CH_2Cl, $CHCl_2$ or CCl_3) with their chemical bond parameters, we report here the ^{35}Cl NQR spectra of N-(substitutedphenyl)-amides, namely, N-(phenyl)-2-chlorobenzamide, N-(2-chlorophenyl)-benzamide, N-(2-chlorophenyl)-2-chlorobenzamide, N-(2-chlorophenyl)-2,2,2-trimethylacetamide and N-(2,6-dichlorophenyl)-benzamide. ^{35}Cl NQR frequencies of all the substituted N-(2-chlorophenyl) and N-(2,6-dichlorophenyl)-amides have also been analysed.

Further, to see how the -NHCO- bond parameters vary with substitution in the side chain, the crystal structures of N-(phenyl)-2-chloro-2-methylacetamide

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Table 1. Effect of substitution on the ^{35}Cl NQR ring C-Cl frequencies in arylamides (Temp. 77 K, Assignment o).

Compounds	γ (MHz)	Ref.	S/N
$C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$	35.347	(present)	4 to 5
$2\text{-}ClC_6H_4NHCOC_6H_5$	35.465	(present)	7 to 8
$2\text{-}ClC_6H_4NHCO\text{-}(2\text{-}ClC_6H_4)$	35.771	(present)	2 to 3
	35.412	(present)	2 to 3
$2\text{-}ClC_6H_4NHCOH$	34.860	[11]	–
$2\text{-}ClC_6H_4NHCOCH_3$	35.153	[6]	5
$2\text{-}ClC_6H_4NHCOC(CH_3)_3$	34.829	(present)	3
$2\text{-}ClC_6H_4NHCOCH_2Cl$	35.351	[6]	4
$2\text{-}ClC_6H_4NHCOCHCl_2$	35.425	[6]	1 to 2
$2\text{-}ClC_6H_4NHCOCCl_3$	34.943	[6]	3
$2,6\text{-}Cl_2C_6H_3NHCOH$	35.992	[18]	30
$2,6\text{-}Cl_2C_6H_3NHCOCH_3$	35.943	[18]	20
$2,6\text{-}Cl_2C_6H_3NHCOCH_2CH_3$	35.745	[18]	25
$2,6\text{-}Cl_2C_6H_3NHCOCH(CH_3)_2$	35.775	[18]	30
$2,6\text{-}Cl_2C_6H_3NHCOC(CH_3)_3$	35.739	[18]	20
$2,6\text{-}Cl_2C_6H_3NHCOC_6H_5$	36.412	(present)	1 to 2
$2,6\text{-}Cl_2C_6H_3NHCOCH_2Cl$	35.967	[6]	30
$2,6\text{-}Cl_2C_6H_3NHCOCHCl_2$	36.090	[18]	20
$2,6\text{-}Cl_2C_6H_3NHCOCCl_3$	36.324	[18]	30

All N-(2,6-dichlorophenyl)-amides except N-(2,6-dichlorophenyl)-benzamide showed two ^{35}Cl NQR ring C-Cl frequencies for the two chlorine atoms present in them. Hence the average of two frequencies are given.

($C_6H_5NHCO\text{-}CHClCH_3$) and N-(phenyl)-2-chlorobenzamide ($C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$) have been determined and the data analysed along with crystal structures of N-(phenyl)-acetamide ($C_6H_5NHCO\text{-}CH_3$), N-(phenyl)-2,2-dichloroacetamide ($C_6H_5\text{-}NHCOCHCl_2$), N-(phenyl)-2,2,2-trichloroacetamide ($C_6H_5NHCOCCl_3$), N-(phenyl)-2,2,2-trimethylacetamide ($C_6H_5NHCO\text{-}C(CH_3)_3$) and N-(phenyl)-benzamide ($C_6H_5NHCOC_6H_5$).

2. Experimental Section

2.1. Preparation and Characterization of the Compounds

The N-(substitutedphenyl)-amides were prepared from substituted anilines, substituted benzoic acids or pivalic acid (Aldrich, Germany) and thionyl chloride. N-(Phenyl)-2-chloro-2-methylacetamide and N-(phenyl)-2-chlorobenzamide were prepared from aniline, 2-chloro-2-methylacetic acid or 2-chlorobenzoic acid (Aldrich, Germany) and thionyl chloride with constant stirring [6, 7, 20]. The resulting mixtures were slowly warmed to expel HCl. Excess thionyl chloride was hydrolysed by adding cold water dropwise under ice cold conditions. HCl produced was removed by treating with excess of 2M NaOH. The solids separated were filtered under suction, washed thoroughly with water and dried. The substitutedphenyl acetamides thus prepared were recrystallised from ethanol several times. Purity of the compounds was checked by elemental analysis and by determining their melting points. The compounds have been further characterised by recording their infrared spectra on a JASCO-430 (Japan) FT/IR spectrometer. The resolution of the spectrometer was set to 4 cm^{-1} and the scanning range was 400 to 4000 cm^{-1}. The spectra were measured in the solid state as pressed KBr pellets (13 mm).

Table 2. Experimental conditions for the crystal structure determination and crystallographic data of N-(phenyl)-2-chloro-2-methylacetamide ($C_6H_5NHCO\text{-}CHClCH_3$) and N-(phenyl)-2-chlorobenzamide ($C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$). Diffractometer: Stoe-Stadi 4; Monochromator: Graphite (002); scan 2 θ/ω = 1/1; Refinement method: Full- matrix least-squares on F^2.

Description	C_6H_5NHCO $CHClCH_3$-	C_6H_5NHCO $(2\text{-}ClC_6H_4)$
Chemical formula	$C_9H_{10}ClNO$	$C_{13}H_{10}ClNO$
Formula weight	183.63	231.67
Temperature, K	299(2)	296(2)
Wavelength, pm	71.069	71.069
Crystal system	Monoclinic	Tetragonal
Space group	$P2_1/c$	P4(3)
a, Å	10.879(2)	8.795(4)
b, Å	9.561(2)	8.795(4)
c, Å	10.067(2)	15.115(6)
β, deg.	116.080(10)	90
Volume, Å^{33}	940.5(3)	1169.2(9)
Z	4	4
D_{calcd}, g cm^{-3}	1.297	1.316
Abs. coeff., cm^{-1}	3.57	3.03
$F(000)$	384	480
Crystal size, mm^3	$2.40 \times 0.46 \times 0.21$	$1.00 \times 0.32 \times 0.28$
θ range, deg.	2.08 to 29.99	2.32 to 27.46
Index ranges	$-14 \le h \le 15$ $0 \le k \le 13$ $-14 \le l \le 2$	$-11 \le h \le 2$ $0 \le k \le 11$ $-19 \le l \le 19$
Reflections coll.	3251	3635
Independent refl.	2745	2668
$[R(\text{int})]$	0.0066	0.0150
Absorption corr.	–	Numerical
Max. and min. transm.	0.9288 and 0.4810	0.924 and 0.900
Data	2745	2668
Restraints/parameters	0 / 123	1 / 150
Goodness-of-fit on F^2	1.045	1.048
Final $R[I > 2\sigma(I)]$	$R1 = 0.0414$ $wR2 = 0.1127$	$R1 = 0.0475$ $wR2 = 0.1135$
R indices (all data)	$R1 = 0.0530$ $wR2 = 0.1242$	$R1 = 0.0683$ $wR2 = 0.1338$
Abs. str. parameter	–	0.27(14)
Extinction coeff.	0.015(3)	0.024(4)
Largest diff. peak and hole $e^-\text{Å}^3$	0.284 and -0.235	0.177 and -0.180

Table 3. Atomic coordinates ($\cdot 10^4$) and equivalent isotropic displacement parameters ($Å^2 \cdot 10^3$) of $C_6H_5NHCO\text{-}CHClCH_3$ and $C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$. U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

Atom	*x*	*y*	*z*	*U*(eq)
$C_6H_5NHCOCHClCH_3$:				
Cl(1)	−1758(1)	1103(1)	3865(1)	76(1)
C(2)	−2965(5)	871(6)	850(6)	108(3)
Cl(1’)	−3016(15)	945(13)	716(17)	89(3)
C(2’)	−1960(40)	770(40)	3340(50)	115(14)
C(3)	−1617(1)	508(1)	2257(2)	55(1)
C(4)	−405(1)	1264(1)	2211(2)	46(1)
O(5)	−356(1)	2537(1)	2193(2)	65(1)
N(6)	559(1)	421(1)	2161(1)	48(1)
C(7)	1785(1)	893(1)	2130(2)	44(1)
C(8)	1780(2)	1986(2)	1233(2)	59(1)
C(9)	3001(2)	2431(2)	1258(2)	75(1)
C(10)	4216(2)	1795(2)	2157(2)	75(1)
C(11)	4211(2)	680(2)	3009(2)	72(1)
C(12)	2998(2)	222(2)	3004(2)	60(1)
$C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$:				
Cl(1)	1006(2)	7404(3)	1335(1)	129(1)
C(2)	2955(6)	7344(5)	1187(3)	80(1)
C(3)	3589(9)	8360(6)	590(4)	103(2)
C(4)	5138(9)	8263(7)	450(4)	109(2)
C(5)	6021(7)	7211(9)	851(4)	112(2)
C(6)	5365(6)	6221(7)	1447(3)	91(1)
C(7)	3816(6)	6282(5)	1628(2)	67(1)
C(8)	3177(5)	5318(4)	2364(2)	63(1)
O(9)	2627(4)	5919(3)	3025(2)	75(1)
N(10)	3292(4)	3814(4)	2237(2)	64(1)
C(11)	2799(4)	2637(4)	2811(2)	60(1)
C(12)	3381(6)	1188(5)	2661(3)	78(1)
C(13)	2849(7)	−21(5)	3157(4)	94(2)
C(14)	1766(7)	201(6)	3797(3)	89(1)
C(15)	1211(6)	1608(7)	3957(3)	92(2)
C(16)	1717(5)	2866(5)	3468(3)	71(1)

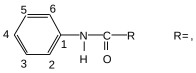
Table 4. Bond lengths (Å) (standard deviation) of $C_6H_5\text{-}NHCOCHClCH_3$ and $C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$.

$C_6H_5NHCOCHClCH_3$:			
Cl(1)-C(3)	1.785(16)	C(2)-C(3)	1.565(6)
Cl(1’)-C(3)	1.680(14)	C(2’)-C(3)	1.320(5)
C(3)-C(4)	1.522(18)	C(4)-O(5)	1.219(15)
C(4)-N(6)	1.341(16)	N(6)-C(7)	1.421(16)
$C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$:			
C(8)-O(9)	1.230(5)	Cl(1)-C(2)	1.730(6)
C(2)-C(7)	1.375(6)	C(2)-C(3)	1.386(7)
C(3)-C(4)	1.382(9)	C(4)-C(5)	1.352(9)
C(5)-C(6)	1.379(8)	C(6)-C(7)	1.390(7)
C(7)-C(8)	1.508(5)	C(8)-N(10)	1.341(5)
N(10)-C(11)	1.419(5)		

Table 5. Bond lengths (Å) (standard deviation) of $C_6H_5\text{-}NHCOCHClCH_3$ and $C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$.

$C_6H_5NHCOCHClCH_3$:			
C(2’)-C(3)-C(4)	119.9(18)	C(2’)-C(3)-C(2)	102.3(17)
C(4)-C(3)-C(2)	110.3(2)	C(2’)-C(3)-Cl(1’)	104.2(18)
C(4)-C(3)-Cl(1’)	107.4(5)	C(2)-C(3)-Cl(1’)	2.9(7)
C(2’)-C(3)-Cl(1)	12.6(17)	C(4)-C(3)-Cl(1)	107.3(9)
C(2)-C(3)-Cl(1)	109.3(3)	Cl(1’)-C(3)-Cl(1)	110.8(6)
O(5)-C(4)-N(6)	124.0(12)	O(5)-C(4)-C(3)	121.2(12)
N(6)-C(4)-C(3)	114.7(11)	C(4)-N(6)-C(7)	124.5(11)
$C_6H_5NHCO\text{-}(2\text{-}ClC_6H_4)$:			
C(7)-C(2)-C(3)	122.1(5)	C(7)-C(2)-Cl(1)	120.2(3)
C(3)-C(2)-Cl(1)	117.6(4)	C(4)-C(3)-C(2)	117.2(6)
C(5)-C(4)-C(3)	122.6(5)	C(4)-C(5)-C(6)	119.0(6)
C(5)-C(6)-C(7)	120.9(6)	C(2)-C(7)-C(6)	118.1(4)
C(2)-C(7)-C(8)	122.3(4)	C(6)-C(7)-C(8)	119.3(4)
O(9)-C(8)-N(10)	124.8(4)	O(9)-C(8)-C(7)	120.3(3)
N(10)-C(8)-C(7)	114.9(3)	C(8)-N(10)-C(11)	127.6(3)

Table 6. Bond lengths (Å) of N-(phenyl)-substitutedamides.

	Bond length for  R=,						
Connections	CH_3	$CHCl_2$	CCl_3	$C(CH_3)_3$	$CHClCH_3$	C_6H_5	$2\text{-}ClC_6H_4$
C(s)-C(O)	1.476	1.528	1.564	1.532	1.522	1.484	1.508
C-O	1.226	1.226	1.211	1.219	1.219	1.282	1.230
C(O)-N	1.330	1.337	1.337	1.348	1.341	1.345	1.340
N-C(1r)	1.426	1.425	1.424	1.420	1.421	1.414	1.419
C(1r)-C(2r)	1.384	1.392	1.390	1.383	1.380	1.415	1.392
C(1r)-C(6r)	1.366	1.392	1.386	1.379	1.379	1.406	1.390
C(2r)-C(3r)	1.413	1.385	1.386	1.384	1.388	1.391	1.403
C(3r)-C(4r)	1.402	1.379	1.382	1.375	1.373	1.383	1.372
C(4r)-C(5r)	1.369	1.369	1.382	1.362	1.369	1.346	1.352
C(5r)-C(6r)	1.388	1.384	1.373	1.379	1.384	1.338	1.382
Cl(1)-C(s)	–	1.766	1.767	–	1.785	–	1.729

^{35}*Cl NQR Frequency Measurements*

Polycrystalline samples of the title compounds were employed. The ^{35}Cl NQR of the N-(phenylsubstituted)-amides were measured at 77 K. The spectra were registered by the continuous wave method with a superregenerative spectrometer. Temperature at the sample site was produced by a stream of temperature and flow regulated nitrogen gas or with a liquid nitrogen bath at 77 K. The temperatures at the sample site were measured by copper-constantan thermocouples to ±1 K. The resonance frequencies were measured via a frequency counter to an accuracy of ±5 kHz. The latter accuracy was determined by the line width of the resonances, which was between 10 and 20 kHz.

X-ray Diffraction Studies

Small crystals were selected for structure determinations, which were carried out at room temperature. The collected intensity data were corrected for Lorentz polarisation and absorption. The positional parameters were determined by direct methods and least squares refinement (SHELXL-97) [21–27]. For locating the

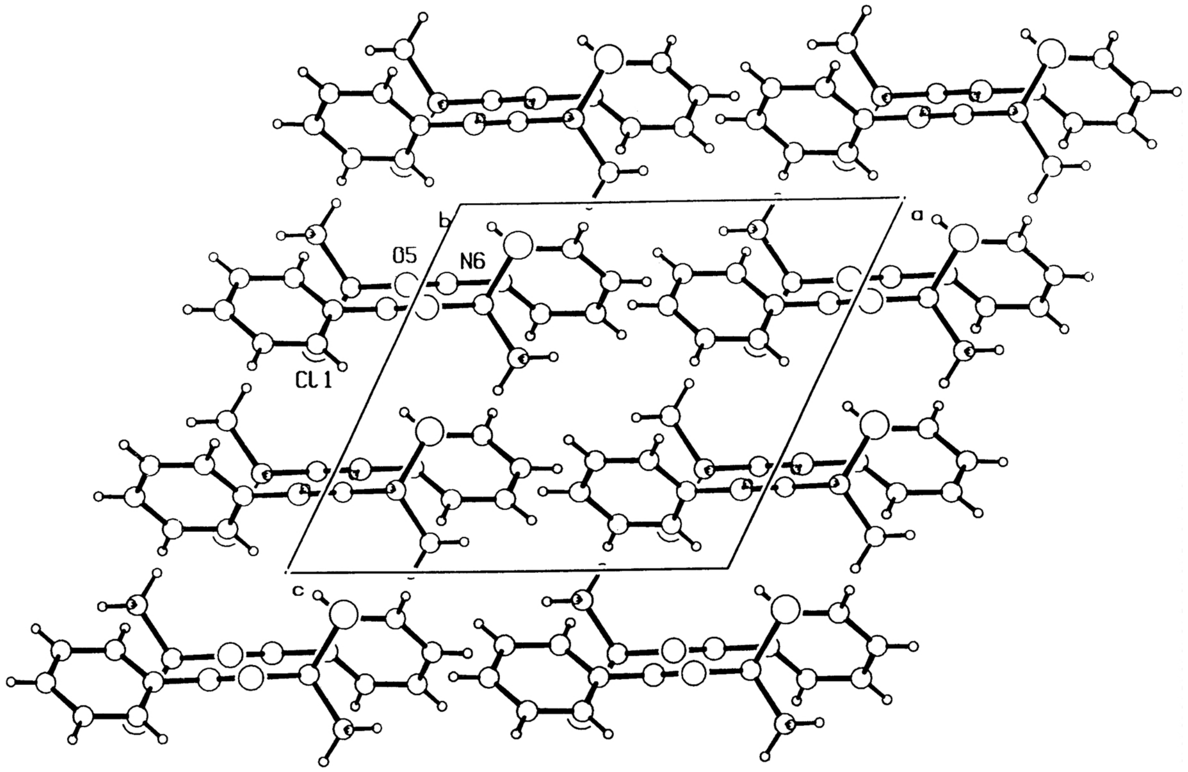
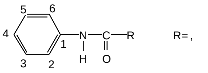


Fig. 1. Projection of the unit cell of N-(phenyl)-2-chloro-2-methylacetamide ($C_6H_5NHCOCHClCH_3$) along [010] on to the *ac* plane.

Table 7. Bond angles (°) of N-(phenyl)-substitutedamides.

	Bond angles for  R=,						
Connections	CH_3	$CHCl_2$	CCl_3	$C(CH_3)_3$	$CHClCH_3$	C_6H_5	$2\text{-}ClC_6H_4$
C(s)-C(O)-N	117.7	113.6	114.8	116.1	114.7	118.3	114.9
C(s)-C(O)-O	120.4	121.0	118.9	121.9	121.3	120.3	120.3
O-C(O)-N	121.7	125.4	126.3	122.1	124.0	121.3	124.8
C(O)-N-C(1r)	129.3	126.8	125.4	126.8	124.5	124.3	127.6
N-C(1r)-C(2r)	122.7	122.3	122.1	122.4	121.4	124.8	122.7
N-C(1r)-C(6r)	115.7	117.8	117.9	117.9	118.8	114.1	117.1
C(2r)-C(1r)-C(6r)	121.2	119.9	119.9	119.7	119.8	120.6	120.1

hydrogen atom positions, the C-H distances were fixed to 0.93 Å for the ring hydrogen atoms, while the side chain C-H distances were fixed to 0.96 Å for the CH_3 group. Further experimental conditions for structure determinations and refinements are given in Table 2.

3. Results and Discussion

The ^{35}Cl NQR frequencies from the literature [28, 29] or the present studies are listed in Table 1.

There was no problem in assigning the frequencies as there are only ^{35}Cl NQR frequencies of the ring C-Cl in all benzanilides. N-(2,6-Dichlorophenyl)-benzamide showed one C-Cl ^{35}Cl NQR frequency at 36.412 MHz for the two chlorine atoms present. The two atoms may be crystallographically equivalent. Further, ^{35}Cl NQR frequency of C-Cl in the ring attached to the amide group is slightly higher than that attached to the carbonyl oxygen.

Comparison of ^{35}Cl NQR frequencies of all the N-(2-chlorophenyl)- and N-(2,6-dichlorophenyl)-amides in Table 1 revealed that the presence of alkyl groups in the side chain lowers the frequency, while that of aryl or chlorosubstituted alkyl groups enhance the frequencies to some extent, with one exception, when compared to the frequencies of either N-(2-chlorophenyl)-acetamide or N-(2,6-dichlorophenyl)-acetamide. The exception of N-(2-chlorophenyl)-2,2,2-trichloroacetamide may be due to the fact that the chemically equivalent chlorine atoms may exhibit different NQR frequencies due to the crystal field effect.

The crystallographic data for the compounds, N-(phenyl)-2-chloro-2-methylacetamide and N-(phenyl)-2-chlorobenzamide are given in Table 2. The atomic

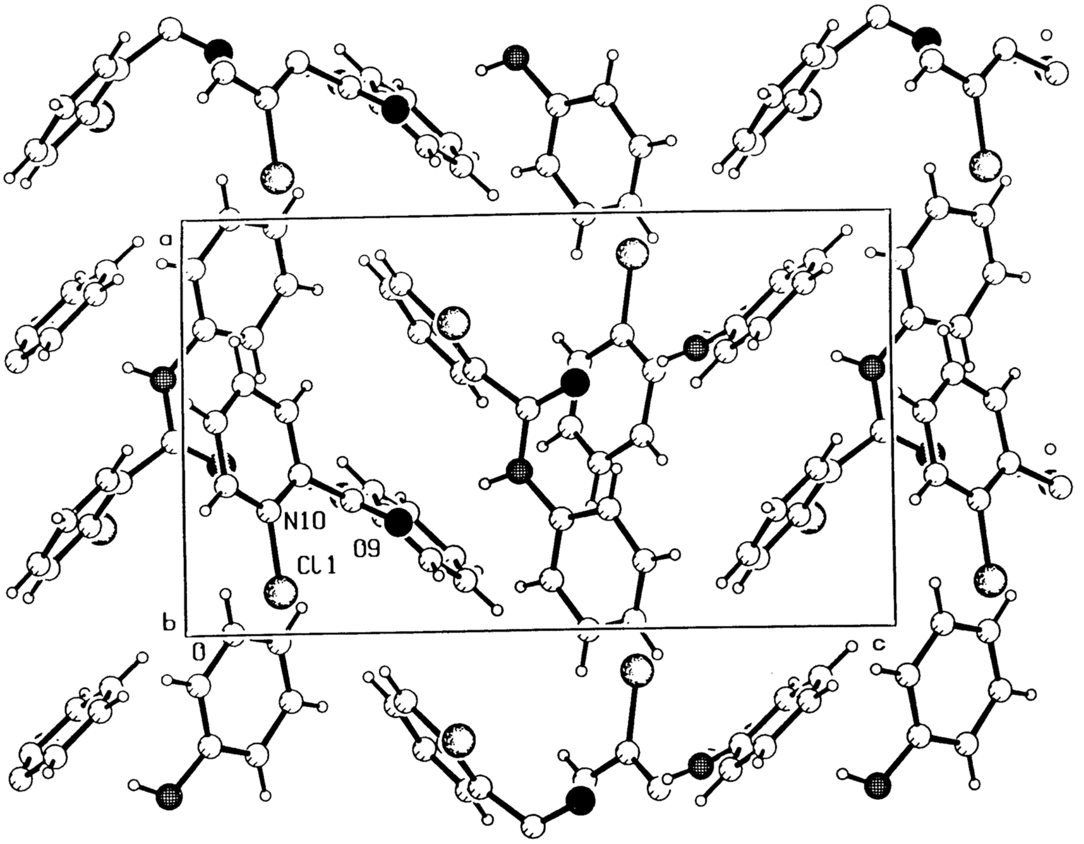


Fig. 2. Projection of the unit cell of N-(phenyl)-2-chlorobenzamide (C_6H_5NHCO-(2-ClC_6H_4)) along [010] on to the *ac* plane.

coordinates and the mean displacement parameters are listed in Table 3, while Tables 4 and 5 give the intermolecular bond distances and bond angles, respectively. The hydrogen coordinates, anisotropic displacement parameters and further informations on the crystal structure determinations have been deposited at the Cambridge Crystallographic Data Centre (CCDC). The CCDC numbers are 200386 and 200387 for N-(phenyl)-chlorobenzamide and N-(phenyl)-2-chloro-2-methyl-acetamide, respectively. The projections of the unit cells of the compounds are shown in Figures 1 and 2.

To analyse the effect of substitution in the side chain on the crystal structures, the structural data of the compounds, N-(phenyl)-2-chloro-2-methylacetamide and N-(phenyl)-2-chlorobenzamide have been compared with those of N-(phenyl)-acetamide, N-(phenyl)-2,2-dichloroacetamide, N-(phenyl)-2,2,2-trichloroacetamide, N-(phenyl)-2,2,2-trimethylacetamide and N-(phenyl)-benzamide (Tables 6 and 7). It is evident from the data that the side chain substitution influences the C(S)-C(O) bond length, while the effect on other bond lengths is not significant except for benzanilide. Similarly only the side chain angles are affected to some extent, while the other angles are not much influenced by the side chain substitution. The variations do not show a definite trend, probably due to differences in crystallisation of the compounds.

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