Abstract

Ollinger, Christian G.: A Study of Intermolecular and Intramolecular Horner Wadsworth-Emmons Olefination of Substituted 5-(Diethoxyphosphoryl)-*N*-acylpiperidin-4-ones. (Under the direction of Dr. Daniel L. Comins)

This research has focused on extending the utility of previously synthesized 5-iodo-2,3-dihydro-4-pyridones. The initial conversion involved introducing a phosphonate group by nickel-catalyzed cross-coupling with the vinyl iodide at the C-5 position. Using a Horner-Wadsworth-Emmons reaction, the phosphonate ester was then converted to various olefins in moderate to high yields. Having an aldehyde present on the substituted 5-(diethoxyphosphoryl)-*N*-acylpiperidin-4-one allowed cyclization to occur thereby providing a bicyclic enone.

A Study of Intermolecular and Intramolecular

Horner-Wadsworth-Emmons Olefination of Substituted

5-(Diethoxyphosphoryl)-*N*-acylpiperidin-4-ones

by

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Biography

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List of Abbreviations and Terms

Abbreviation or Term	Explanation
$A^{(1,3)}$	1,3-allylic
AcOH	acetic acid
anal.	analysis
anhyd	anhydrous
aq	aqueous
bp	boiling point
br	broad
brine	saturated aqueous sodium chloride
Bu	butyl
n-BuLi	*n*-butyllithium
t-BuLi	*tert*-butyllithium
°C	degree(s) Celsius
^{13}C NMR	carbon-13 nuclear magnetic resonance spectroscopy
calcd.	calculated
$CDCl_3$	deuterated chloroform
$CHCl_3$	chloroform
CH_2Cl_2	methylene chloride
cm^{-1}	reciprocal centimeters
δ	chemical shift in ppm from tetramethylsilane

d	doublet
DCM	dichloromethane
dd	doublet of doublets
de	diastereomeric excess
DMSO	dimethyl sulfoxide
ee	enantiomeric excess
eq.	equivalent(s)
Et	ethyl
Et_2O	diethyl ether
EtOAc	ethyl acetate
EtOH	ethanol
g	gram(s)
GC	gas chromatography
h	hour(s)
H^+	proton or protic acid
^1H NMR	proton nuclear magnetic resonance spectroscopy
HWE	Horner-Wadsworth-Emmons
HPLC	high-performance liquid chromatography
HRMS	high resolution mass spectrometry
HTIB	hydroxy(tosyloxy)iodobenzene
Hz	hertz
i-Pr	isopropyl
IR	infrared spectroscopy

J	coupling constant
kcal	kilocalorie(s)
K-Selectride®	potassium tri-*sec*-butylborohydride
L	liter(s)
L-Selectride®	lithium tri-*sec*-butylborohydride
LDA	lithium diisopropylamide
lit.	literature
M	molar
m	multiplet
Me	methyl
MeCN	acetonitrile
MeOH	methanol
mg	milligram(s)
MHz	megahertz
min	minute(s)
mL	milliliter(s)
mmol	millimole(s)
mol	mole(s)
mp	melting point
NIH	National Institutes of Health
NIS	*N*-iodosuccinimide
NMR	nuclear magnetic resonance
NOE	nuclear Overhauser effect

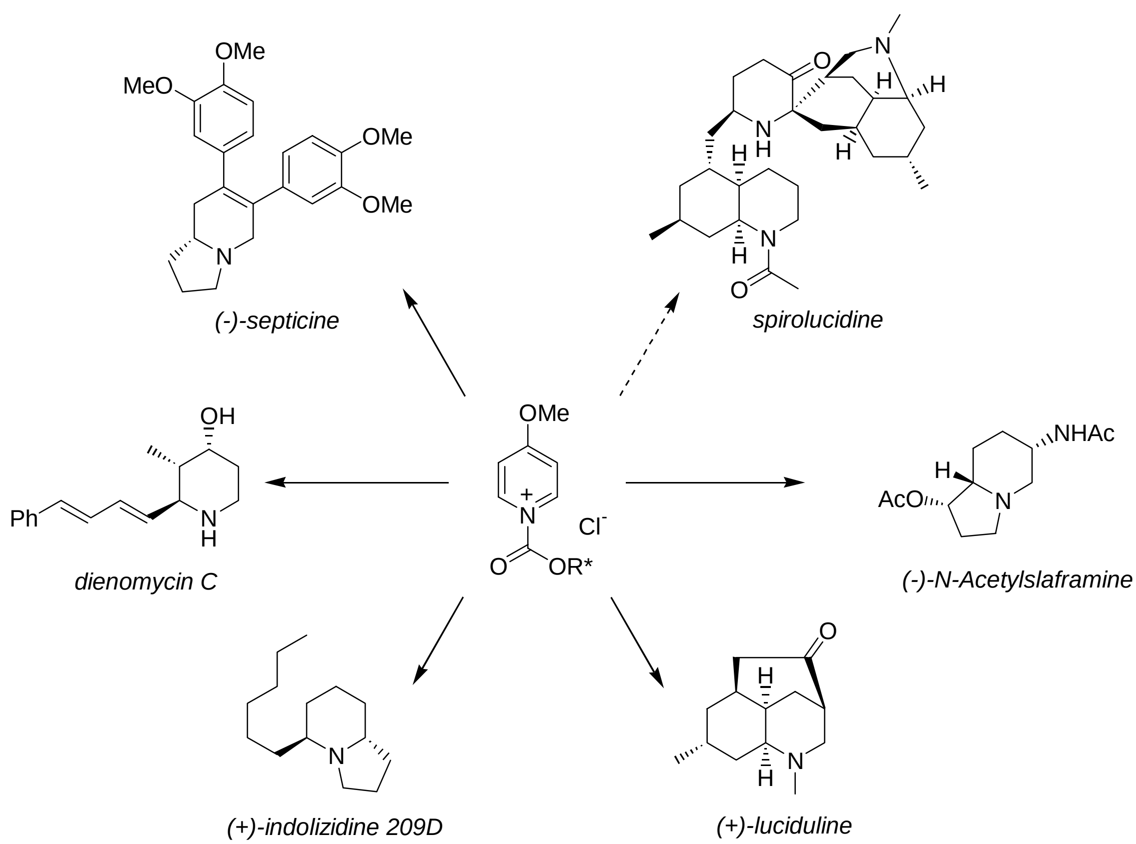
PCC	pyridinium chlorochromate
Ph	phenyl
ppm	parts per million
Pr	propyl
q	quartet
R	alkyl group
radial PLC	radial preparative-layer chromatography
rt	room temperature
s	singlet
t	triplet
t-Bu	*tert*-butyl
TCC	*trans*-2-(α-cumyl)cyclohexyl
TEA	triethylamine
THF	tetrahydrofuran
TIPS	triisopropylsilyl
TLC	thin-layer chromatography
TMS	trimethylsilyl
t_R	retention time
X	halogen atom

Introduction

N-Acyl-2,3-dihydro-4-pyridones

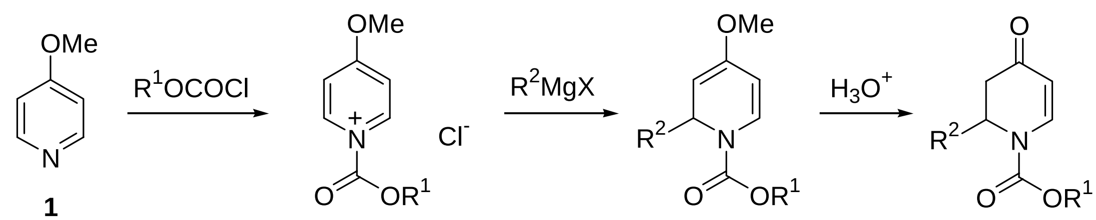
The Comins group has conducted extensive studies on 1-acyl-2,3-dihydro-4-pyridones and has used these unique heterocycles as building blocks for alkaloid synthesis. This unique ring system has been a valuable intermediate for synthesizing enantiopure indolizidine,[1] quinolizidine,[2] and piperidine[3] alkaloids (Figure 1).

Figure 1: Past and Present Targets Using *N*-Acylpyridinium Salt Chemistry



N-Acylpyridinium salts **2** are generated from commercially available 4-methoxypyridine (**1**) and a chloroformate. Treatment of the pyridinium salt, formed *in situ*, with an organometallic such as a Grignard reagent or a metallo enolate affords the 2-substituted 1-acyl-2,3-dihydro-4-pyridone **4** after subsequent quenching with aqueous acid (Scheme 1). Yields for these reactions are generally high, ranging from 85 to 95%.

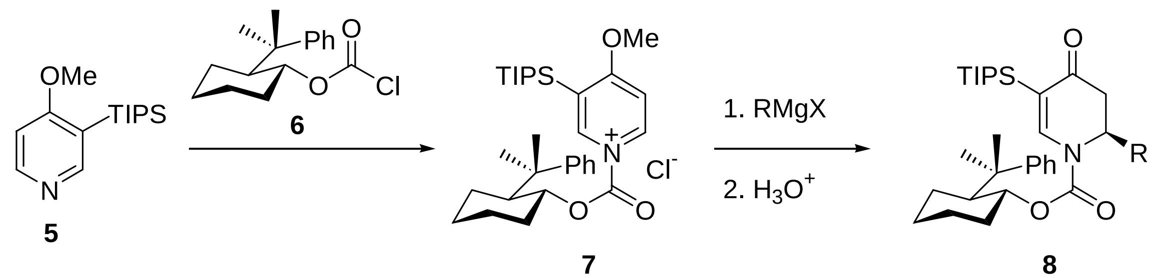
Scheme 1: Synthesis of Dihydropyridones



1 **2** **3** **4**

This methodology has been expanded to include the formation of highly enantiopure dihydropyridones by adding chloroformates of chiral alcohols, such as (-)- or (+)-*trans*-2-(α-cumyl)cyclohexyl chloroformate[4] or TCC chloroformate (**6**), to 4-methoxy-3-triisopropylsilylpyridine (**5**), followed by a Grignard reagent (Scheme 2). Diastereoselectivities range from 60 to 94% de with yields, comparable to that of the achiral dihydropyridone synthesis.[5] The triisopropylsilyl group at the C-3 position of the pyridine ring increases the de when a nucleophile, such as a Grignard reagent, is added to the ring. The bulkiness of the silyl group aids in blocking one of the sites at the carbon adjacent to the nitrogen, thereby regulating the site of attack of the nucleophile at the C-6 position. With the TIPS group present on the ring system at C-3, a rather poor de of 30% is increased considerably to 94%.

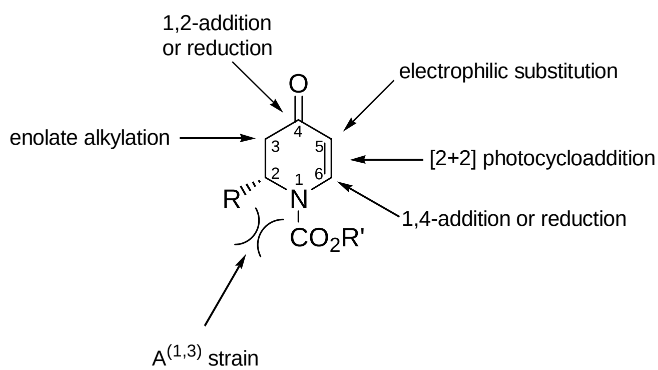
Scheme 2: Synthesis of Chiral Dihydropyridones



These dihydropyridones are excellent building blocks due to the useful functionality present on the heterocyclic ring (Figure 2). Several transformations can be carried out on the dihydropyridone. The olefin on the enone portion can undergo [2+2] photocycloaddition with other alkenes to form cyclobutanes.[6] The C-3 position can be alkylated by enolate formation.[1c,3d, 7] The enone functionality can serve as a Michael acceptor at C-6, undergo 1,4-reduction,[8] or undergo 1,2-addition to the carbonyl.[8b, 9] Finally, electrophilic substitution can occur at the C-5 position of the ring.[9, 10]

The stereochemistry is effected by the $A^{(1,3)}$ strain that is present between the carbamate and the R group shown in Figure 2. A conformational bias is observed because the $A^{(1,3)}$ strain causes the substituent at C-2 to be forced in the axial position.[11] This phenomenon is advantageous to these dihydropyridones because it allows stereochemical control of functionalities introduced at C-3, C-4, and C-6.

Figure 2: The Versatility of *N*-Acyl-2,3-dihydro-4-pyridones

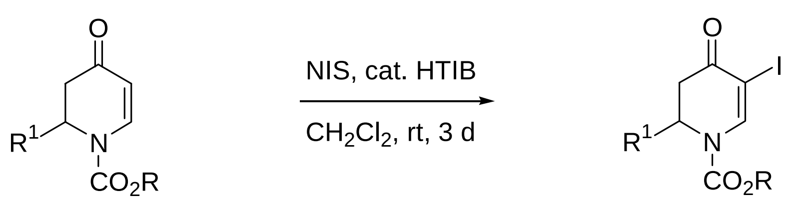


Literature Review

5-Iodo-2,3-dihydro-4-pyridones

Electrophilic substitution at the C-5 position has been thoroughly examined on these particular dihydropyridones.[9, 10] Studies conducted within the Comins group have successfully shown that dihydropyridones halogenate at the C-5 position with an electropositive source of a halide, such as *N*-iodo- or *N*-bromosuccinimide.[10b] Using *N*-iodosuccinimide (NIS) and a catalytic amount of [hydroxy(tosyloxy)iodo]benzene (Koser's reagent, HTIB),[11] various α-iodoenones have been prepared in moderate to high yields as shown in Table 1.[10b]

Table 1: Preparation of 5-Iodo-2,3-dihydro-4-pyridones



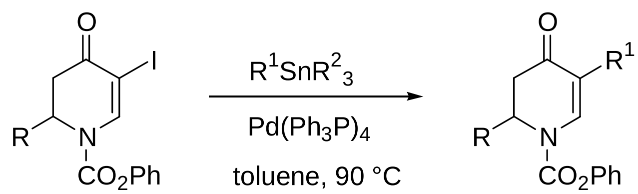
9 R, R^1 = Me, Ph **10** R, R^1 = Me, Ph

R	R^1	Yield,[a,b] (%)
Ph	CH_3	84
Ph	Ph	79
CH_3	CH_3	98
CH_3	Ph	90

[a] The reactions were performed on a 1.5-2.0-mmol scale. [b] Yield of purified product obtained from radial PLC (silica gel, EtOAc, hexanes).

With the halogen in place, various cross-coupling reactions can be carried out. Stille coupling reactions can be performed with vinyltin compounds in the presence of a Pd(0) reagent, such as tetrakis-triphenyl phosphine palladium (0).[12] In a study conducted with these dihydropyridones, vinyl iodide **11** was subjected to these conditions and the results are presented in Table 2.[10b]

Table 2: Stille Coupling Reactions of 5-Iodo-2,3-dihydro-4-pyridones



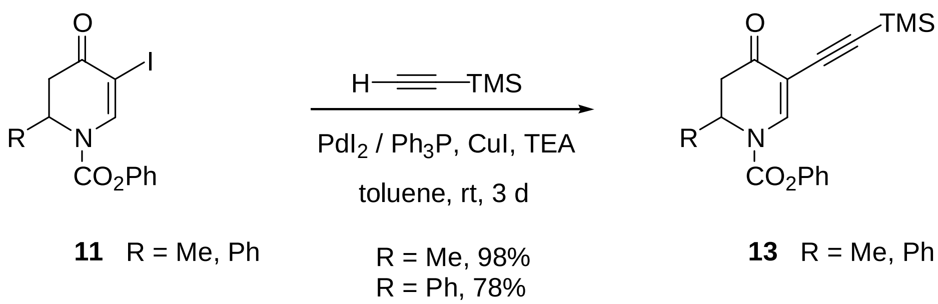
11 **12**

R	R^1	R^2	Yield,[a,b] (%)
CH_3	$CH=CH_2$	CH_3	79
CH_3	Ph	Bu	70
Ph	$CH=CH_2$	CH_3	70
Ph	Ph	Bu	61

[a] The reactions were performed on a 1.5-2.0-mmol scale. [b] Yield of purified product obtained from radial PLC (silica gel, EtOAc, hexanes).

Other functionality can also be introduced at the C-5 position using vinyl iodide **11** (Scheme 3).[13] Trimethylsilylacetylene (TMS-acetylene) can undergo coupling with a vinyl halide in the presence of a palladium catalyst and CuI to introduce acetylenes in high yields.[14] Moderate to high yields have been achieved using this palladium-catalyzed acetylenation condition with **11**.

Scheme 3: Cross-coupling of 5-Iodo-2,3-dihydro-4-pyridones with TMS-acetylene



11 R = Me, Ph

13 R = Me, Ph

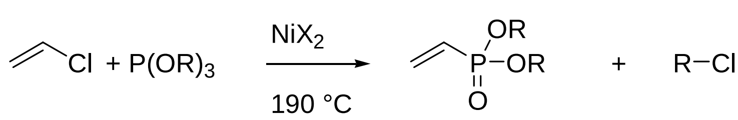
Cross-coupling Reactions with Vinyl and Aryl Halides

Most phosphonates are easily obtained by using the Arbuzov-Michaelis reaction.[15] This reaction involves the addition of an alkyl halide to triethyl phosphite to form a phosphonium salt. Upon heating, the salt is converted to a monoalkyl phosphonic ester. This ester can then be used in olefination reactions. Unfortunately, using this reaction with vinyl halides has resulted in little success.[16] Thus, an alternate route must be selected.

Cross-coupling reactions are commonly used for establishing carbon-heteroatom bonds, such as C-S,[17] C-N,[18] and C-P.[19] For the past three decades, numerous studies have been performed that combine aryl or vinyl halides with various phosphorus reagents through the use of metal catalyzed cross-coupling reactions.[20-22] This innovative transformation gives rise to phosphonate esters and phosphine oxides, which are capable of undergoing further transformations.

Initial studies involving vinyl chlorides, trialkyl phosphites, and catalytic amounts of nickel chloride or bromide have been performed by Tavs and coworkers (Scheme 4).[20a] These reactions were done neat and were heated in a sealed tube for approximately four hours to give the phosphonate ester **16** with yields ranging from approximately 50 to 90%. Various trialkyl phosphites were employed. Other investigations were carried out using aryl halides under similar conditions.[20b]

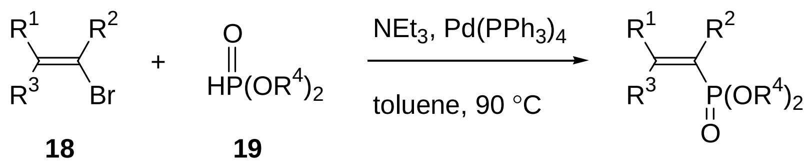
Scheme 4: Phosphorylation of Vinyl Chloride Using Nickel Catalysts



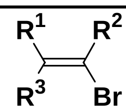
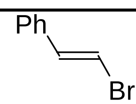
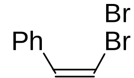
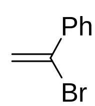
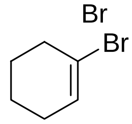
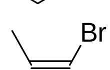
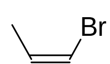
14 **15** **16** **17**

Hirao and coworkers have used an alternate method in making vinyl phosphonate esters **20**.[21] The compounds were generated by reacting vinyl bromides **18** with dialkylphosphites **19** in the presence of triethylamine and a catalytic amount of tetrakis(triphenylphosphine)palladium. Several different bromides were selected, thereby providing the corresponding phosphonate ester in yields ranging from 66 to 98% as shown in Table 3. Reactions were run at 90 °C for 0.2 to 30 hours depending on the bromide used. Unlike Tav's procedure, these reactions were run in a solvent such as toluene.

Table 3: Phosphorylation of Vinyl Bromides Using Palladium Catalysts



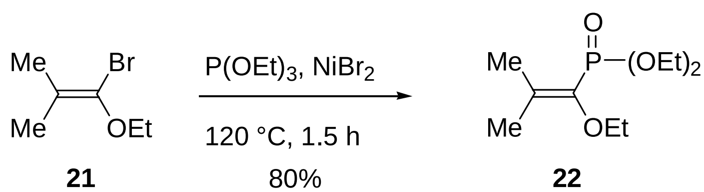
18 **19** **20**

Entry		HP (O) $(OR^4)_2$ R^4	Reaction Time,[a] (h)	Yield,[b] (%)
1		Et	0.2	93
2		Et	1.5	91
3		*i*-Pr	12	81
4		n-Bu	1.5	92
5		Et	30	66
6		Et	9	69
7		Et	1.5	98
8		*i*-Pr	6	92

[a] Reaction temperature; 90 °C. [b] Yield was based on isolated product

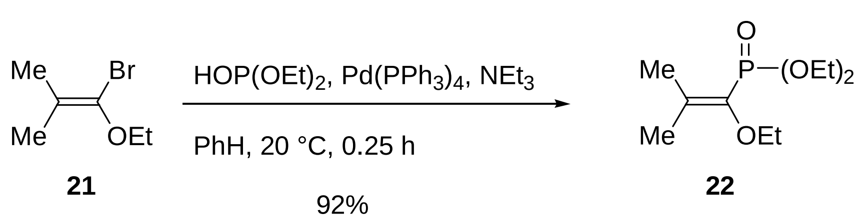
Recently, Kazankova and coworkers have synthesized a number of vinylphosphonates by transition metal catalyzed reactions using previous chemistry that has been established by Tavs and Hirao.[22] Specifically, they synthesized 1- and 2-alkoxy- and 1- and 2-dialkylaminoalkenylphosphonates using substituted vinyl halides with triethyl phosphite catalyzed by nickel halides or with diethyl phosphite catalyzed by palladium complexes in the presence of a base. Schemes 5 and 6 show one example of each reaction type.

Scheme 5: Cross-coupling Reactions of 1-Alkoxyalkenes with Triethyl Phosphite



Both routes produced high yields despite the varied reaction conditions between the two. The nickel-catalyzed reaction required heating (75-120 °C) and was run neat in triethyl phosphite. The palladium-catalyzed reaction performed run in benzene, diethyl phosphite, and a stoichiometric amount of base with no heating necessary.

Scheme 6: Cross-coupling Reactions of 1-Alkoxyalkenes with Diethyl Phosphite

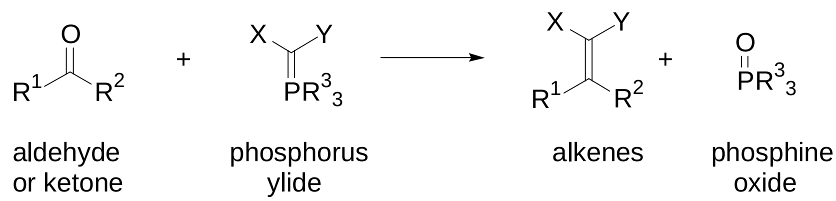


Wittig Reaction

Until 40 years ago, chemists had to suffer the difficulties concerning the location and geometry at which an olefin was introduced. In the early 1950s, however, Georg Wittig developed methodology using phosphorus chemistry in which alkenes could be synthesized and no ambiguity would exist as to the position of the double bond in the product.[23] Approximately ten years later, Horner, Wadsworth, and Emmons modified Wittig's methodology to increase the utility of the phosphorus-based olefination reactions.[24, 25]

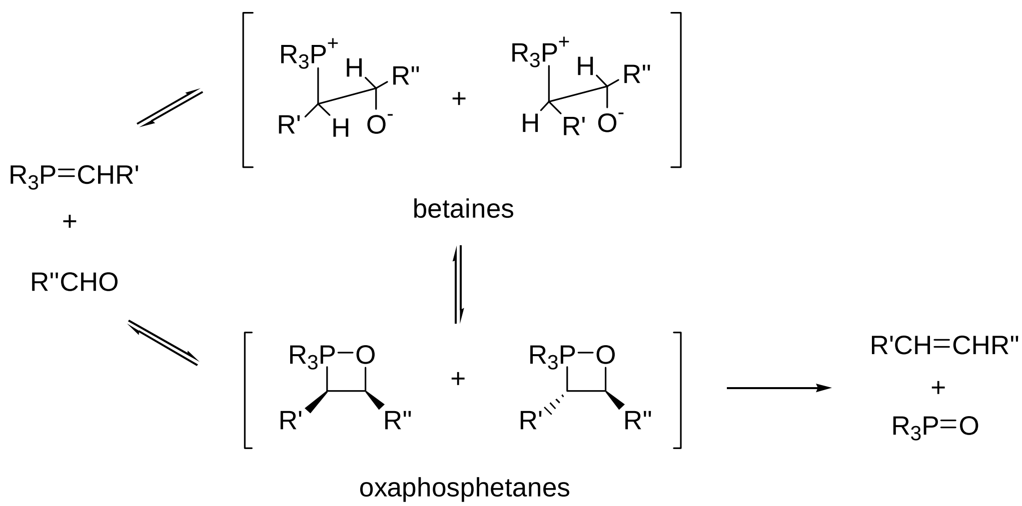
The typical Wittig method involves the reaction of an aldehyde or ketone with a phosphonium ylide to give an alkene (Scheme 7). Depending on the type of ylide, type of carbonyl, or reaction conditions employed, the formation of the alkene can favor either the (*Z*)- or (*E*)-alkene.[26] The (*E*)-alkene generally arises from "stabilized" ylides which contain strongly conjugating substituents, such as COOMe, CN, or SO_2Ph, on the ylidic carbon. Conversely, (*Z*)-alkenes are usually generated from ylides without such functionalities present, known as "nonstabilized" ylides. Ylides stabilized by mild conjugating substituents ("semistabilized" ylides), such as a phenyl or allyl group, normally show no partiality for one geometric isomer over the other.

Scheme 7: The Wittig Reaction



The mechanism of the Wittig reaction includes the nucleophilic addition of the phosphorus ylide to the aldehyde or ketone via a betaine or a 1,2-oxaphosphetane intermediate (Figure 3). The type of intermediate has been debated and studied extensively.[23, 26, 33] Initially, Wittig proposed an oxaphosphetane, a four-membered cyclic phosphorane, intermediate.[23a] After further study, he suggested the formation of the alkene proceeded by a zwitterionic phosphorus betaine.[23b]

Figure 3: Mechanism of the Wittig Reaction



Originally, evidence showed the dipolar betaine intermediate was the more probable of the two to occur. Firstly, the betaines had been successfully trapped as β-hydroxy phosphonium salts when acid was added at low temperatures.[26] In addition, studies conducted primarily by Schlosser showed that alkene stereochemistry was greatly effected when lithium salts were utilized.[27] However, shortly after, Vedejs was successful in isolating the oxaphosphetane and verified it by ^{31}P NMR as the only intermediate present with nonstabilized ylides.[28] Additional support for this mechanism was that uncomplexed betaine intermediates were not observed. Also, molecular modeling calculations were carried out with methylene phosphane and formaldehyde showing the betaine intermediate had a higher activation energy (ca. 32 kcal/mol) than the oxaphosphetane (ca. 7 kcal/mol), making the oxaphosphetane energetically more favorable.[29]

The Wittig reaction allowed, for the first time, stereoselective and facile formation of olefins. Altering the substituents on the ylidic carbon of an ylide can cause one geometric isomer to be favored over the other. The *E*-*Z* selectivity can be controlled by using Schlosser's modification of the Wittig reaction. The lithium salt of an ylide has been reported to selectively give the (*E*)-alkene,[27] whereas salt-free conditions normally give the (*Z*)-alkene.[30] This stereoselective control and alkene incorporation makes this type of reaction indispensable for organic synthesis.

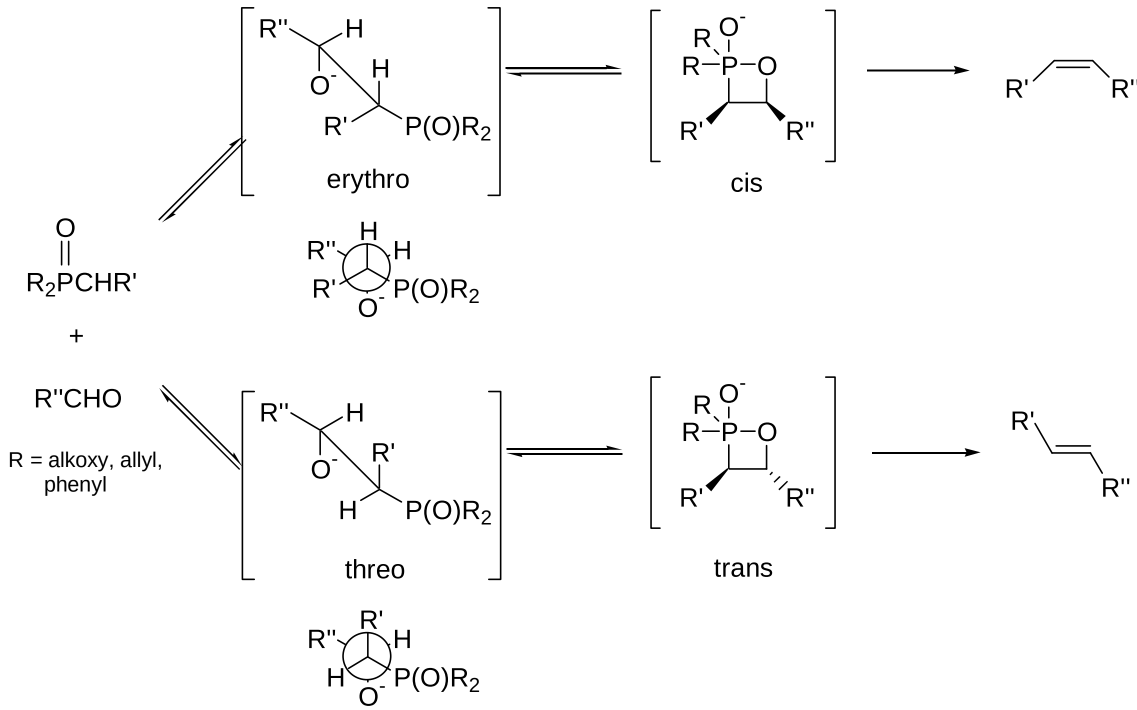
Horner-Wadsworth-Emmons Olefination

Horner and coworkers were the first to modify the Wittig reaction. They reacted phosphoryl-stabilized carbanions with ketones and aldehydes to make alkenes.[25a, 31] Derivatives of diphenylphosphine oxide and diethylbenzylphosphonate were used to obtain these particular carbanions. Wadsworth and Emmons, on the other hand, focused primarily on phosphonate carbanions to produce olefins.[24] Both phosphonate-mediated modifications of the Wittig reaction were combined and referred to as the Horner-Wadsworth-Emmons (HWE) olefination reaction.

The HWE reaction provides many advantages over the traditional Wittig reaction. Phosphonate carbanions are recognized to be more nucleophilic than phosphonium ylides, providing a higher diversity of aldehydes and ketones to react with these particular carbanions.[24, 32] A problem associated with the Wittig reaction is the phosphine oxide formed is often difficult to separate from the desired product. The phosphate ion formed from the HWE reaction is water-soluble thus providing easier separation from the desired product. Finally, smooth alkylation is possible on the α-carbon of the phosphonate carbanion, whereas phosphonium ylides generally do not alkylate well.[33]

The mechanism for olefin formation from stabilized phosphonate carbanions is similar to the conventional Wittig reaction (Figure 4).[26] The phosphonate carbanion reacts with the aldehyde or ketone in a reversible first step to form either the threo or erythro betaine intermediate. This intermediate then decomposes to give an olefin via a four-membered cyclic transition state.[33]

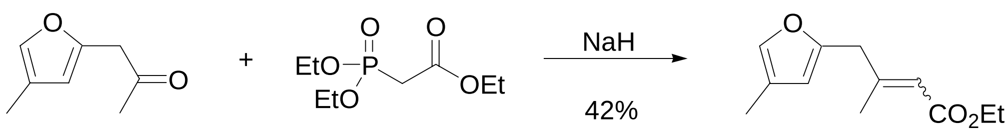
Figure 4: Mechanism of the Horner-Wadsworth-Emmons Reaction



The initial step of the HWE reaction governs the stereochemistry of the olefin. The threo betaine is the precursor to the *trans* product via a *syn* elimination, and in the same way the *cis* product is generated from the *cis* betaine. Generally seen with the HWE reaction, the *trans* olefin is favored over the *cis* olefin.[33] To support this observation, the formation and decomposition of the threo betaine must take place faster than that of the erythro betaine. This phenomenon can be explained by sterics. The erythro betaine in the eclipsed conformation is more hindered than the threo analog. Furthermore, the rate of decomposition from the threo betaine intermediate to the corresponding olefin is more rapid than the *cis* betaine to its resulting olefin due to the conjugative stabilization of the developing double bond in the transition state of the threo betaine.[33] This particular transition state is more difficult to achieve with the erythro betaine because R' and R" (Figure 4) cannot become coplanar with the incipient double bond.

The Horner-Wadsworth-Emmons olefination has found widespread applications in organic synthesis. A vital intermediate in the synthesis of longifolin utilized this convenient and versatile chemistry. Fukamiya and Yasuda treated ketone **23** with phosphonate **24** in the presence of sodium hydride to form ester **25** in 42% yield.[34] The selectivity was moderate with a *trans* to *cis* ratio of 3 : 1.

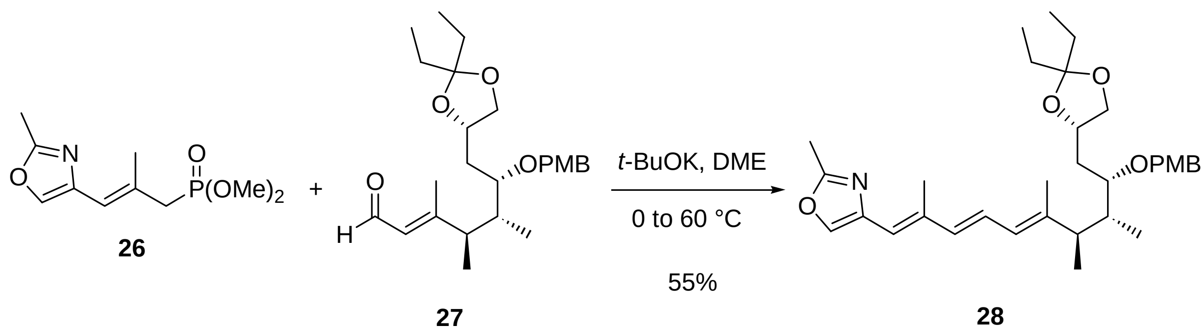
Scheme 8: Synthesis of Intermediate for Longifolin



23 **24** **25**

E : *Z* = 3 : 1

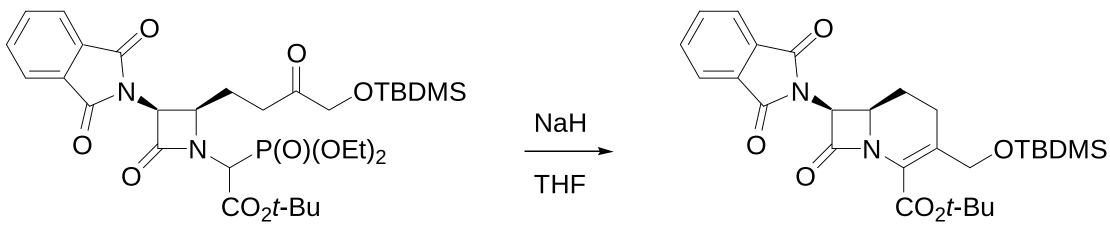
Scheme 9: Synthesis of Intermediate for Rhizoxin



In another synthesis Burke and coworkers used a HWE reaction to couple two critical components to form a key intermediate in the synthesis of macrolide rhizoxin.[35] Phosphonate **26** and aldehyde **27** were reacted with potassium *tert*-butoxide in DME to form *E*,*E*,*E*-triene **28** in 55% yield (Scheme 9). Excellent selectivity was achieved providing only the desired (*E*)-alkene. Interestingly, the solvent played a significant role in this reaction. When this same reaction was run in THF, a 1:1 mixture of alkene stereoisomers was formed.

Olefination reactions can be carried out in an intramolecular manner with reasonably stable ylides. The phosphonate ylide is more stable than the phosphorus ylide used in the Wittig reaction. The relatively stable phosphonate ylide can thus be subjected to intramolecular olefination reactions more easily than with Wittig reagents.[26] Cyclization, especially formation of a macrocyclic ring system, is very common using the intramolecular HWE reaction. Rings containing 5, 6, 12, 13, 14, 15, 16, 18, 20, and 38 atoms have successfully been made using this chemistry.[26]

Scheme 10: Synthesis of Intermediate for 3-(Hydroxymethyl)carbacephalosporin

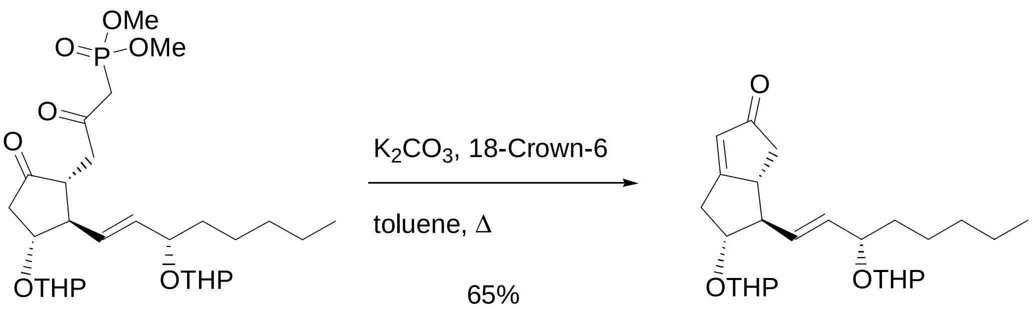


29 30

Miller and coworkers successfully cyclized the sodium salt of ketone **29** affording bicyclic β-lactam **30** (Scheme 10). This lactam is a key intermediate for the total synthesis of 3-(hydroxymethyl)carbacephalosporin, an important antibacterial agent.[36] The reaction afforded the cyclized product (**30**) in 85% yield producing a single diastereomer.

Another example demonstrating the versatility of the Horner-Wadsworth-Emmons reaction is found within the synthesis of 6a-carbaprostaglandin I_2 by Aristoff. The highly strained bicyclo[3.3.0]octenone **32** is synthesized from ketone **31** (Scheme 11).[37] These particular octenones are normally formed via an intramolecular aldol-type condensation from the corresponding 1,4-diketone.[37] In this case, double bond migration around the cyclopentenone ring could occur which would result in very low yields of the formation of **32**. The HWE reaction became a viable alternative for this transformation. Treatment of ketone **31** with potassium carbonate and 18-crown-6 afforded the desired bicyclic compound (**32**) in 65% yield. The function of the crown ether was to increase the nucleophilicity of the β-ketophosphonate anion.

Scheme 11: Synthesis of Intermediate for 6a-Carbaprostaglandin I_2



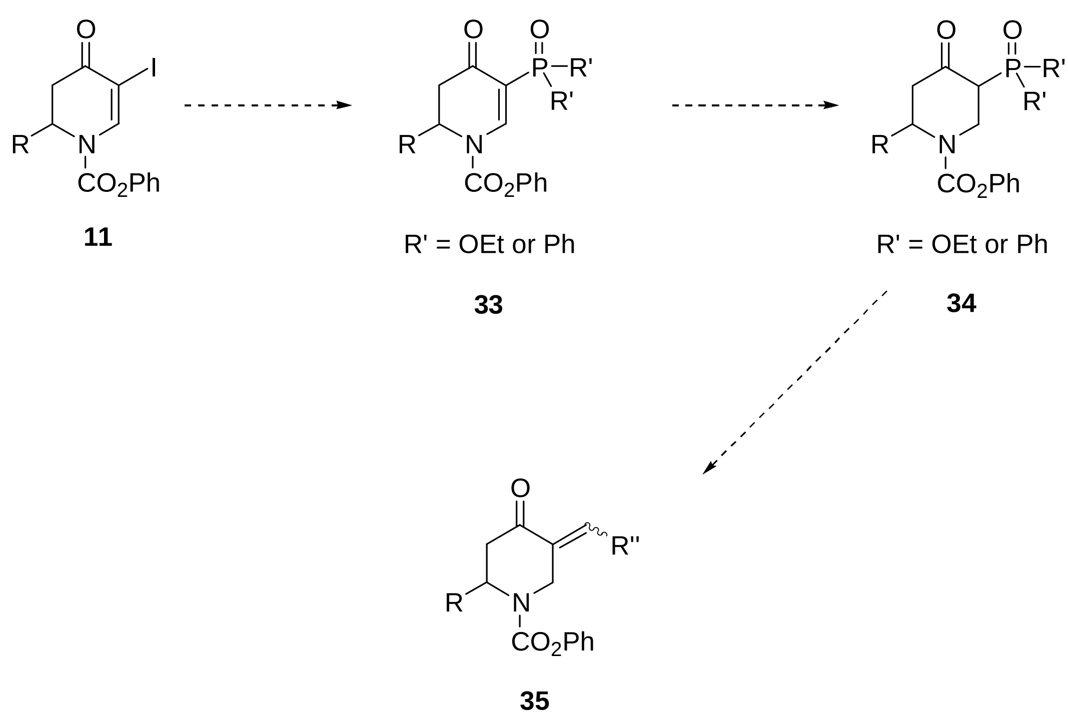
31 **32**

Results and Discussion

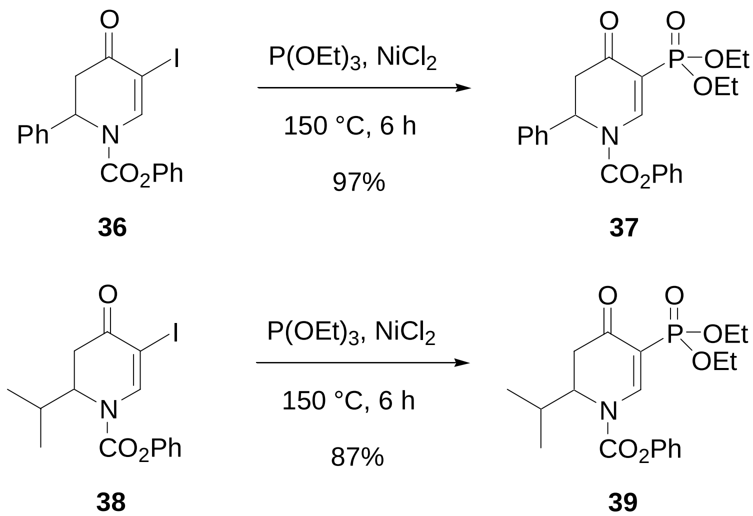
Intermolecular Horner-Wadsworth-Emmons Olefination Study

2,3-Dihydro-4-pyridones have been the primary focus of the Comins research group for the past number of years. With the functionality present in 2,3-dihydro-4-pyridones (Figure 2), many functional groups can be selectively introduced onto the ring system. Introducing an olefin at the C-5 position would prove valuable in supplementing this chemistry. This transformation could be accomplished by first introducing a phosphine oxide or a phosphonate group by cross-coupling with the iodide **11** at the C-5 position. After selectively reducing the double bond, a modified Wittig reaction could then be used to obtain enone **35** (Scheme 12).

Scheme 12: Synthetic Plan for Intermolecular HWE Olefination



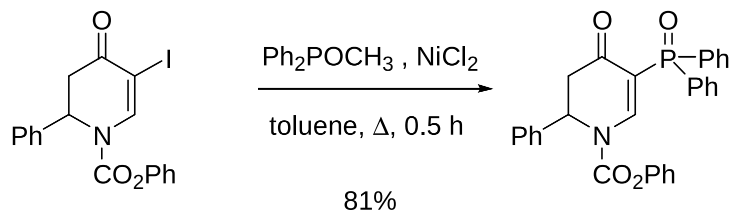
Scheme 13: Phosphorylation Using Triethyl Phosphite and Nickel Chloride



The synthesis of 5-iodo-2,3-dihydro-4-pyridones has been previously developed by the Comins group.[10b] The work recently completed by Kazankova and coworkers allows phosphonates and phosphine oxides to be incorporated in a compound formed from vinyl and aryl halides.[22]

Both palladium and nickel catalyzed cross-coupling reactions from Kazankova's procedure were attempted on α-iododihydropyridones **36** and **38**. The first attempt was with triethyl phosphite and a catalytic amount of nickel chloride at a constant temperature of 150 °C for six hours. Initially, the reactions were run in several different solvents without success. It was later found that the reaction must be run neat. The yields were nearly quantitative (97%) for the phenyl-substituted dihydropyridone **36** and reasonably high (87%) for the isopropyl-substituted dihydropyridone **38** (Scheme 13).

Scheme 14: Phosphorylation Using Methyl Diphenylphosphinite and Nickel Chloride

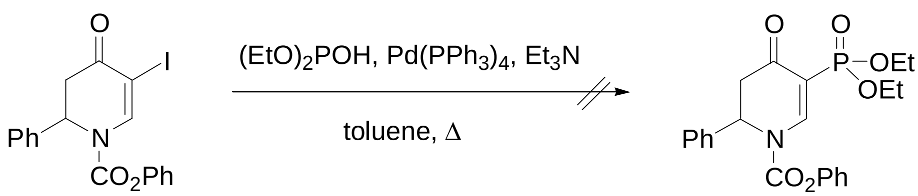


36 **40**

To obtain a Horner precursor as a phosphine oxide, methyl diphenylphosphinite was substituted for triethyl phosphite. In this case, the reaction was run in refluxing toluene, and TLC monitoring illustrated that the reaction went to completion after only 30 minutes (Scheme 14). Unfortunately, an inseparable by-product was observed with this reaction. Increasing the number of equivalents of methyl diphenylphosphinite showed higher levels of the undesired by-product via NMR analysis. Therefore, the number of equivalents of the phosphorus reagent was varied. It was found that fewer than 4 equivalents led to some unreacted starting material, while increasing the number of equivalents led to a greater amount of the by-product. All attempts at using chromatography with several different solvent systems failed. The phosphine oxide purification was finally resolved through crystallization. The compound was finally purified by crystallization using a seeding technique.

This specific conversion was not found in current literature. Advantages of using this novel transformation over using triethyl phosphite were that the reaction could be run in a solvent and that the reaction time was significantly decreased. However, methyl diphenylphosphinite is considerably more expensive than triethyl phosphite and purification proved to be problematic. For this reason, we decided to continue using the phosphonate ester instead of the phosphine oxide for further study.

Scheme 15: Attempted Phosphorylation Using a Palladium Catalyst

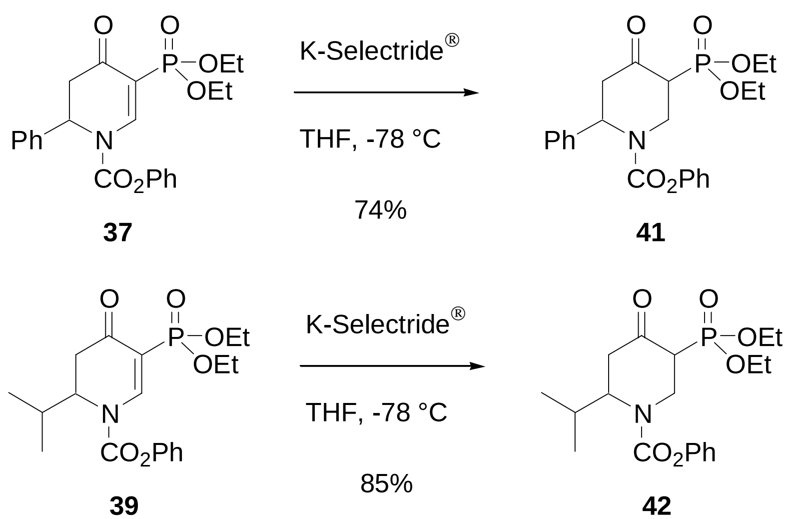


36 37

Using chemistry reported by Kazankova,[22] the last attempt of phosphorylation with the vinyl iodide of dihydropyridones used palladium catalysts in place of nickel halides. This conversion used diethyl phosphite in the presence triethylamine and a catalytic amount of tetrakis(triphenylphosphine)palladium (Scheme 15). After 24 hours at room temperature, starting material still remained. When the reaction was heated the compound decomposed. Due to these negative results and the success with nickel catalysts, this route was abandoned.

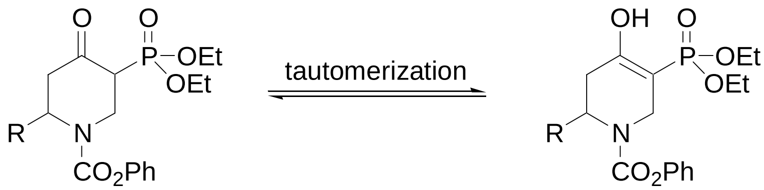
With the phosphonate in place, 1,4-reduction of α,β-unsaturated ketones **37** and **39** provided precursors **41** and **42** for the HWE olefination reactions. Selectrides® (or tri-*sec*-butylborohydrides) have been common reagents used for the reduction of the enone portion of these dihydropyridones.[38] Both L- and K-Selectrides® were attempted on the two enones. K-Selectride® (potassium tri-*sec*-butylborohydride) gave better yields than its lithium derivative. Enones **37** and **39** were stirred with K-Selectrides® at -78 °C for thirty minutes to give reduced compounds **41** and **42** with yields of 74% and 85%, respectively (Scheme 16).

Scheme 16: 1,4-Reduction of α,β-Unsaturated Ketones with K-Selectride®



β-Ketophosphonates **41** and **42** exhibited an unexpected singlet at approximately 11.0 ppm in the ^{1}H NMR spectra. We discovered that this singlet corresponded to the corresponding tautomers in the enol form (Scheme 17). It was later found that this tautomerization proved useful in a future transformation.

Scheme 17: Tautomerization of the β-Ketophosphonates



41, **42** R = Ph or *i*-Pr **43, 44** R = Ph or *i*-Pr

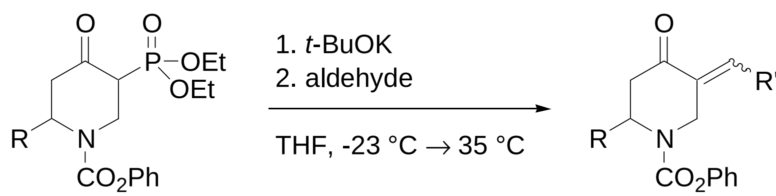
With the reduced compounds in hand (**41**, **42**), we could use the Horner-Wadsworth-Emmons reaction to olefinate at the C-5 position. A base had to be added to form the anion. Once the anion was prepared, various aldehydes could be added to introduce different olefins.

For all the intermolecular reactions carried out in this study, the same reaction conditions were employed and only the aldehyde was varied. We added potassium *tert*-butoxide in a solution of THF to the reduced β-ketophosphonate at -23 °C to produce the anion *in situ*. After ten minutes, the aldehyde was added and the mixture was heated at 35 °C overnight (Table 4). The aldehydes that were used for this study included benzaldehyde, hexanal, hydrocinnamaldehyde, and *trans*-cinnamaldehyde.

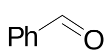
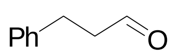
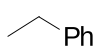
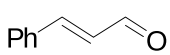
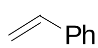
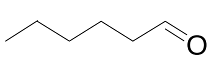
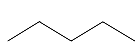
These reaction conditions gave the best results. Sodium hydride was another base that was used but the yields were considerably lower. Temperatures were key when carrying out this transformation. The anion did not seem to survive very well at temperatures greater than -23 °C because yields were much poorer when the reaction was attempted at warmer temperatures. The slight heating was necessary after the addition of the aldehyde, since after 3 days at room temperature, mostly starting material was observed.

The yields for the formation of the alkenes ranged from 61 to 92% depending on the aldehyde used (Table 4). Reactions with benzaldehyde (Entry 1) gave the highest yields, whereas *trans*-cinnamaldehyde (Entry 3) gave the lowest yields. The stereoselectivity for olefin formation varied greatly from aldehyde to aldehyde. The highest selectivity was observed with hexanal and benzaldehyde.

Table 4: Horner-Wadsworth-Emmons Olefination with Various Aldehydes



41, **42** **45-52**

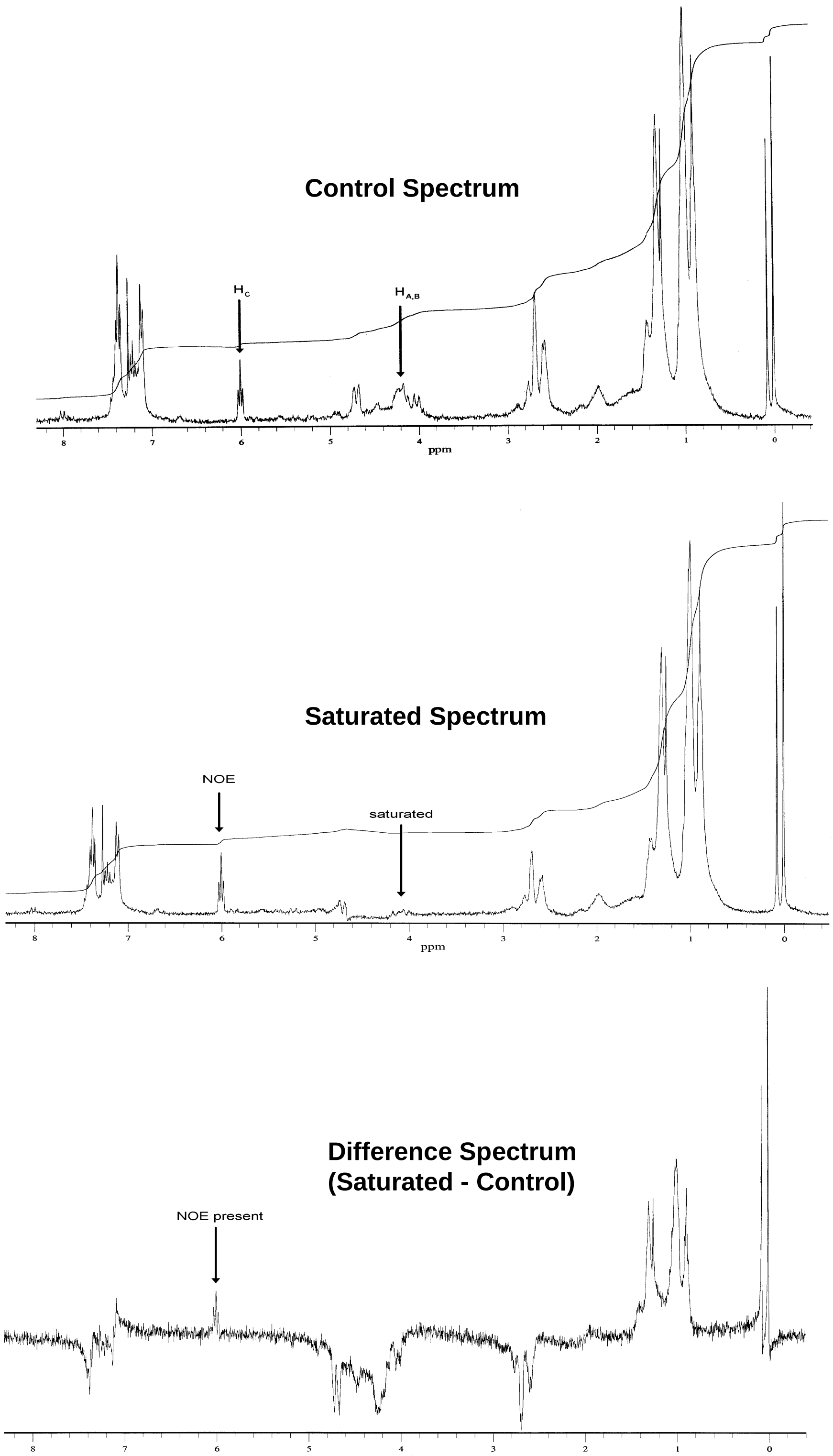
Entry	Aldehyde	R'	Olefin R = Ph	Olefin R = *i*-Pr	Yield,[a,b] (%) R = Ph	Yield,[a,b] (%) R = *i*-Pr	*E* : *Z* Ratio of Olefin[c] R = Ph	*E* : *Z* Ratio of Olefin[c] R = *i*-Pr
1	 *benzaldehyde*	Ph	**45**	**49**	92	84	8.3 : 1	11.6 : 1
2	 *hydrocinnamaldehyde*		**46**	**50**	75	74	6.4 : 1	1.2 : 1
3	 *trans-cinnamaldehyde*		**47**	**51**	64	61	3.1 : 1	3.6 : 1
4	 *hexanal*		**48**	**52**	80	76	8.8 : 1	12.3 : 1

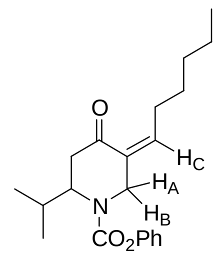
[a] The reactions were performed on a 1.5-3.0-mmol scale. [b] Yield of purified product obtained from radial PLC (silica gel, EtOAc, hexanes). [c] Isomer ratios were determined by HPLC

Despite several attempts, purification of the *E*,*Z*-isomers away from each other proved impossible. Therefore, all of these olefins were characterized as a mixture of both (*E*)- and (*Z*)-isomers. Although purification by simple chromatography failed, separation was achieved by using HPLC, and the data obtained from the chromatogram allowed us to determine the relative ratios of the geometric isomers.

Our hypothesis was that the (*E*)-isomer was the major isomer, and the (*Z*)-isomer was the minor isomer. This was in concurrence with most results obtained from the HWE reaction.[34, 35] We were able to purify olefin **52** into its separate isomers by collecting the first fraction (minor isomer) and last fraction (major isomer) of the chromophore by using radial PLC. This enabled us to perform a nuclear Overhauser effect (NOE) experiment on each of the isomers. A triplet corresponded to H_C at 6 ppm in the ^{1}H NMR spectrum (Figures 5 and 6) for compound **52b**. When proton H_A or H_B (4.0 – 4.2 ppm) was irradiated, an NOE with H_C should have been observed for the (*Z*)-isomer. It was found that the minor isomer exhibited a 6.4% enhancement of the triplet corresponding to H_C for **52b**. The major isomer, on the other hand, exhibited no NOE when the same protons were irradiated. This experiment verified that the (*E*)-isomer was indeed the major isomer formed.

Figure 5: Nuclear Overhauser Effect Experiment on Compound 52b

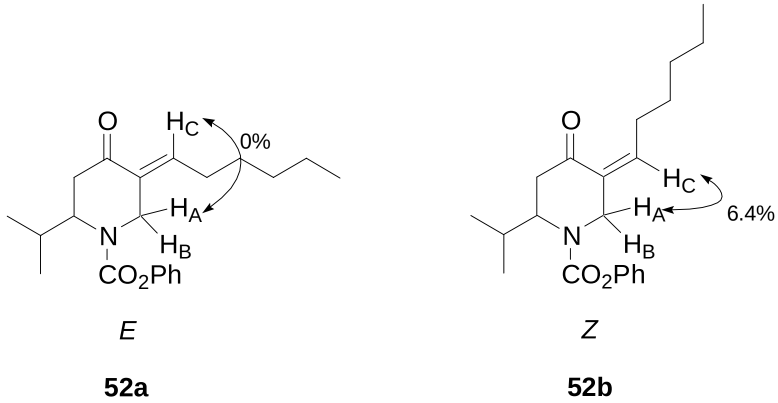




52b

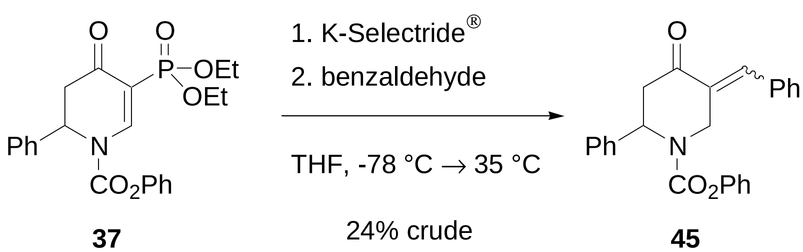
Z

Figure 6: Nuclear Overhauser Effect Experiment on *E* and *Z* Isomers



A one-pot procedure was also attempted that used unreduced phosphonate **37** to obtain olefin **45**. The phosphonate ester was reduced with K-Selectride® at -78 °C, and the aldehyde was added and the mixture was warmed to 35 °C (Scheme 18). The desired olefin **45** was obtained, although many other products were also observed by TLC. With the one-pot procedure having a crude yield of 24%, the two-pot procedure proved to be the more desirable method of olefin formation.

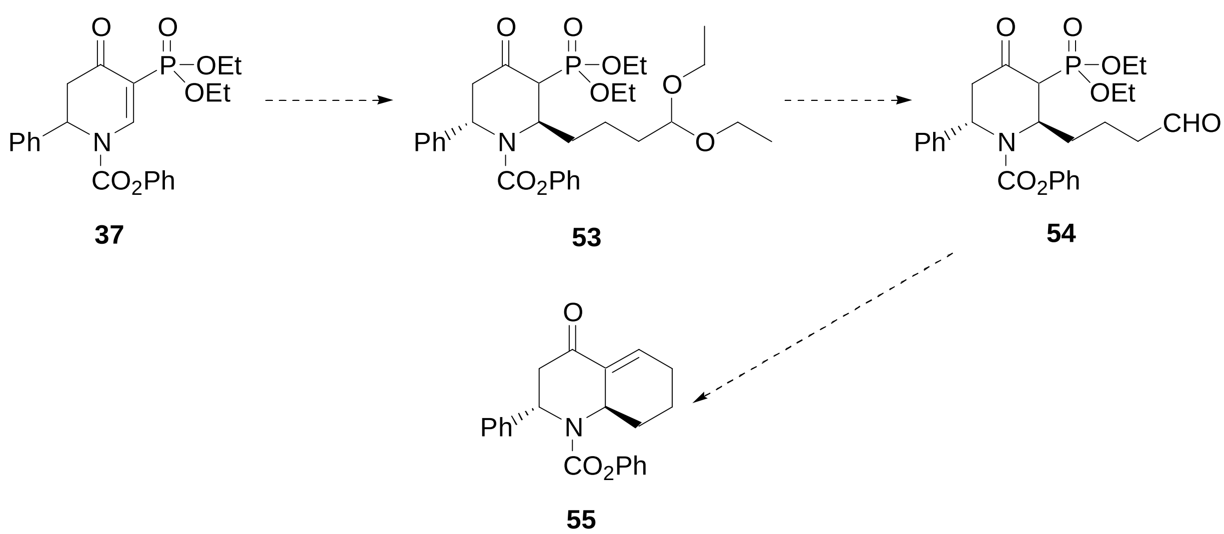
Scheme 18: One-Pot Olefination Procedure



Intramolecular Horner-Wadsworth-Emmons Olefination Study

This methodology can be further extended by using the HWE reaction for ring formation via intramolecular process (Scheme 19). By having an alkyl aldehyde present on the carbon β to the phosphorus group on compound **37**, cyclization might be induced to form a bicyclic ring **55**. We envision forming a protected aldehyde **53**, followed by hydrolysis to aldehyde **54**, and finally using a HWE olefination reaction to obtain bicyclic enone **55**.

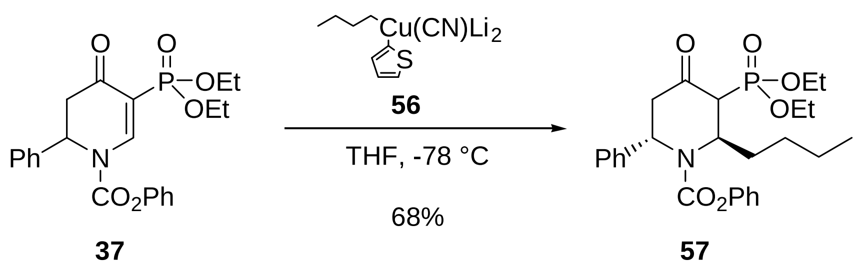
Scheme 19: Synthetic Plan for Intramolecular HWE Olefination



Higher order cuprates have previously been used in the Comins group for 1,4-additions on dihydropyridones.[39] This class of cuprates normally gives excellent yields of conjugate addition with α,β-unsaturated compounds.[40a] Also, it has been observed that higher order cuprates generally proceed in higher yield and selectivity for conjugate additions as compared to the analogous lower order cuprates (Gilman reagents).[41]

In our study, butylcyanocuprate reagent **56** was made from lithium 2-thienylcyanocuprate (Lipshutz reagent)[40b] and *n*-BuLi at -78 °C. After 30 minutes of stirring, phosphonate **37** in a solution of THF was transferred to the organocuprate, and the mixture was allowed to stir for 3 hours. After quenching the reaction with saturated aqueous sodium bicarbonate, the Michael addition product **57** was obtained with a yield of 68%.

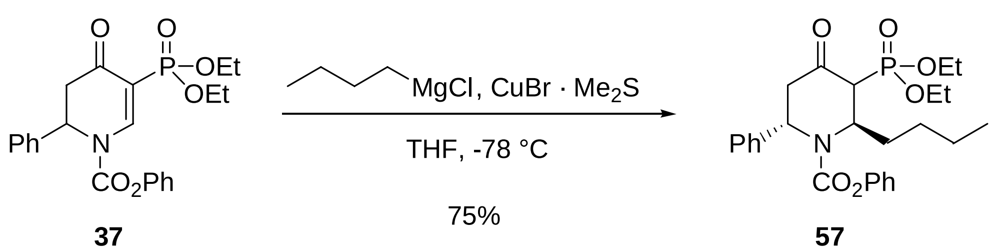
Scheme 20: Conjugate Addition Using a Higher Order Cuprate



Nucleophilic addition of a Grignard reagent can occur at either the acyl carbon (1,2-addition) or the alkenyl carbon (1,4-addition) of an enone. Generally, using cuprous [Cu(I)] salts along with Grignard reagents facilitates conjugate addition. This is due to the increased reactivity of the Grignard reagent and also to the enhanced coordination of copper to the carbonyl.

The same conjugate addition product (**57**) was obtained using butyl Grignard with a Cu(I) salt. This copper-magnesium reagent was formed by stirring butylmagnesium chloride with copper bromide at -78 °C for 5 minutes. The phosphonate enone (**37**) was then added to the Grignard-copper salt suspension and allowed to stir for 45 minutes. Subsequent quenching with saturated aqueous ammonium chloride gave **57** in 75% yield.

Scheme 21: Conjugate Addition Using a Copper-Assisted Grignard

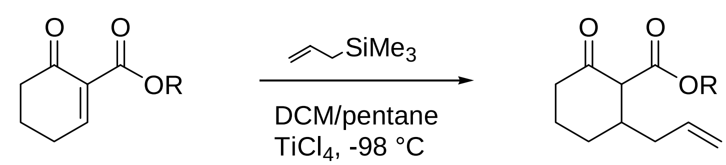


The de's were not able to be determined because **57** exists partially as a tautomer (Scheme 17, page 25). As the β-ketophosphonate **57** is shown in Scheme 21, the phosphonate group can be either up or down since it is an *sp*^{3} center. In the enol form, this group can only exist in one orientation since the double bond is present. The presence of this double bond serves to make the carbon *sp*^{2}-hybridized. For this reason, the diastereomeric excess can not be determined for the conjugate addition product with the phosphonate group present. It was later found that the nucleophilic addition of the Grignard takes place *trans* to the phenyl group.

Three attempts were made to dephosphorylate compound **57**. The first attempt was made using a dissolving metal reduction. The second was made through refluxing Zn^0 and acetic acid. The last attempt was done by adding samarium iodide with HMPA. Each instance resulted in the phosphonate group remaining in tact on the ring.

Introduction of other groups were also attempted onto the same enone **37** using Lewis acids. Recently, Myers and coworkers showed that the addition of allyltrimethylsilane in the presence of a Lewis acid was possible with α-carboalkoxy α,β-unsaturated ketone **58** using the Sakurai reaction (Scheme 22).[42] The yields were high and gave almost exclusively Michael addition product **59**.

Scheme 22: Conjugate Addition to β-Ketoester Using Allyltrimethylsilane



58 **59**

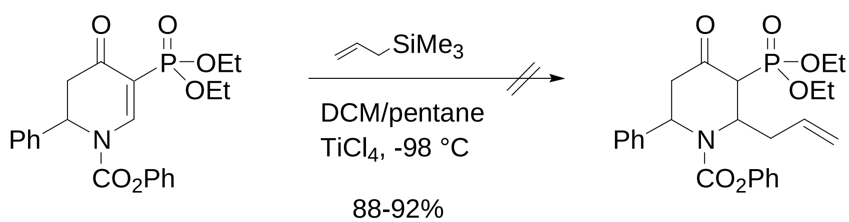
R = CH_3, *t*-Bu R = CH_3, *t*-Bu

(Myers)[42]

These same conditions were employed with enone **37** (Scheme 23). However, allyltrimethylsilane did not add to the enone even when warmer temperatures were tried.

There were sufficient dissimilarities between the two α,β-unsaturated ketones (**58** and **37**) that could account for the nonreactivity of the one enone and the success of the other.

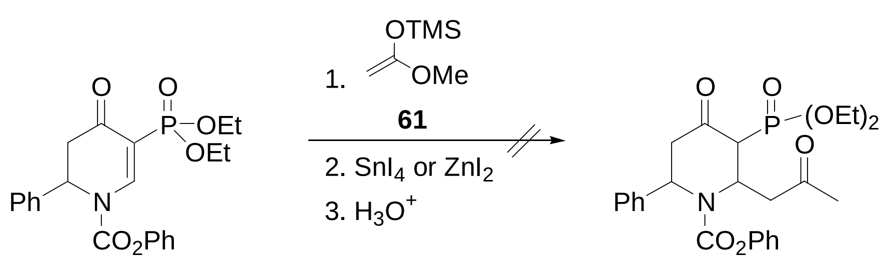
Scheme 23: Conjugate Addition Using Allyltrimethylsilane



37 **60**

Another attempt to conjugately add to enone **37** with a Lewis acid and *O*-silyl ketene acetal was unsuccessful (Scheme 24). This particular enone was unreactive when either $SnCl_4$ or ZnI_2 was used as the Lewis acid. Studies on dihydropyridones without the phosphonate group present were successful, giving yields near 85%.[43]

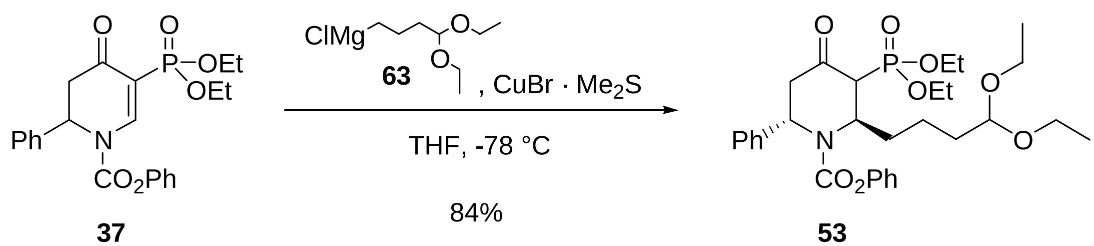
Scheme 24: Conjugate Addition Using *O*-Silyl Ketene Acetal



37 **62**

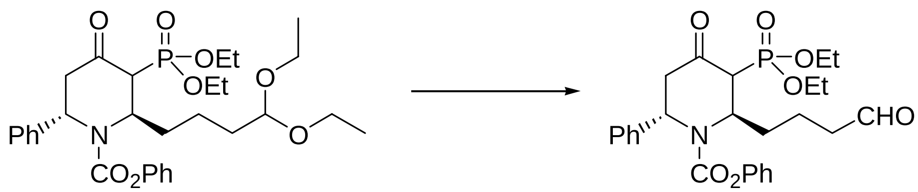
We proceeded with the 1,4-addition reactions of **37** by introducing an acetal side chain using the copper-mediated Grignard reaction as previously done with the butyl Grignard (Scheme 21). The acetal Grignard was formed by refluxing 4-chloro-1,1-diethoxybutane with magnesium in THF for 4 hours (Scheme 25). The newly formed Grignard **63** was added to copper bromide. The electrophile **37** was then added to the Grignard-copper salt suspension, and the mixture was allowed to stir for 45 minutes. Subsequent quenching with saturated aqueous ammonium chloride gave **53** in 84% yield.

Scheme 25: Conjugate Addition with Acetal Using a Copper-Assisted Grignard



With the protected aldehyde in place, the next step involved the hydrolysis of acetal **53** (Table 5). Weaker acidic conditions, represented by entries 1 and 2, were first attempted without much success. Because these reaction conditions were so mild, saturated aqueous oxalic acid was tried. The reaction went to completion after allowing the acetal to stir in saturated aqueous oxalic acid overnight to provide aldehyde **54**.

Table 5: Hydrolysis of Acetal 53

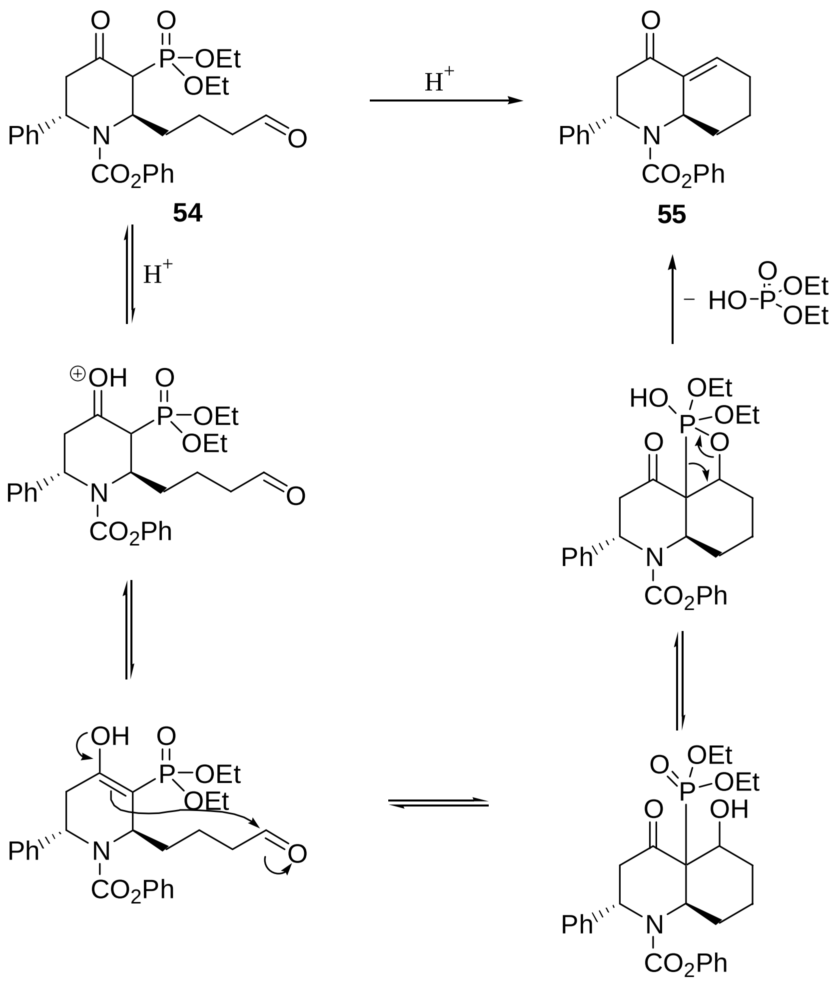


53 **54**

Entry	Conditions	Result
1	silica gel, 10% oxalic acid, CH_2Cl_2, 16 h	very little product formed, mostly SM
2	silica gel, 10% H_2SO_4, CH_2Cl_2, 2 d	little product formed, mostly SM
3	sat'd. oxalic acid, THF, 16 h	reaction went to completion, no SM present

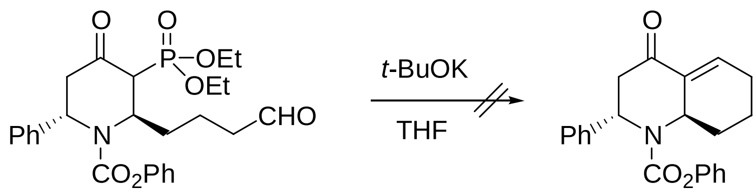
When purification of the aldehyde was carried out using chromatography, partial cyclization to bicyclic enone **55** was observed. This transformation was caused by the slightly acidic silica gel reacting with aldehyde **54** via an acid-catalyzed aldol-type condensation. A HWE reaction followed, to give the annulation (Scheme 26). Unfortunately, only about 20% of the bicyclic enone **55** was obtained in this manner.

Scheme 26: Acid-catalyzed Aldol Condensation Mechanism



In an effort to obtain the cyclized compound **55** with a HWE reaction similar to the intermolecular study, we subjected the crude aldehyde to potassium *tert*-butoxide (Scheme 27). Surprisingly, the crude NMR spectrum showed no formation of the cyclized product, but rather it showed that all starting material remained.

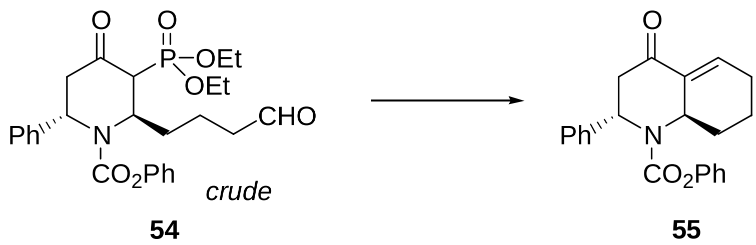
Scheme 27: Attempted Cyclization with Potassium *tert*-Butoxide



54 **55**

Since the conventional method did not work, we turned to other acidic and basic conditions to obtain bicyclic enone **55** (Table 6). The acidic conditions, represented by entries 1 through 3, were tried in efforts to mimic the acid-catalyzed aldol reaction observed with silica gel. The last four entries were basic conditions where deprotonation of the hydrogen alpha to the phosphorus was desired, followed by cyclization.

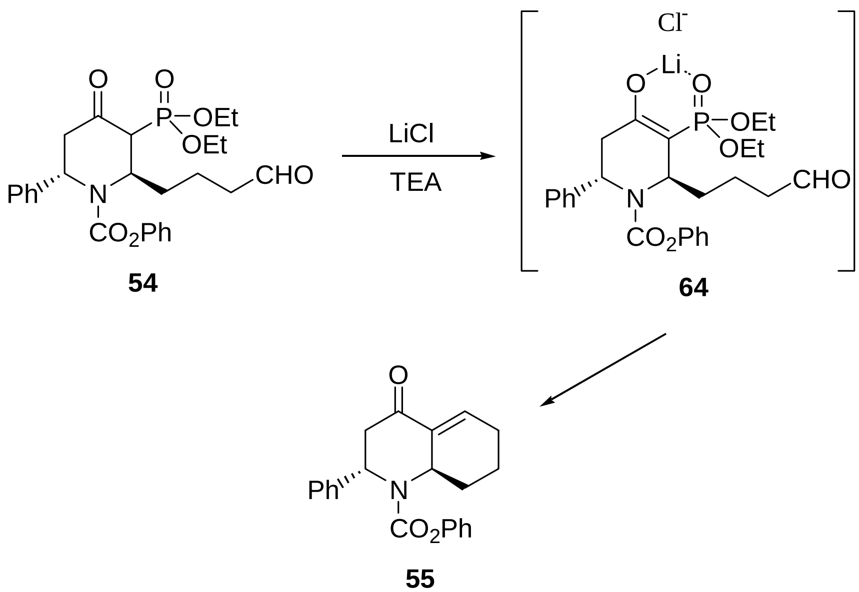
Table 6: Cyclization Using Various Conditions



Entry	Conditions	Result
1	aldehyde **54** run through silica gel column	very little product formed, mostly SM
2	CF_3CO_2H, $CHCl_3$, 1 h	all SM
3	Amberlyst® 15, THF, 3 d	all SM
4	NaOMe, MeOH, 8 h	all SM
5	NEt_3, THF, 8 h	very little product formed, mostly SM
6	NEt_3, $MgBr_2$, THF, 23 h	very little product formed, mostly SM
7	NEt_3, LiCl, THF, 1.5 h	89% yield of bicylic enone **55** from crude aldehyde

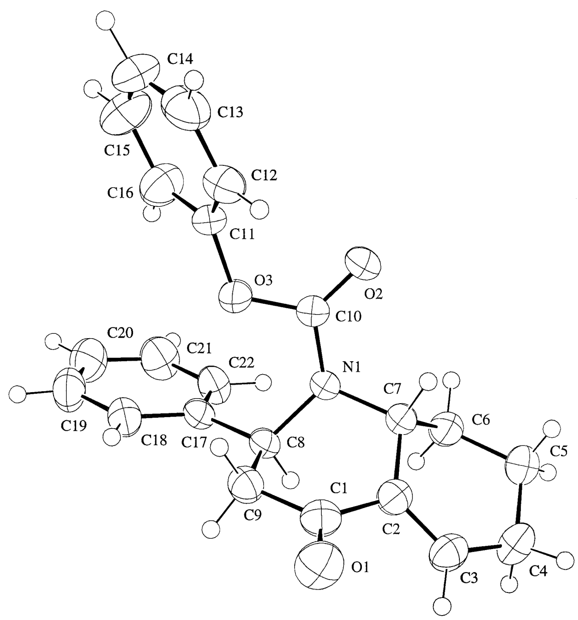
The acidic conditions resulted in little or no product formation. Entries 6 and 7 were reaction conditions reported by Rathke and coworkers[44] in which metal salts with triethylamine were used for deprotonation. The goal was to have the metal coordinate the two oxygens on the β-ketophosphonate to facilitate deprotonation (Scheme 28). The reaction of **54** with magnesium bromide and triethylamine (Table 6, Entry 6) gave low yields of the annulation product. However, treating crude aldehyde **54** with triethylamine in the presence of lithium chloride (Table 6, Entry 7) afforded the bicyclic enone **55** in 89% yield.

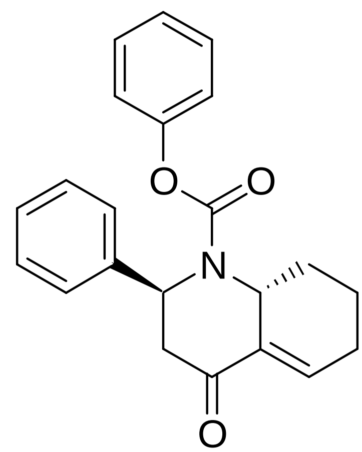
Scheme 28: Cyclization Using Lithium Chloride



The NMR spectrum of bicyclic enone **55** showed only one diastereomer present (> 95% de by HPLC). In order to decipher the relative stereochemistry between the C-2 and C-6 position, an X-ray structure of the bicyclic enone **55** was obtained (Figure 7). Results from this analysis showed that the phenyl group was *trans* to the C-6 (C-7 in Figure 7) alkyl chain on the ring. Also, it confirmed that the 1,4-addition of the acetal Grignard reagent **63** occurred stereoselectively and *trans* to the phenyl group. This high degree of stereoselectivity was concluded based on the high yields obtained from the Michael addition, the hydrolysis of the acetal, and the cyclization reaction. These high yields led to the final *trans* bicyclic enone **55**. Without the phosphonate group present at the C-5 position on the dihydropyridone, nucleophiles have been observed to attack *cis* relative to the C-2 substituent.[1d, 38a, 43] Thus, the phosphonate group appears to contribute to this novel reverse of stereoselectivity.

Figure 7: Crystal Structure of the Bicyclic Enone

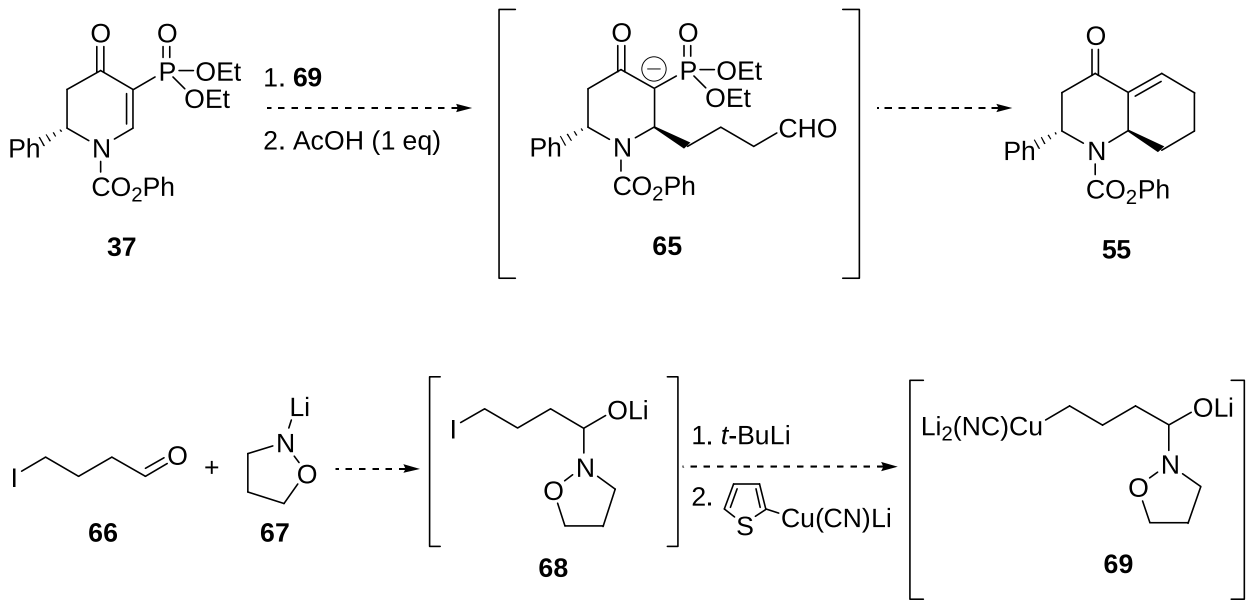




55

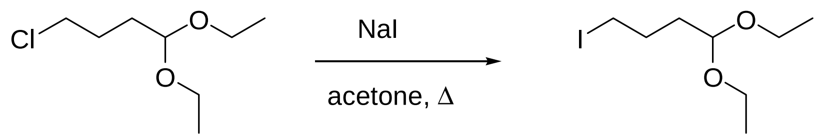
The above synthesis required three steps to obtain the cyclized product **55**. A one-pot cyclization of bicyclic enone **55** was then tried using higher order cuprates. Earlier, it was shown that a butyl higher order cuprate adds to phosphonate **37** in a moderate yield (Scheme 20). We envisioned using a more complex higher order cuprate, formed from isoxazolidine[45] and 4-iodobutanal **66**, to obtain bicyclic enone **55** (Scheme 29).

Scheme 29: Synthetic Plan for a One-Pot Cyclization



The first step was to synthesize 4-iodobutanal **66**. Using the same chloroacetal (4-chloro-1,1-diethoxybutane) as before, the chloride was displaced with an iodide by employing the Finkelstein reaction (NaI / acetone)[46] to obtain 4-iodo-1,1-diethoxy butane (**71**) with a small amount of the corresponding aldehyde **66** (Scheme 30).

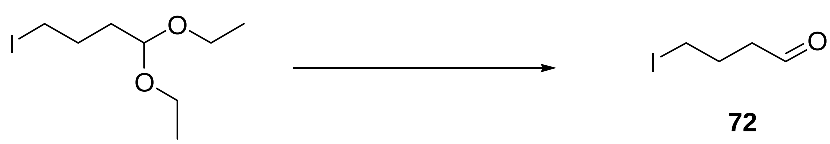
Scheme 30: Finkelstein Reaction on 4-Chloro-1,1-diethoxybutane



70 **71**

The first step of the reaction involved the hydrolysis of iodoacetal **71** to the desired aldehyde **66**. Several different acidic conditions were used. However, all conditions proved disappointing and led to decomposition of the product (Table 7).

Table 7: Attempted Formation of 4-Iodobutanal



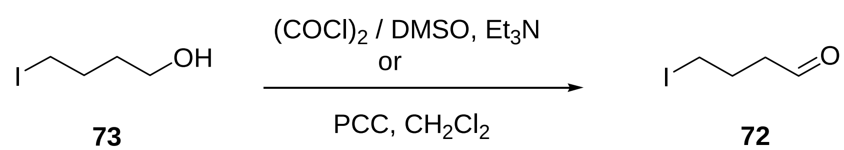
72

71

Entry	Conditions	Results
1	sat. oxalic acid, THF, Δ, 9 h	SM was still present, aldehyde was disappearing
2	sat. oxalic acid, MeOH, rt, 4 h	SM was still present, aldehyde was disappearing
3	10% HCl, THF, rt	No aldehyde or SM present → decomposition
4	10% H_2SO_4, THF, rt	No aldehyde or SM present → decomposition
5	TsOH, acetone, rt	No aldehyde or SM present → decomposition
6	50%, CF_3COOH, $CHCl_3$, H_2O, 0 °C	No aldehyde or SM present → decomposition

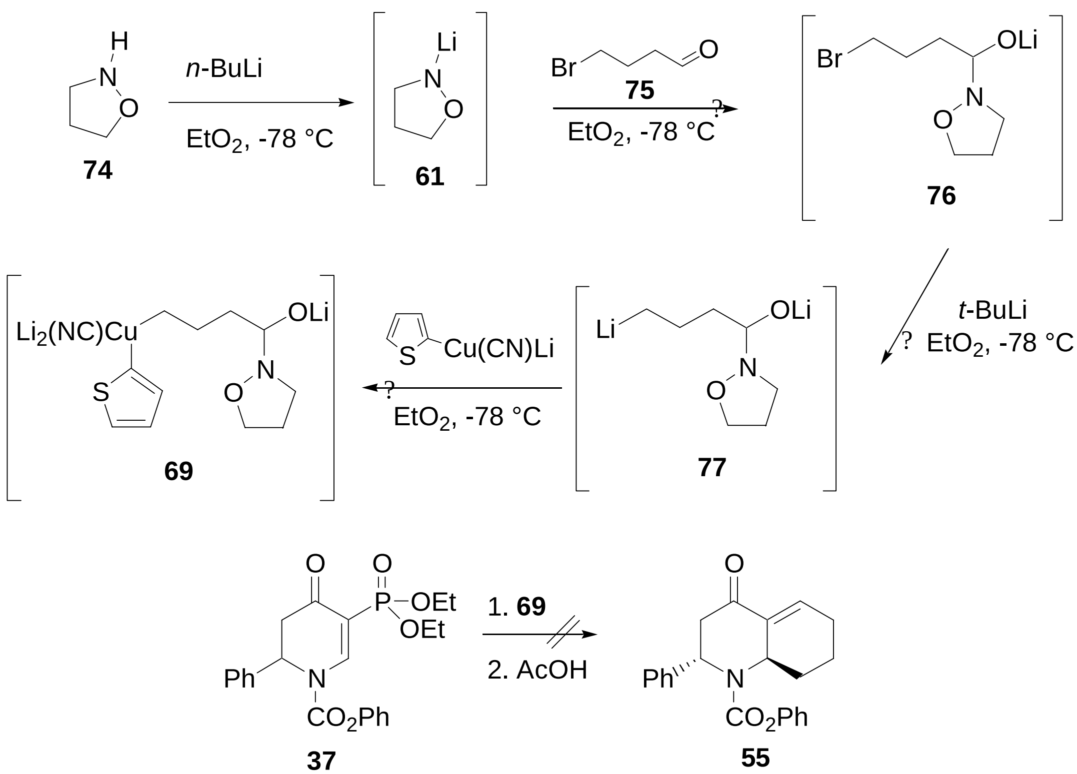
Since this route failed to generate the desired aldehyde, we tried a different approach. We decided to obtain the iodoalcohol and oxidize it to acquire the corresponding iodoaldehyde. Using a procedure developed by Helquist, 4-iodo-1-butanol (**73**) was synthesized by treating THF with TMS-I followed by hydrolysis.[47] The alcohol was then oxidized using pyridinium chlorochromate (PCC), or a Swern oxidation, to give the proper aldehyde (Scheme 31). Unfortunately, isolation of the aldehyde proved to be impossible. Purification by distillation and flash chromatography, containing silica gel or neutral alumina, led to significant decomposition with no recovery of the starting material. The iodoaldehyde was too unstable for it to be obtained in its pure form. Therefore, we chose the less reactive bromoaldehyde for our study.

Scheme 31: Oxidation of 4-Iodo-1-butanol



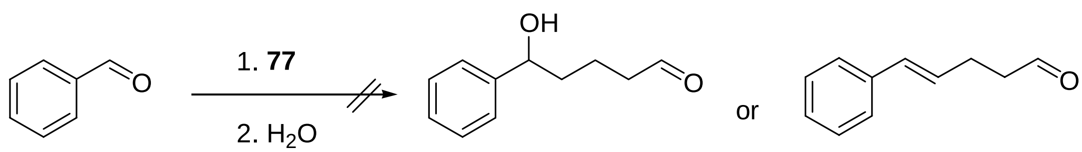
4-Bromobutanal (**75**) was made by oxidizing commercially available 4-bromo-1-butanol with PCC according to Vedejs's procedure.[48] With the aldehyde **75** in hand, the one-pot cyclization was attempted with phosphonate **37** and higher order cuprate **69** (Scheme 32). 4-Bromobutanal (**75**) was added to the lithiated form of isoxazolidine (**61**) at -78 °C in ether. The subsequent step involved a lithium-halogen exchange with the bromide, and then addition of the Lipshutz reagent to obtain higher order cuprate **69**. The cuprate was finally added to the electrophile **37**. After 10 minutes, acetic acid was added to the reaction mixture.

Scheme 32: Attempted One-Pot Cyclization



The crude NMR spectrum, after addition of all reagents, showed that the phosphonate **37** remained present. The bromoaldehyde **75** and isoxazolidine (**74**) were completely consumed in the sequence of reactions. These findings suggested that the higher order cuprate probably did not form, and the problem was within its synthesis. To determine if the steps leading to the lithium halogen-exchange were successful, we added **77** to benzaldehyde (**78**) hoping to obtain either **79** or **80** (Scheme 33). Regrettably, the crude NMR spectrum showed that benzaldehyde remained unreactive, and the rest of the NMR spectrum showed significant decomposition. This observation suggested it was difficult to form **76** or **77**. Due to the success with the other route and the difficulty presented in this synthetic plan, we decided to abandon this approach.

Scheme 33: Attempted Addition to Benzaldehyde



78 **79** **80**

Conclusion

The nickel-catalyzed cross-coupling reaction with triethyl phosphite allows a vinyl phosphonate group to be introduced with near quantitative conversion. The reaction involving the cross-coupling of vinyl iodide **36** with nickel chloride and methyl diphenylphosphinite to afford vinyl phosphine oxide **40** is a transformation not found in current literature.

Conjugate addition to *N*-acyl-5-(diethoxy-phosphoryl)-2,3-dihydro-4-pyridones is possible by the use of either higher order cuprates or copper-assisted Grignard reagents. An intermolecular Horner-Wadsworth-Emmons olefination reaction allows olefins to be introduced at the C-5 position of *N*-Acyl-2,3-dihydro-4-pyridones in moderate to high yields with preference for the (*E*)-alkene. Use of an intramolecular Horner-Wadsworth-Emmons olefination induces cyclization of 6-substituted-3-(diethoxy-phosphoryl)-2-(4-oxo-butyl)-*N*-acylpiperidin-4-ones to give a *trans* bicyclic enone.

Experimental Section

All reactions were performed in oven-dried glassware under argon atmosphere. Tetrahydrofuran (THF) and toluene were distilled from sodium/benzophenone ketyl prior to use. Other solvents and reagents purchased from commercial sources were stored under argon and used directly. *n*-Butyllithium and *tert*-butyllithium were titrated periodically against diphenylacetic acid according to the procedure of Kofron and Baclanski.[49] Melting points were obtained using a Thomas-Hoover capillary melting point apparatus and are uncorrected. Radial preparative layer chromatography (radial PLC) was performed on a model 7924T chromatotron (Harrison Research, Palo Alto, CA) using glass plates coated with 1-, 2-, or 4-mm thicknesses of Kieselgel 60 PF_{254} containing gypsum. Elemental analyses were performed by Atlantic Microlab, Inc. (Norcross, GA). NMR spectra were obtained on either a Varian XL-300 or GE GN 300 spectrometer. IR spectra were collected on a Perkin-Elmer 7500 spectrometer. High resolution mass spectra were obtained using a JEOL HX110HF mass spectrometer. diastereomeric excess was determined by HPLC using a Waters and Assoc. (Miliford, MA) 600 E multi solvent delivery system/486 tunable detector equipped with a μ-PORAIL analytical column.

5-(Diethoxyphosphoryl)-1-(phenoxycarbonyl)-2-phenyl-2,3-dihydro-4-pyridone (37). A mixture of 5-iodo-1-(phenoxycarbonyl)-2-phenyl-2,3-dihydro-4-pyridone (**36**) (2.00 g, 4.77 mmol), triethyl phosphite (10 mL, 85.0 mmol), and $NiCl_2$ (0.050 g, 0.477 mmol) was heated to 150 °C for 6 h with stirring. The excess triethyl phosphite was removed by distillation, and the remaining liquid was purified by radial PLC (silica gel, 75% EtOAc/hexanes) to give 1.98 g (97%) of **37** as a white solid: mp 134-135 °C; IR ($CHCl_3$) 2985, 1750, 1681, 1584, 1301, 1243, 1024 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ 1.25 (t, 3H, J = 6.6 Hz), 1.33 (t, 3H, J = 6.6 Hz), 2.95 (d, 1H, J = 16.5 Hz), 3.27 (dd, 1H, J = 7.3 and 17.1 Hz), 4.10 (m, 4H), 5.90 (d, 1H, J = 7.2 Hz), 7.25 (m, 10H), 8.92 (d, 1H, J = 14.7 Hz); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 16.9, 17.0, 17.1, 42.8, 42.9, 57.5, 63.1, 63.1, 63.2, 63.3, 107.2, 109.7, 121.8, 126.4, 127.4, 129.3, 129.8, 130.4, 137.9, 150.8, 151.6, 152.2, 152.5, 189.7. Anal. Calcd. for $C_{22}H_{24}NO_6P$: C, 61.54; H, 5.63; N, 3.26. Found: C, 61.61; H, 5.74; N, 3.23.

5-(Diethoxyphosphoryl)-1-(phenoxycarbonyl)-2-isopropyl-2,3-dihydro-4-pyridone (39). A mixture of 5-iodo-1-(phenoxycarbonyl)-2-isopropyl-2,3-dihydro-4-pyridone (**38**) (1.62 g, 4.21 mmol), triethyl phosphite (8.92 mL, 75.8 mmol), and $NiCl_2$ (0.0546 g, 0.421 mmol) was heated to 150 °C for 6 h with stirring. The excess triethyl phosphite was removed by distillation and the remaining liquid was purified by radial PLC (silica gel, 75% EtOAc/hexanes) to give 1.45 g (87%) of **39** as a white solid: mp 91-92 °C; IR ($CHCl_3$) 2966, 1749, 1678, 1584, 1308, 1249, 1196, 1026, 961, 750 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ 1.00 (t, 6H, J = 5.73 Hz), 1.34 (q, 6H, J = 6.8 Hz), 2.16 (m, 1H), 2.82 (m, 2H), 4.16 (m, 4H), 4.52 (t, 1H, *J* = 7.2 Hz), 7.30 (m, 5H), 8.77 (d, 1H, *J* = 14.6 Hz); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 6.5, 18.9, 19.7, 29.9, 38.4, 59.7, 62.7, 121.3, 126.9, 129.9, 150.5, 151.5, 190.3. Anal. Calcd. for $C_{19}H_{26}NO_6P$: C, 57.72; H, 6.63; N, 3.54. Found: C, 57.88; H, 6.70; N, 3.53.

5-(Diphenylphosphinoyl)-1-(phenoxycarbonyl)-2-phenyl-2,3-dihydro-4-pyridone (40). To a stirred solution of 5-iodo-1-(phenoxycarbonyl)-2-phenyl-2,3-dihydro-4-pyridone (**36**) (1.33 g, 3.18 mmol) in toluene (50 mL) was added methyl diphenylphosphinite (2.55 mL, 12.7 mmol) and $NiCl_2$ (0.0206 g, 0.159 mmol). The mixture was heated to reflux for 30 min with stirring. The reaction was allowed to cool and then concentrated under reduced pressure. Purification by radial PLC (silica gel, 75% EtOAc/hexanes), followed by recrystallization with seeding, gave 1.27 g (81%) of **40** as a white solid: mp 219-220 °C; IR ($CHCl_3$) 3060, 2249, 1749, 1672, 1572, 1296, 1243, 1190, 908, 732 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ 2.89 (d, 1H, *J* = 16.4 Hz), 3.24 (dd, 1H, *J* = 7.1 and 16.3 Hz), 5.94 (d, 1H, *J* = 6.7 Hz), 7.11-7.84 (m, 20H), 9.13 (d, 1H, 12.4 Hz); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 42.5, 56.9, 110.5, 112.0, 121.3, 126.0, 128.1, 128.2, 128.3, 128.4, 128.5, 128.7, 129.2, 129.8, 131.2, 131.7, 131.8, 132.1, 132.2, 132.6, 133.6, 137.3, 150.4, 151.0, 151.9, 152.1, 152.1, 190.1. Anal. Calcd. for $C_{30}H_{24}NO_4P$: C, 73.01; H, 4.90; N, 2.84. Found: C, 72.97; H, 4.92; N, 2.94.

5-(Diethoxyphosphoryl)- 1-(phenoxycarbonyl)-2-phenylpiperidin-4-one (41). To a stirred solution of **37** (115 mg, 0.268 mmol) in THF (15 mL) at -78 °C was added K-Selectride® (0.29 mL, 0.299 mmol, 1.0 M solution in THF). The reaction was stirred for 30 min and then quenched with saturated aqueous $NaHCO_3$. The reaction was allowed to warm to room temperature and then extracted with ethyl acetate. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by PLC (silica gel, 50% EtOAc/hexanes) to give 85 mg (74%) of **41** as a colorless oil: IR ($CHCl_3$) 3436, 3013, 2249, 1819, 1796, 1713, 1643, 1467, 1420, 1384, 1214, 908, 744 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ 1.07 (t, 3H, *J* = 7.2 Hz), 1.34 (t, 3H, *J* = 6.3 Hz), 2.82-3.63 (m, 2H), 3.85 (m, 1H), 4.17 (m, 4H), 4.43 (m, 1H), 4.80 (m, 1H), 5.90 (d, 1H, *J* = 7.2 Hz), 6.05 (br s, 1H), 7.13-7.42 (m, 10H), 10.99 (s, 1H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 15.7, 16.1, 37.7, 37.9, 39.3, 40.9, 49.1, 50.8, 61.6, 61.7, 62.0, 62.4, 62.7, 62.9, 76.6, 85.2, 87.6, 121.4, 125.2, 125.3, 125.4, 125.6, 126.3, 126.5, 126.7, 127.5, 127.8, 128.5, 128.7, 128.9, 129.0, 129.1, 138.3, 151.1, 153.6, 201.6. HRMS Calcd. for $C_{22}H_{26}NO_6P$: 432.1576 $[M+H]^+$. Found: 432.1581 $[M+H]^+$.

5-(Diethoxyphosphoryl)-2-isopropyl-1-(phenoxycarbonyl)piperidin-4-one (42). To a stirred solution of **39** (508 mg, 1.29 mmol) in THF (30 mL) at -78 °C was added K-Selectride® (1.42 mL, 1.41 mmol, 1.0 M solution in THF). The reaction was stirred for 30 min and then quenched with saturated aqueous $NaHCO_3$. The reaction was allowed to warm to room temperature and then extracted with ethyl acetate. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 50% EtOAc/hexanes) to give 436 mg (85%) of **42** as a colorless oil: IR ($CHCl_3$) 2968, 1715, 1639, 1592, 1417, 1242, 1202, 1160, 1021, 968, 747 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ 1.02 (d, 6H, J = 6.3 Hz), 1.35 (t, 6H, J = 5.7 Hz), 1.78 (br s, 1H), 2.04 (br s, 1H), 2.38 (br s, 1H), 2.44 (br s, 1H), 2.65 (d, 2H, J = 14.1 Hz), 2.94 (dd, 1H, J = 4.3 and 24.0 Hz), 3.06 (m, 1H), 3.22 (m, 1H), 4.15 (m, 4H), 4.44 (m, 1H), 4.68 (dd, 1H, J = 6.2 and 14.2 Hz), 4.97 (q, 1H, J = 15.9 Hz), 7.08-7.39 (m, 5H), 10.86 (s, 1H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 16.5, 18.4, 19.1, 19.7, 20.3, 29.0, 30.8, 38.0, 39.7, 40.9, 42.2, 42.7, 49.4, 51.1, 55.5, 56.4, 58.9, 59.8, 62.5, 63.1, 63.3, 121.8, 125.5, 129.5, 151.6, 154.1, 202.5. HRMS Calcd. for $C_{19}H_{28}NO_6P$: 398.1733 $[M+H]^+$. Found: 398.1729 $[M+H]^+$.

5-Benzylidene-1-(phenoxycarbonyl)-2-phenylpiperidin-4-one (45). To a stirred solution of **41** (110 mg, 0.255 mmol) in THF (5 mL) was added potassium *tert*-butoxide (0.26 mL, 0.255 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. Benzaldehyde (0.03 mL, 0.255 mmol) was then added dropwise and the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 90 mg (92%) of an inseparable mixture of geometric isomers as a yellow oil: IR ($CHCl_3$) 3017, 1713, 1622, 1494, 1403, 1371, 1323, 1210, 756 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 3.16 (d, 2H, J = 6.0 Hz), 4.80 (br s, 1H), 5.31 (d, 1H, J = 17.1 Hz), 5.78 (br s, 1H), 6.82-7.70 (m, 16H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 30.0, 42.7, 49.9, 104.4, 121.8, 125.7, 126.2, 128.1, 129.0, 129.2, 129.5, 129.9, 130.7, 134.1, 137.2, 151.2, 154.2, 196.2. HRMS Calcd. for $C_{25}H_{21}NO_3$: 384.1600 $[M+H]^+$. Found: 384.1610 $[M+H]^+$; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 4.79, 5.38 min) determined the ratio of *E* : *Z* isomers to be 8.3 : 1.

1-(Phenoxycarbonyl)-2-phenyl-5-(3-phenylpropylidene)piperidin-4-one (46). To a stirred solution of **41** (68 mg, 0.136 mmol) in THF (5 mL) was added potassium *tert*-butoxide (0.14 mL, 0.136 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. Hydrocinnamaldehyde (0.02 mL, 0.136 mmol) was added dropwise and then the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 42 mg (74%) of an inseparable mixture of geometric isomers as a colorless oil: IR ($CHCl_3$) 3023, 2925, 2849, 2361, 1719, 1697, 1491, 1453, 1393, 1198 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 2.50 (m, 2H), 2.75 (m, 2H), 3.00 (d, 2H, J = 20.8 Hz), 3.99 (br s, 1H), 5.70 (br s, 1H), 6.87 (t, 1H, J = 7.5 Hz), 6.94-7.69 (m, 15H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 29.9, 30.9, 34.6, 35.2, 41.4, 48.1, 121.8, 125.7, 125.8, 125.9, 126.0, 126.1, 16.3, 126.5, 127.8, 128.9, 128.0, 128.6, 128.6, 128.7, 129.1, 129.1, 129.3, 129.5, 140.0, 140.6, 151.3, 154.2, 195.4. HRMS Calcd.

for $C_{25}H_{21}NO_3$: 412.1913 $[M+H]^+$. Found: 412.1904 $[M+H]^+$; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 5.08, 5.92 min) determined the ratio of *E* : *Z* isomers to be 3 : 1.

2-Phenyl-5-(3-phenylallylidene)-1-(phenoxycarbonyl)piperidin-4-one (47). To a stirred solution of **41** (111 mg, 0.256 mmol) in THF (7 mL) was added potassium *tert*-butoxide (0.25 mL, 0.256 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. *Trans*-cinnamaldehyde (0.03 mL, 0.256 mmol) was then added dropwise and the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 67 mg (64%) of an inseparable mixture of geometric isomers as a yellow oil: IR ($CHCl_3$) 3022, 2921, 2863, 1715, 1687, 1602, 1579, 1398, 1201 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 1.27 (m, 2H), 3.12 (d, 2H, J = 4.0 Hz), 4.40 (br s, 1H), 5.17 (d, 1H, J = 16.4 Hz), 5.79 (br s, 1H), 6.89-7.56 (m, 16H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 14.7, 29.9, 41.9, 48.2, 121.9, 125.7, 125.8, 126.1, 126.3, 126.3, 126.4, 126.5, 126.8, 127.8, 127.9, 128.1, 128.3, 128.8, 129.0, 129.1, 129.2, 129.6, 129.9, 136.1, 136.4, 144.3, 154.3, 195.4. HRMS Calcd. for $C_{27}H_{23}NO_3$: 410.1756 $[M+H]^+$. Found: 410.1762 $[M+H]^+$; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 4.84, 5.58 min) determined the ratio of *E* : *Z* isomers to be 3.1 : 1.

5-Hexylidene-1-(phenoxycarbonyl)-2-phenylpiperidin-4-one (48). To a stirred solution of **41** (72 mg, 0.166 mmol) in THF (5 mL) was added potassium *tert*-butoxide (0.17 mL, 0.166 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. Hexanal (0.02 mL, 0.166 mmol) was then added dropwise, and the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 50 mg (80%) of an inseparable mixture of geometric isomers as a colorless oil: IR ($CHCl_3$) 3156, 2956, 2923, 2856, 1711, 1628, 1456, 1394, 1200, 906, 733, 646 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 0.87 (t, 3H, *J* = 6.8 Hz), 1.36 (m, 5H), 2.17 (t, 2H, *J* = 6.9 Hz), 2.60 (m, 1H), 3.07 (d, 2H, *J* = 5.4 Hz), 4.16 (br s, 1H), 4.94 (d, 1H, *J* = 16.1 Hz), 6.09 (br s, 1H), 6.85 (t, 1H, *J* = 7.7 Hz), 7.00-7.36 (m, 10H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 14.1, 22.6, 28.1, 29.0, 29.4, 29.9, 31.7, 41.6, 54.9, 121.8, 125.7, 126.0, 127.8, 129.1, 129.5, 130.7, 131.4, 141.8, 151.3, 154.2, 195.6. HRMS Calcd. for $C_{24}H_{27}NO_3$: 378.2069 [M+H]. Found: 378.2077 [M+H]; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 3.50, 4.24 min) determined the ratio of *E* : *Z* isomers to be 8.8 : 1.

5-Benzylidene-2-isopropyl-1-(phenoxycarbonyl)piperidin-4-one (49). To a stirred solution of **42** (148 mg, 0.371 mmol) in THF (5 mL) was added potassium *tert*-butoxide (0.37 mL, 0.371 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. Benzaldehyde (0.04 mL, 0.371 mmol) was then added dropwise, and the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 109 mg (84%) of an inseparable mixture of geometric isomers as a yellow oil: IR ($CHCl_3$) 2959, 2873, 1715, 1593, 1491, 1411, 1197 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 1.04 (m, 6H), 1.97 (m, 1H), 2.84 (m, 2H), 4.33 (d, 2H, J = 15.8 Hz), 5.33 (d, 1H, J = 17.1 Hz), 7.05-7.68 (m, 11H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 18.9, 19.2, 19.3, 30.7, 41.4, 42.7, 56.6, 57.2, 121.7, 125.6, 128.2, 128.9, 129.4, 129.8, 134.2, 136.5, 136.8, 151.3, 154.3, 196.6. HRMS Calcd. for $C_{22}H_{23}NO_3$: 350.1756 [M+H]. Found: 350.1751 [M+H]; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 4.79, 5.29 min) determined the ratio of *E* : *Z* isomers to be 11.6 : 1.

2-Isopropyl-5-(3-phenylpropylidene)-1-(phenoxycarbonyl)piperidin-4-one (50). To a stirred solution of **42** (97 mg, 0.245 mmol) in THF (5 mL) was added potassium *tert*-butoxide (0.24 mL, 0.245 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. Hydrocinnamaldehyde (0.03 mL, 0.245 mmol) was then added dropwise, and the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 69 mg (74%) of an inseparable mixture of geometric isomers as a colorless oil: IR ($CHCl_3$) 2931, 1713, 1690, 1614, 1414, 1255, 1202 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 0.96 (t, 6H, *J* = 6.3 Hz), 1.7 (m, 1H), 2.50 (q, 2H, *J* = 6.5 Hz), 2.80 (m, 4H), 3.71 (d, 1H, *J* = 16.9 Hz), 4.21 (m, 1H), 4.83 (d, 1H, *J* = 16.9 Hz), 6.87 (t, 1H, *J* = 7.6 Hz), 7.07-7.40 (m, 10H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 19.2, 19.7, 19.9, 30.0, 30.5, 34.6, 41.2, 41.5, 56.8, 57.4, 121.8, 125.7, 126.5, 128.6, 128.7, 129.5, 129.8, 131.5, 139.6, 140.1, 140.8, 151.4, 154.4, 195.7. HRMS Calcd. for $C_{24}H_{27}NO_3$: 378.2069 [M+H]. Found: 378.2074 [M+H]; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 3.96, 4.42 min) determined the ratio of *E* : *Z* isomers to be 1.2 : 1.

2-Isopropyl-5-(3-phenylallylidene)-1-(phenoxycarbonyl)piperidin-4-one (51). To a stirred solution of **42** (124 mg, 0.311 mmol) in THF (7 mL) was added potassium *tert*-butoxide (0.31 mL, 0.311 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. *Trans*-cinnamaldehyde (0.04 mL, 0.311 mmol) was then added dropwise, and the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 71 mg (61%) of an inseparable mixture of geometric isomers as a yellow oil: IR ($CHCl_3$) 3041, 2966, 2872, 1713, 1678, 1572, 1408, 1202, 750 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 1.06 (m, 6H), 1.27 (s, 1H), 1.93 (m, 1H), 2.78 (m, 2H), 4.29 (m, 2H), 5.20 (t, 1H, *J* = 15.0 Hz), 6.53 (d, 1H, *J* = 11.3 Hz), 6.86-7.52

(m, 10H), 8.15 (m, 1H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 19.0, 19.4, 19.7, 29.9, 30.6, 56.8, 57.4, 121.8, 125.4, 127.7, 128.4, 128.8, 128.9, 129.0, 129.5, 136.1, 143.5, 144.0, 151.4, 154.4, 195.6. HRMS Calcd. for $C_{24}H_{25}NO_3$: 376.1913 $[M+H]^+$. Found: 376.1914 $[M+H]^+$; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 3.52, 4.30 min) determined the ratio of *E* : *Z* isomers to be 3.6 : 1.

5-Hexylidene-2-isopropyl-1-(phenoxycarbonyl)piperidin-4-one (52). To a stirred solution of **42** (67 mg, 0.168 mmol) in THF (5 mL) was added potassium *tert*-butoxide (0.17 mL, 0.168 mmol, 1.0 M solution in THF) at -23 °C, and the mixture was allowed to stir for 10 min. Hexanal (0.02 mL, 0.168 mmol) was then added dropwise, and the mixture was heated to 35 °C for 16 h. The reaction was quenched with brine and extracted with methylene chloride. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 44 mg (76%) of an inseparable mixture of geometric isomers as a colorless oil: IR ($CHCl_3$) 2955, 2861, 1719, 1408 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ (major isomer) 0.97 (m, 9H), 1.3 (m, 4H), 1.46 (m, 2H), 1.90 (m, 1H), 2.16 (d, 2H, J = 6.8 Hz), 2.70 (d, 2H, J = 16.2 Hz), 4.09 (m, 2H), 6.85 (t, 1H, J = 7.4 Hz), 7.09-7.40 (m, 5H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ (mixture of *E*,*Z* isomers) 1.2, 14.1, 18.8, 19.3, 19.9, 22.6, 28.0, 29.1, 29.4, 29.9, 30.8, 31.7, 32.6, 35.3, 41.5, 43.0, 47.3, 47.9, 56.9, 57.4, 57.7, 121.8, 125.7, 129.5, 130.8, 141.3, 141.9, 144.6, 145.0, 151.4, 154.4, 198.8. HRMS Calcd. for $C_{21}H_{29}NO_3$: 344.2147 [M+H]. Found: 344.2222 [M+H]; Separation of diastereomers by HPLC (Nova-Pak Silica, flow rate 1.0 mL/min, 10% EtOAc, 90% hexanes, t_R, 4.82, 5.35 min) determined the ratio of *E* : *Z* isomers to be 12.3 : 1.

(6*S,2*R**)-2-(4,4-Diethoxybutyl)-3-(diethoxy-phosphoryl)-1-(phenoxycarbonyl)-6-phenylpiperidin-4-one (53).** 1,2-Dibromoethane (0.2 mL, 2.32 mmol) was added to magnesium powder (1.36 g, 55.9 mmol) in THF (3.7 mL). 4-Chloro-1,1-diethoxybutane (1.4 mL, 6.99 mmol) was added dropwise over 10 min while refluxing. The suspension was refluxed for an additional 4 h. After cooling to rt, the prepared Grignard reagent (**63**) was transferred *via* a double-tipped stainless steel needle to a flask containing copper(I) bromide-dimethyl sulfide complex (72 mg, 3.50 mmol) in THF (5 mL) at -78 °C. In another flask, phosphonate **37** (1.25 g, 2.91 mmol) in THF (15 mL) was added dropwise to the flask containing the newly formed copper-magnesium suspension at -78 °C, and the mixture was allowed to stir for 30 min. After 45 min, the reaction was quenched with saturated aqueous ammonium chloride. The aqueous layer was extracted with diethyl ether. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. Purification by radial PLC (silica gel, 20% EtOAc/hexanes) yielded 1.408 g (84%) of the desired **53** as a colorless oil: IR ($CHCl_3$) 2964, 2917, 1712, 1630, 1495, 1448, 1383, 1342, 1242, 1207, 1160, 1052, 1021, 971 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ 1.22 (m, 6H), 1.38 (q, 6H, *J* = 7.0 Hz), 1.68 (m, 5H), 2.05 (m, 1H), 2.58 (d, 1H, *J* = 13.29 Hz), 3.16 (t, 1H, *J* = 5.8 Hz), 3.22 (dt, 1H, *J* = 5.8 and 15.8 Hz), 3.50 (m, 2H), 3.67 (q, 2H, *J* = 7.4 Hz), 4.11 (m, 4H), 4.51 (t, 1H, *J* = 5.4 Hz), 4.96 (m, 1H), 5.42 (br s, 1H), 6.55-7.41 (m, 10H), 10.92 (s, 1H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 15.5, 16.5, 21.1, 34.0, 36.1, 36.5, 52.6, 52.8, 55.9, 61.3, 62.5, 91.2, 93.7, 103.1, 121.7, 125.4, 125.6, 126.6, 127.3, 128.6, 129.2, 151.1, 168.7. HRMS Calcd. for $C_{26}H_{34}NO_6P$: 574.2570 $[M-H]^-$. Found: 574.2570 $[M-H]^-$.

3-(Diethoxyphosphoryl)-2-(4-oxo-butyl)-1-(phenoxycarbonyl)-6-phenylpiperidin-4-one (54). To a stirred solution of **53** (100 mg, 0.17 mmol) in THF (5 mL) was added saturated aqueous oxalic acid (5 mL) at rt. After 16 h, the reaction was quenched with saturated aqueous sodium bicarbonate. The aqueous layer was extracted with diethyl ether. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure to give 89 mg of the crude product. (This crude material was used directly in the next step).

(2*S,8a*R**)-2-Phenyl-3,4,6,7,8,8a-hexahydro-1-(phenoxycarbonyl)-2*H*-quinolin-4-one (55).** To a stirred solution of crude **54** (14 mg, 0.0287 mmol) in THF (5 mL) was added lithium chloride (1.2 mg, 0.0287 mmol) and triethylamine (4 μL, 0.0287 mmol) at rt. After 1.5 h, the reaction was quenched with deionized water. The aqueous layer was extracted with diethyl ether. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. Purification by radial PLC (silica gel, 20% EtOAc/hexanes) yielded 9 mg (89%) of desired **55** from crude aldehyde **54** as a white solid: mp 122-123 °C; IR ($CHCl_3$) 3031, 2919, 2867, 1724, 1694, 1620, 1490, 1420, 1266, 1196 cm^{-1}; ^1H NMR ($CDCl_3$, 300 MHz) δ 1.65 (m, 1H), 1.94 (m, 1H), 2.19-2.49 (m, 4H), 3.02 (dd, 1H, J = 5.6 and 5.9 Hz), 3.13 (dd, 1H, J = 7.6 and 7.7 Hz), 4.51 (m, 1H), 5.52 (t, 1H, J = 6.6 Hz), 6.88 (m, 2H), 7.15 (t, 1H, J = 7.3 Hz), 7.27-7.42 (m, 8H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 21.2, 25.6, 27.7, 43.1, 54.4, 55.8, 121.8, 125.6, 126.8, 127.9, 129.0, 129.4, 137.3, 138.9, 139.9, 150.9, 154.2, 197.2. Anal. Calcd. for $C_{22}H_{21}NO_3$: C, 76.06; H, 6.09; N, 4.03. Found: C, 76.06; H, 6.11; N, 3.97.

(6*S,2*R**)-2-Butyl-3-(diethoxyphosphoryl)-6-phenyl-1-(phenoxycarbonyl)piperidin-4-one (57).**

Lipshutz conditions

To a stirred solution of *n*-butyllithium (1.35 mL, 0.337 mmol, 1.5 M solution in hexanes) in THF (10 mL) was added lithium 2-thienylcyanocuprate (0.14 mL, 0.337 mmol, 0.25 solution in THF) at -78 °C, and the mixture was allowed to stir for 30 min. A stirred solution of **37** (96 mg, 0.224 mmol) in THF (5 mL) at -78 °C was transferred *via* a double-tipped stainless steel needle into the above higher order cuprate (**56**). After 3 h, the reaction was quenched with saturated aqueous sodium bicarbonate and extracted with EtOAc. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by radial PLC (silica gel, 10% EtOAc/hexanes) to give 75 mg (68%) of **57** as a colorless oil: IR ($CHCl_3$) 2953, 2857, 1718, 1633, 1387, 1339, 1205, 1018, 970 cm^{-1}; ^{1}H NMR ($CDCl_3$, 300 MHz) δ 0.94 (t, 1H, *J* = 6.7 Hz), 1.39 (m, 10H), 1.78 (m, 1H), 2.01 (m, 1H), 2.57 (d, 1H, *J* = 15.9 Hz), 3.21 (m, 1H), 4.08 (m, 4H), 4.94 (m, 1H), 5.43 (br s, 1H), 6.70-7.43 (m, 10H), 10.92 (s, 1H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 14.3, 16.5, 23.0, 28.0, 28.5, 29.3, 36.2, 45.1, 52.8, 55.9, 62.2, 62.5, 91.3, 93.8, 121.7, 125.4, 127.3, 128.5, 129.3, 143.3, 151.2, 154.3, 168.7. HRMS Calcd. for $C_{26}H_{34}NO_6P$: 488.2202 [M+H]$^+$. Found: 488.2210 [M+H]$^+$.

Copper bromide conditions

To a stirred solution of copper(I) bromide-dimethyl sulfide complex (12 mg, 0.056 mmol) in THF (5 mL) at -78 °C was added butylmagnesium chloride (0.06 mL, 0.112 mmol, 2.0 M solution in THF) for 5 min. A stirred solution of **37** (20 mg, 0.047 mmol) in THF (5 mL) at -78 °C was then transferred *via* a double-tipped stainless steel needle into the above copper-magnesium suspension. After 45 min, the reaction was quenched with saturated aqueous ammonium chloride. The aqueous layer was extracted with diethyl ether. The combined organic extracts were dried over $MgSO_4$ and concentrated under reduced pressure. Purification by radial PLC (silica gel, 20% EtOAc/hexanes) yielded 17 mg (75%) of desired **57** as a colorless oil.

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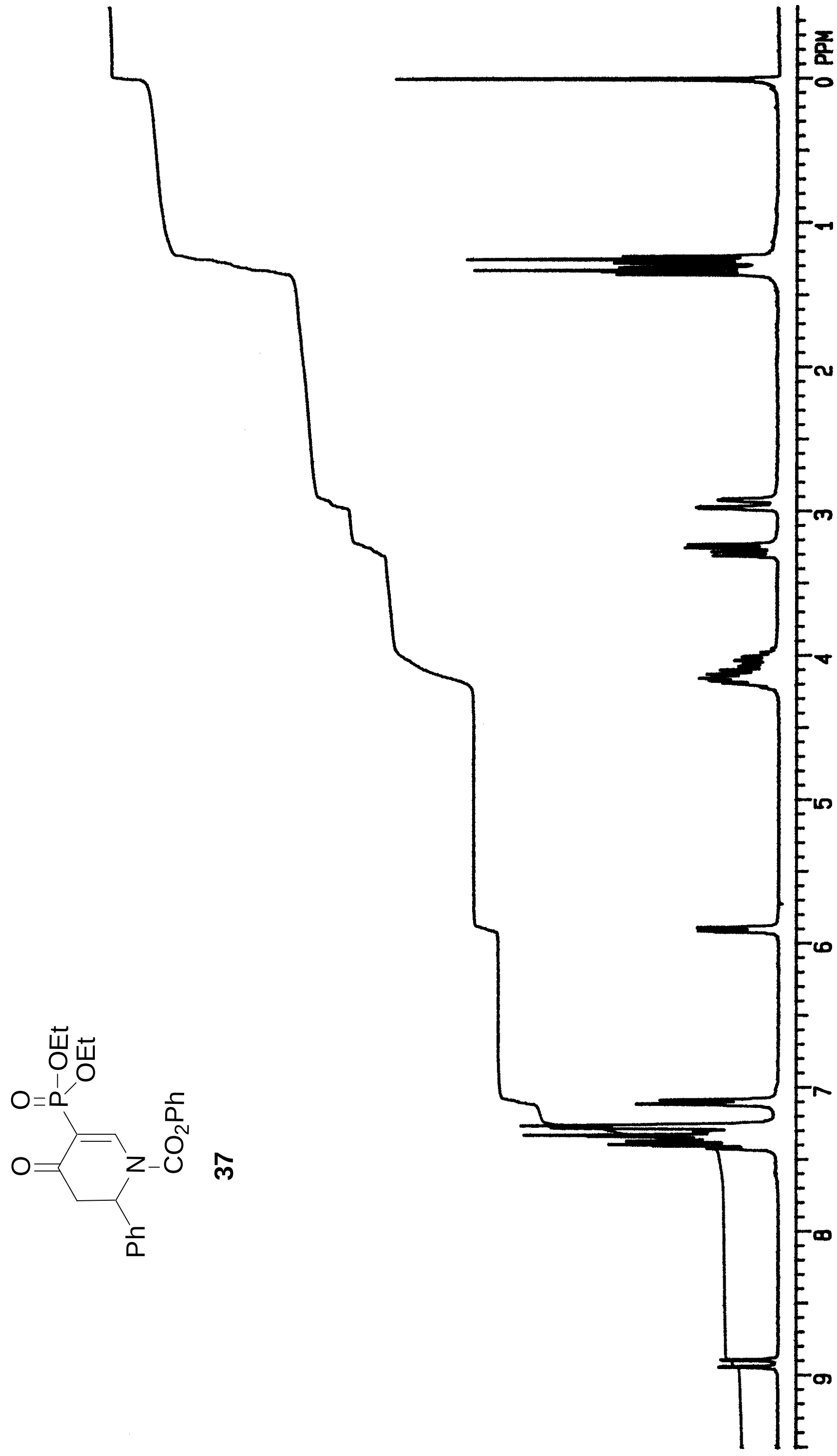
Appendices



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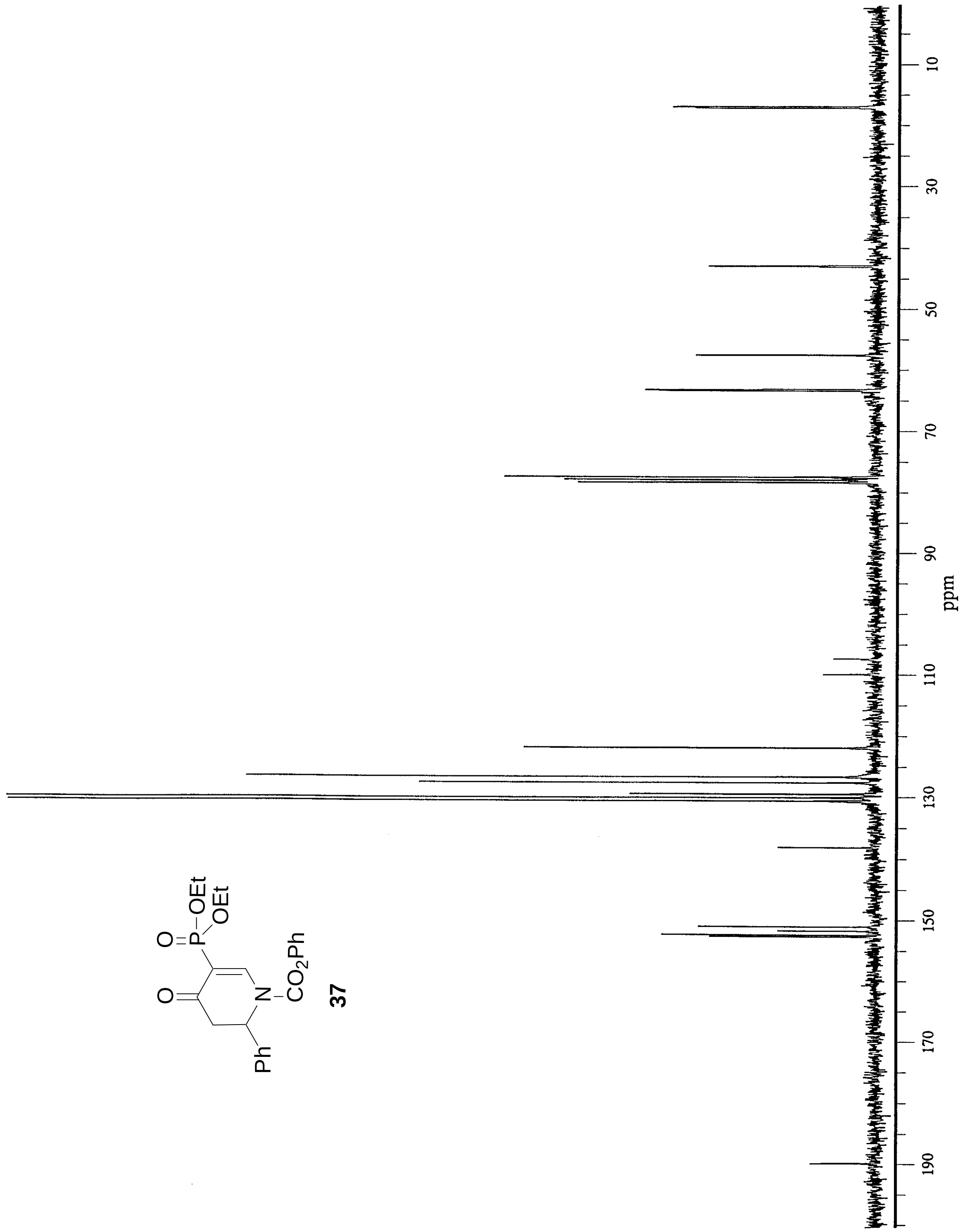

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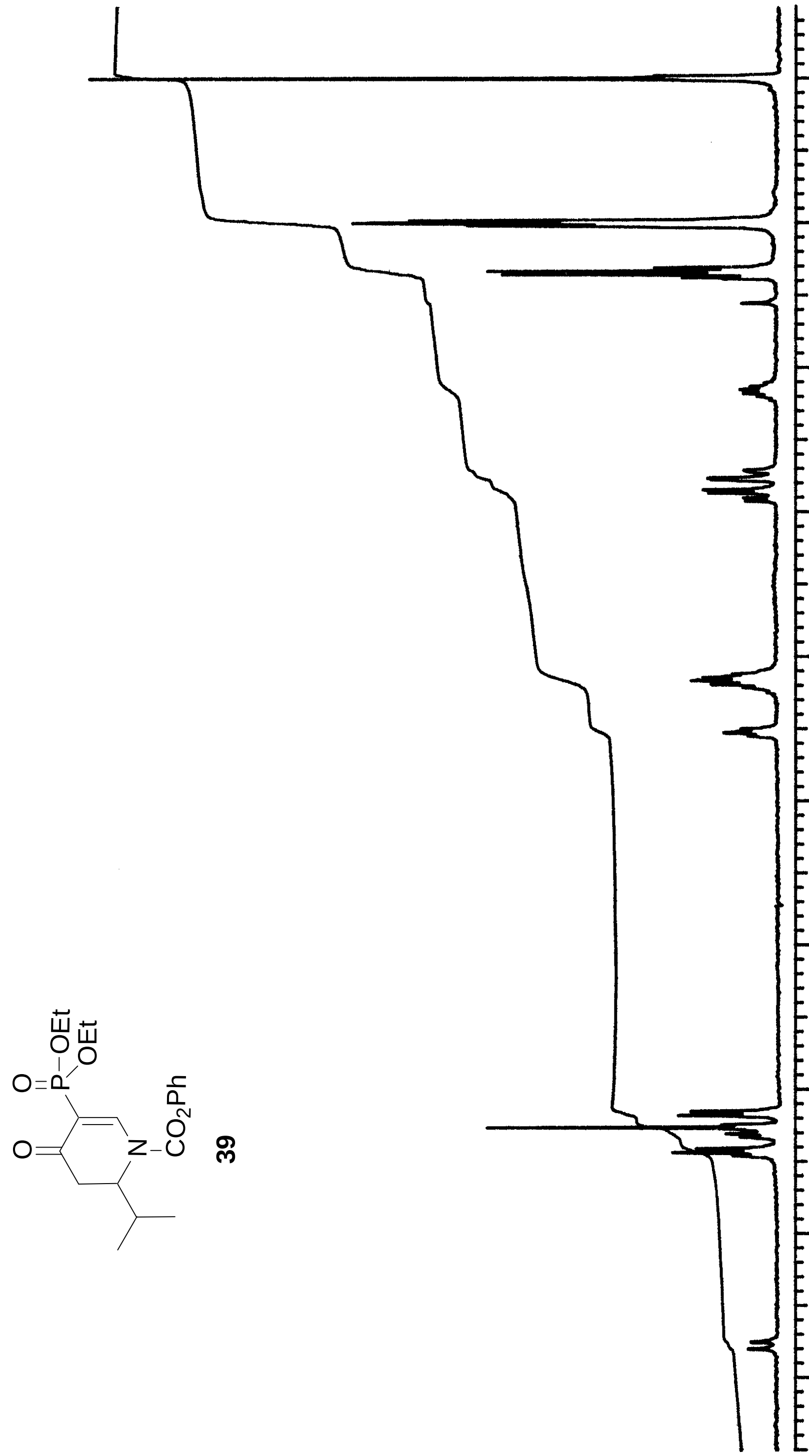

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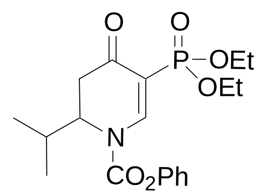

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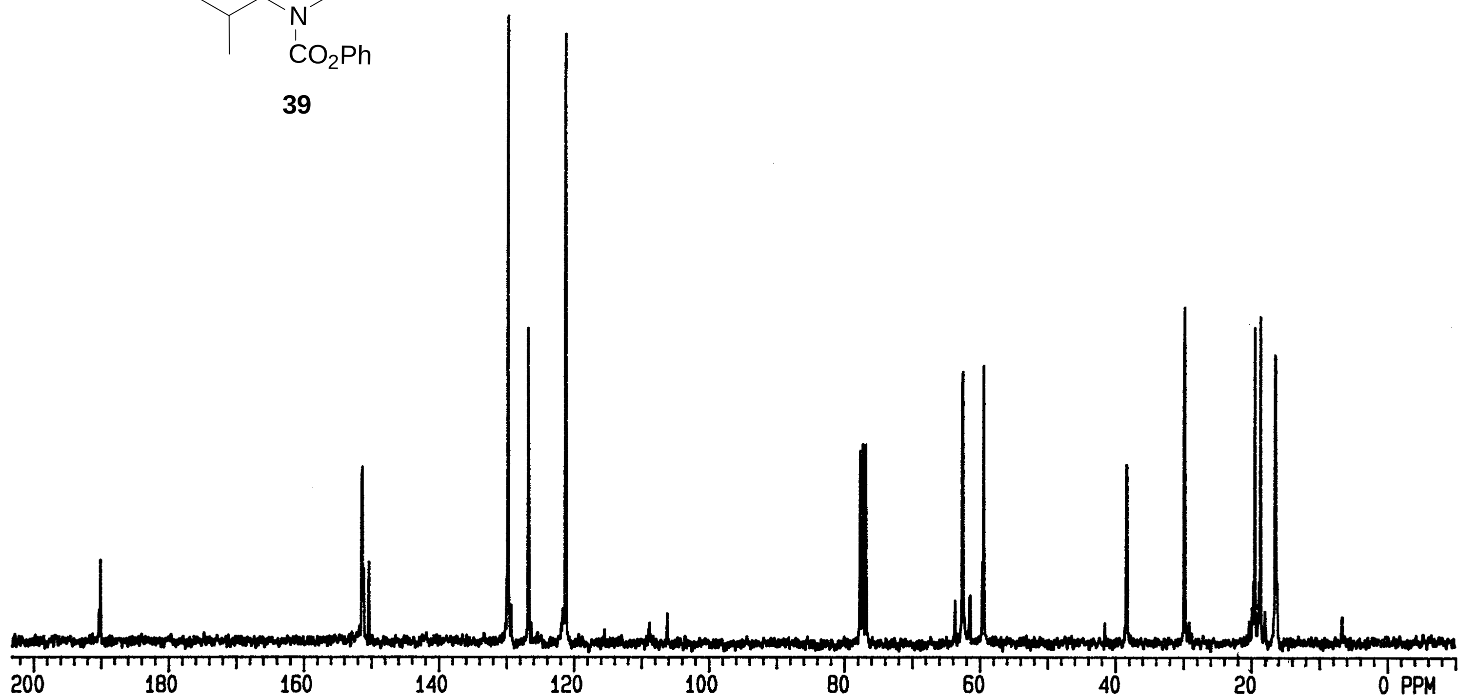

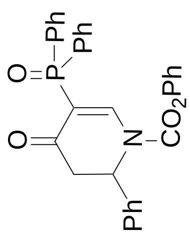

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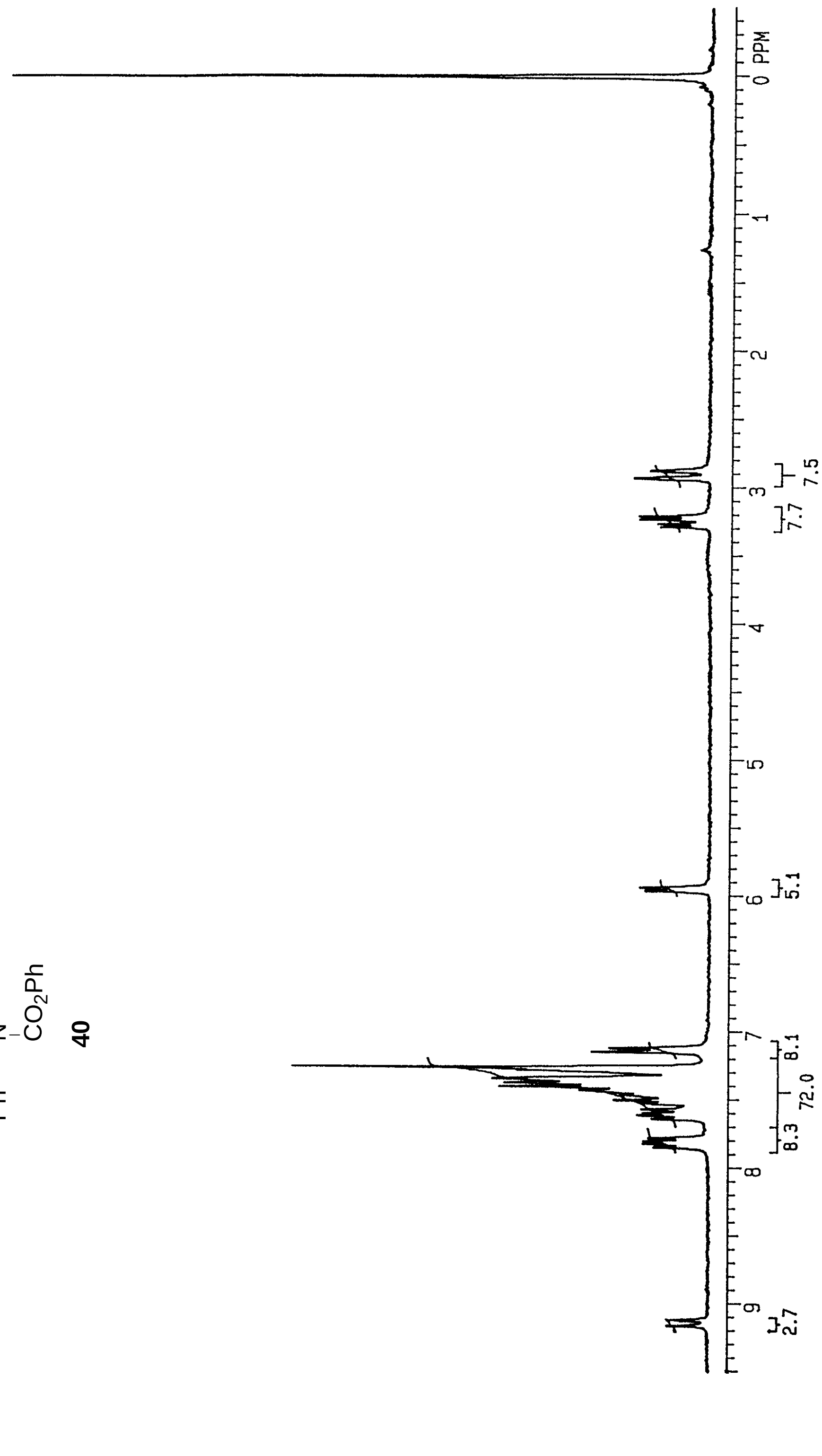

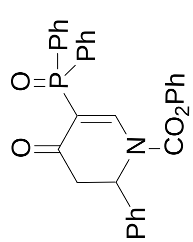

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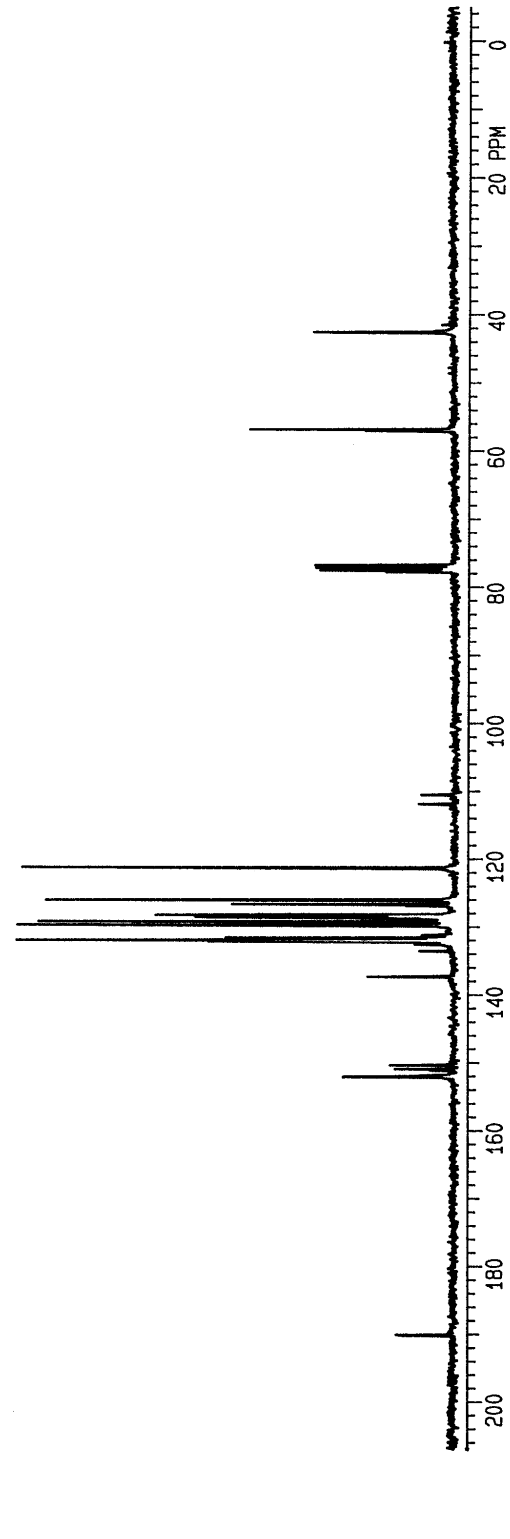

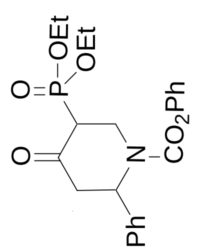

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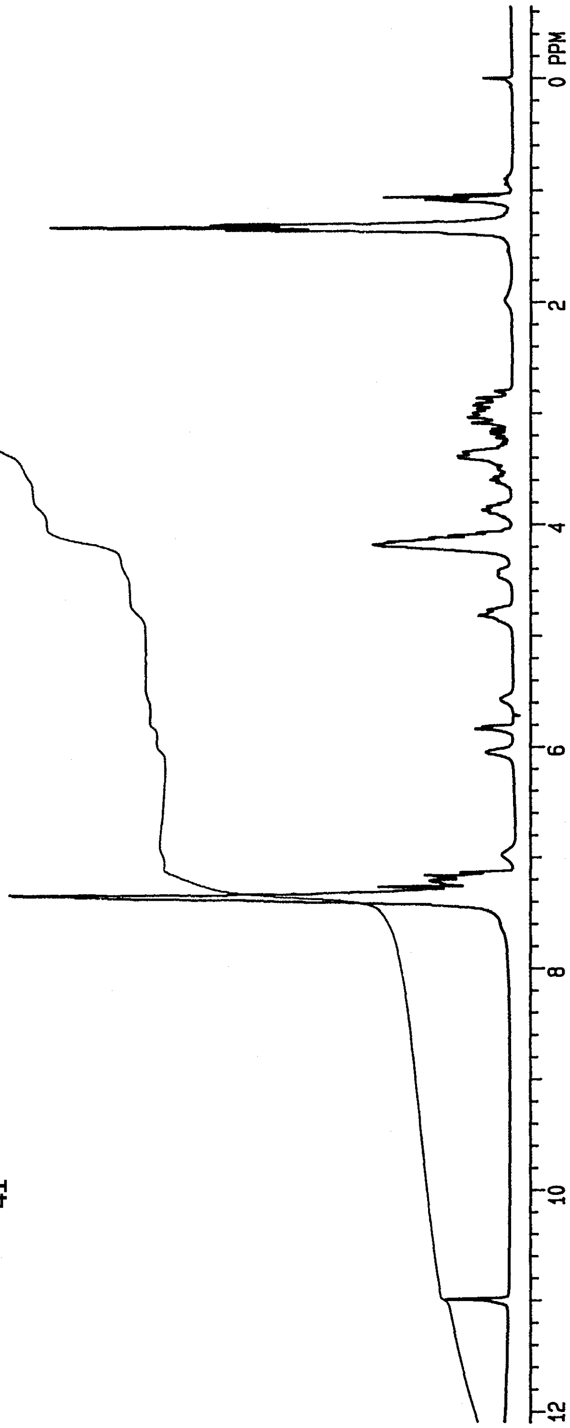

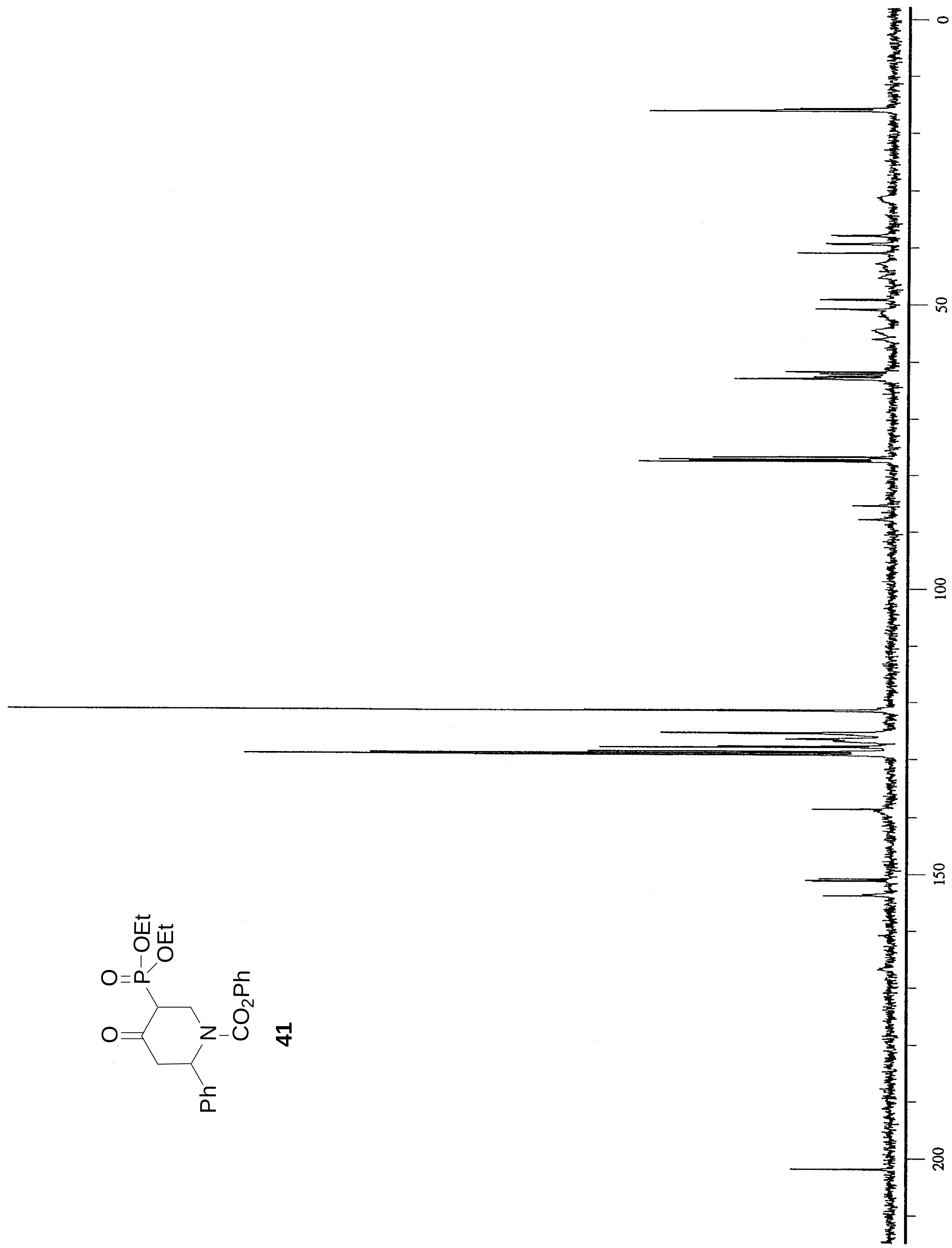

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ppm
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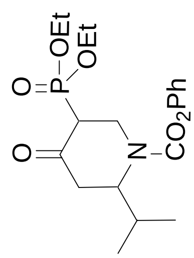

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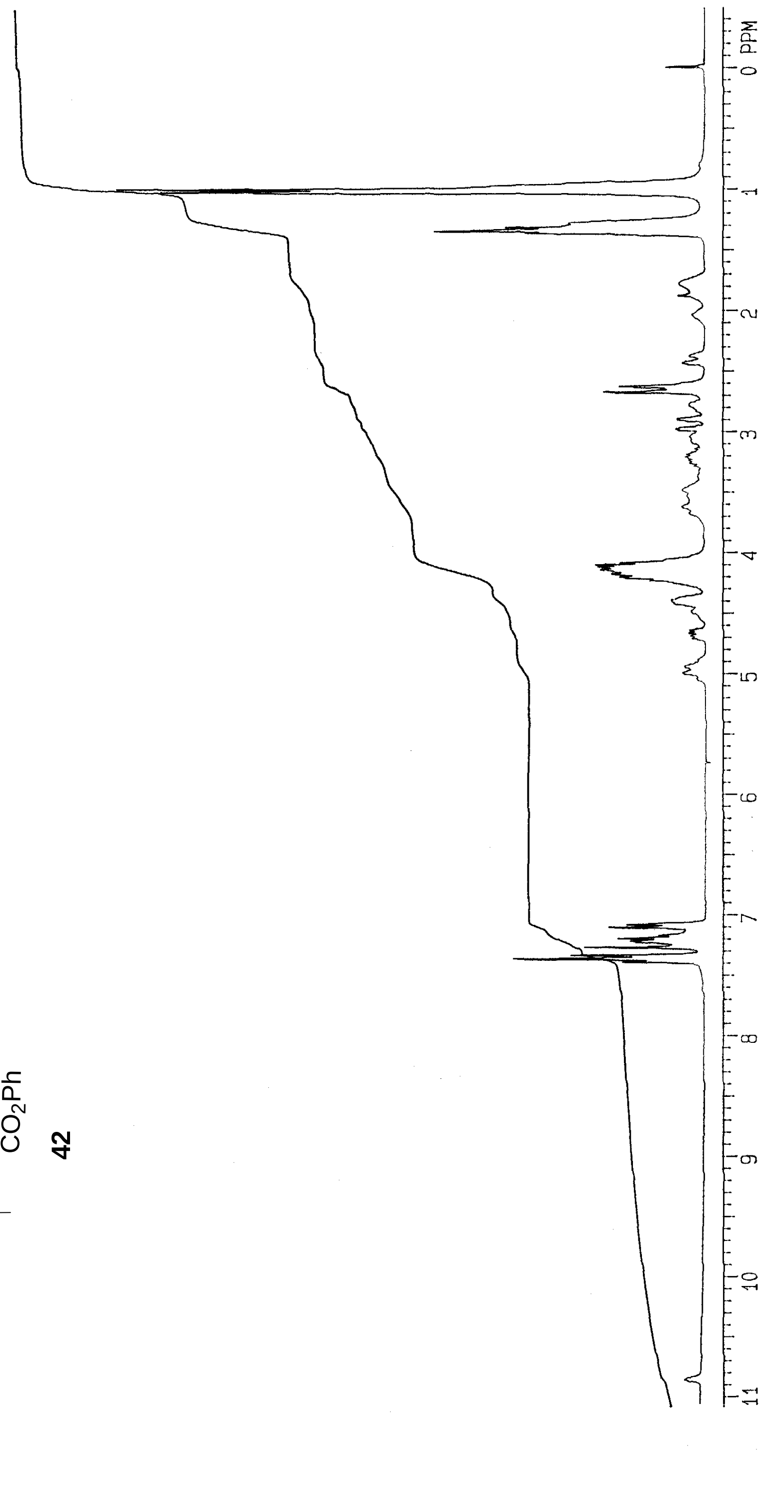

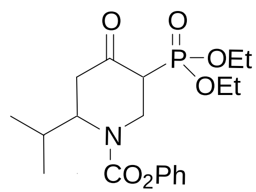

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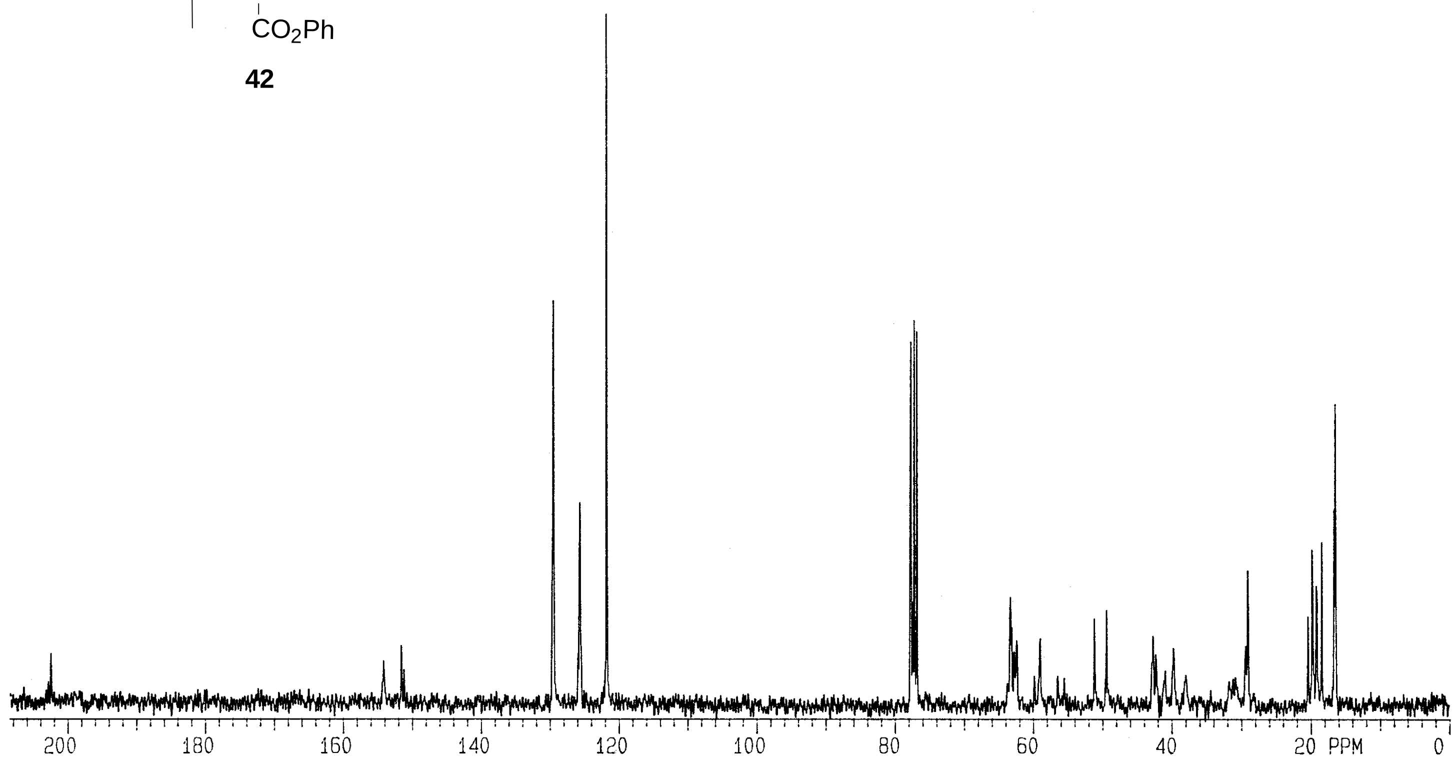

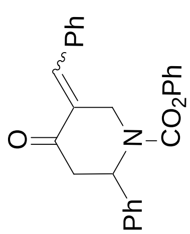

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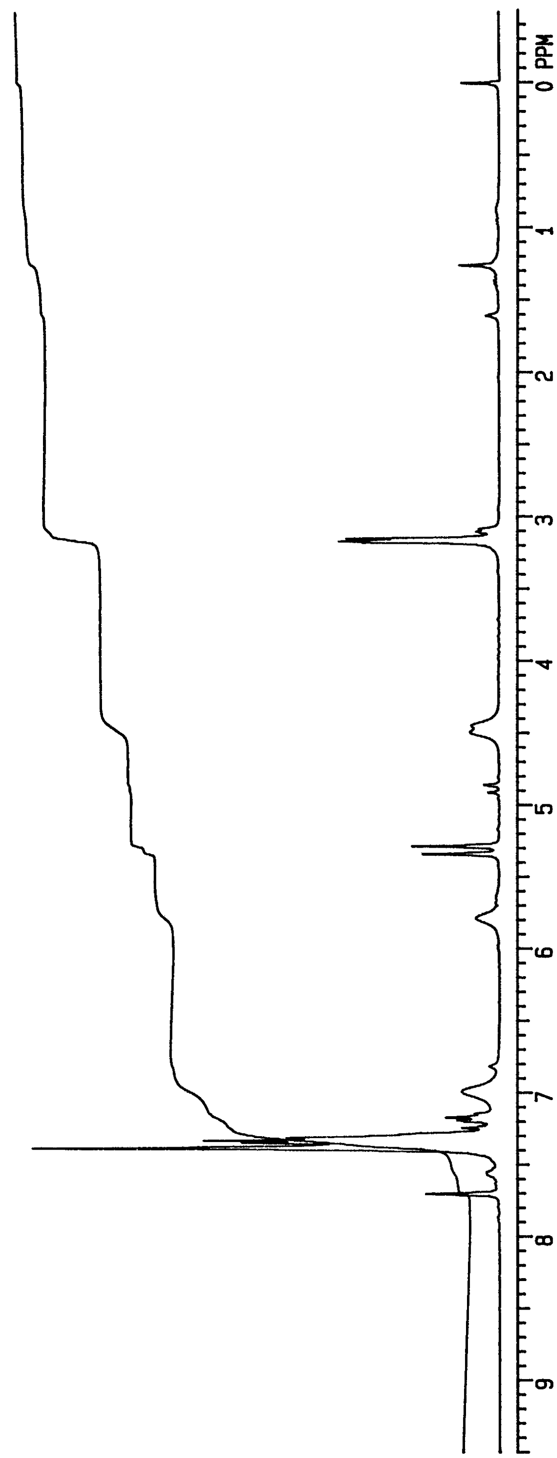

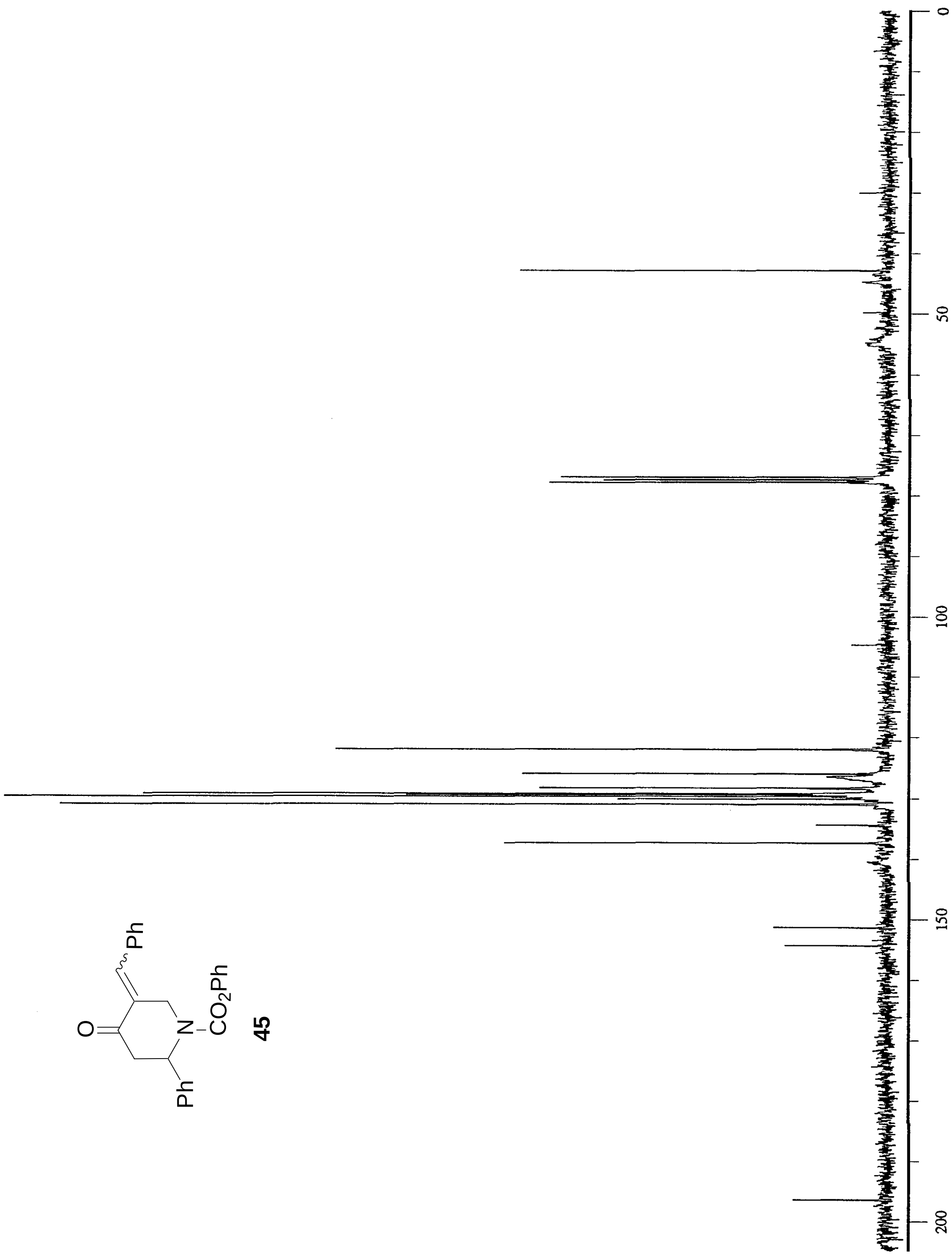

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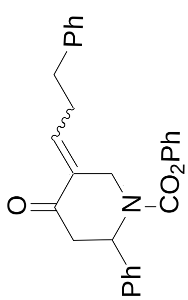

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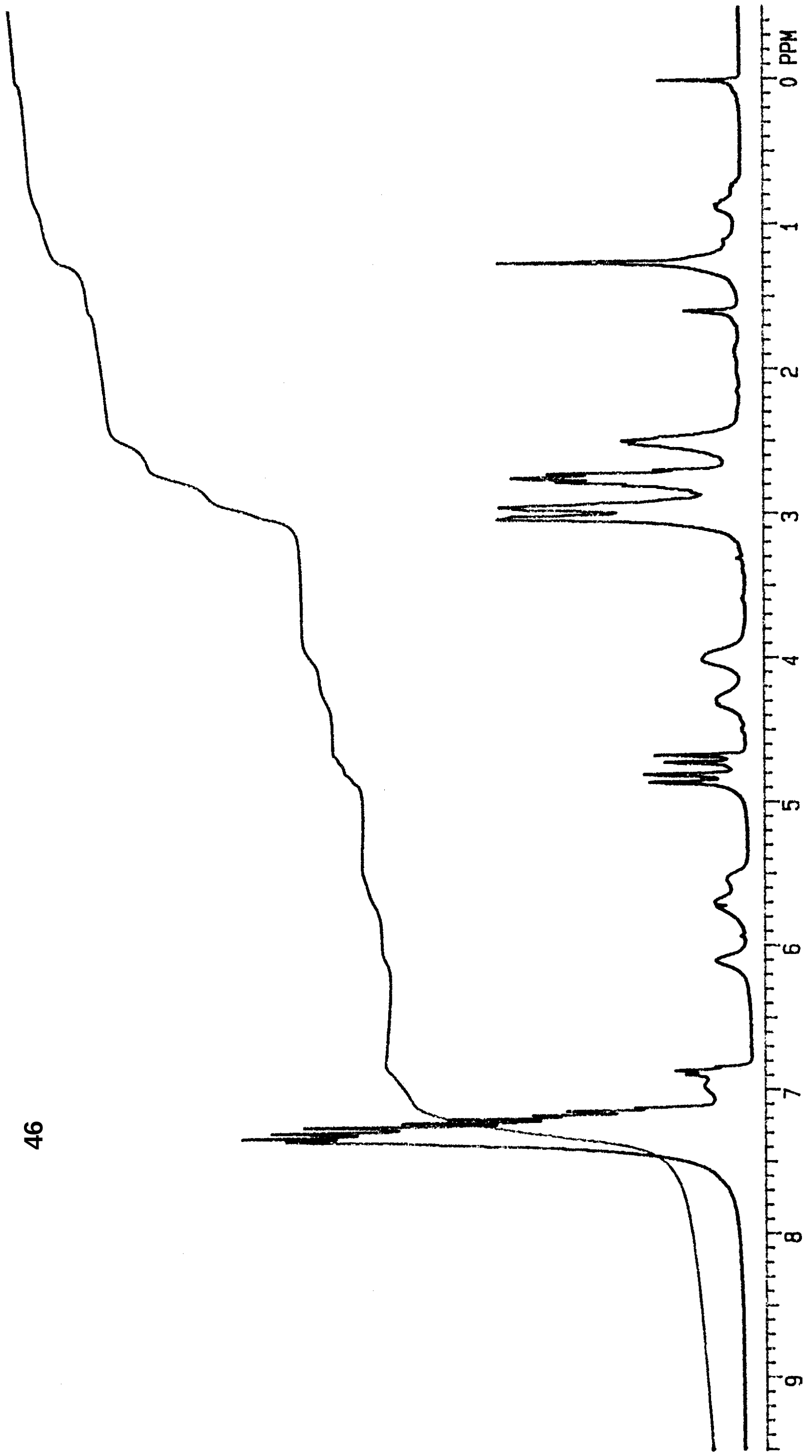

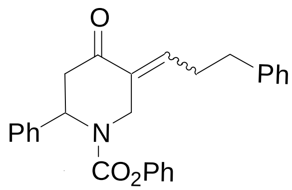

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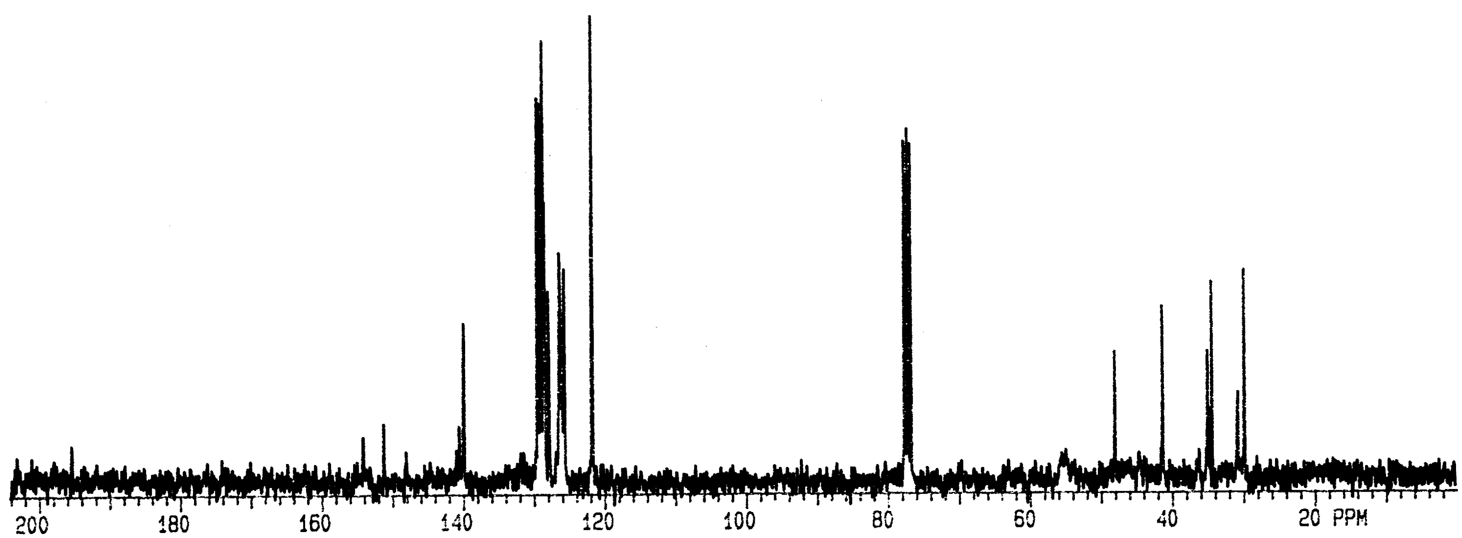

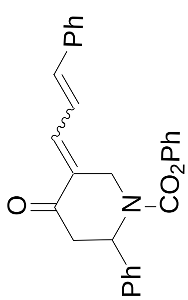

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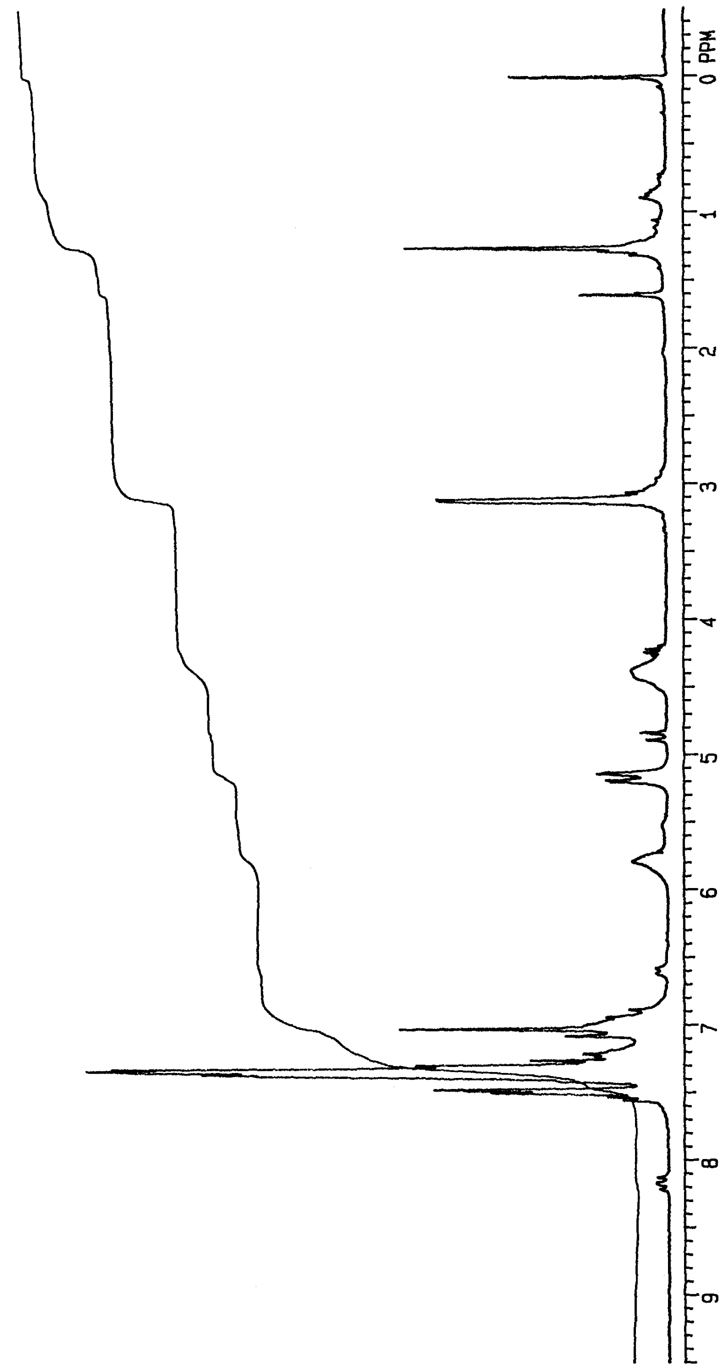

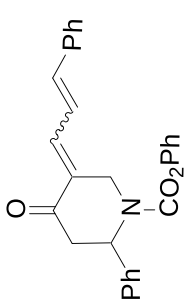

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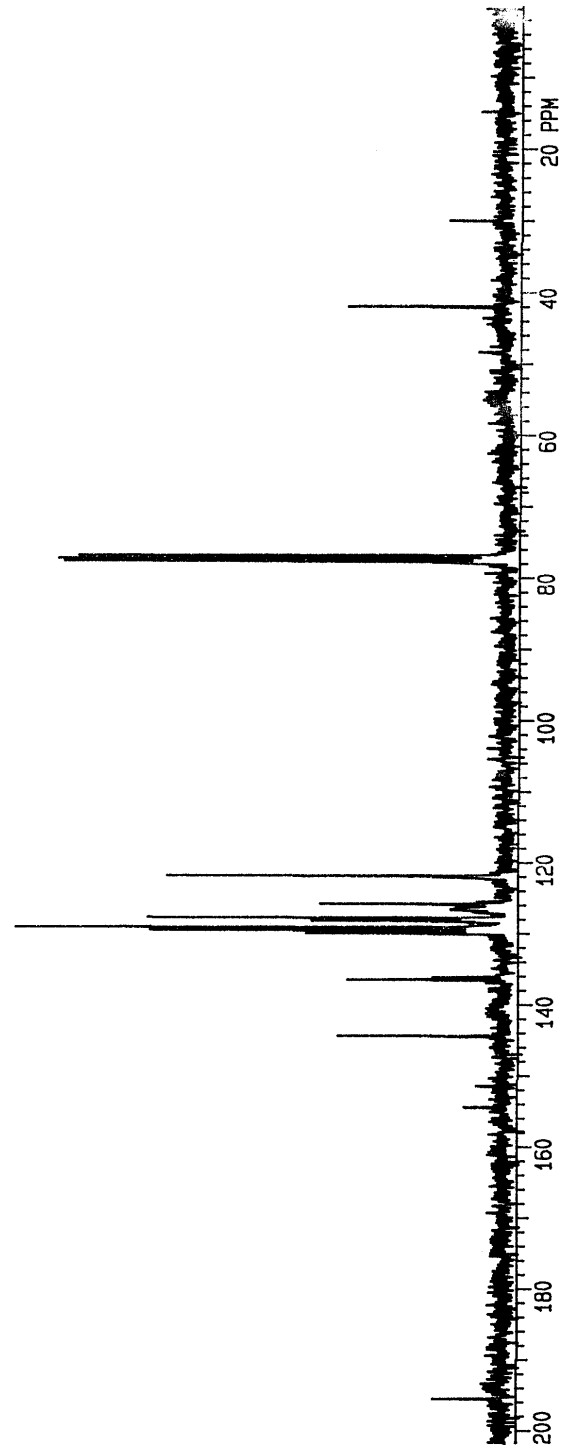

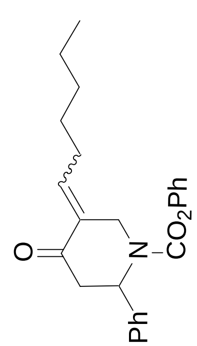
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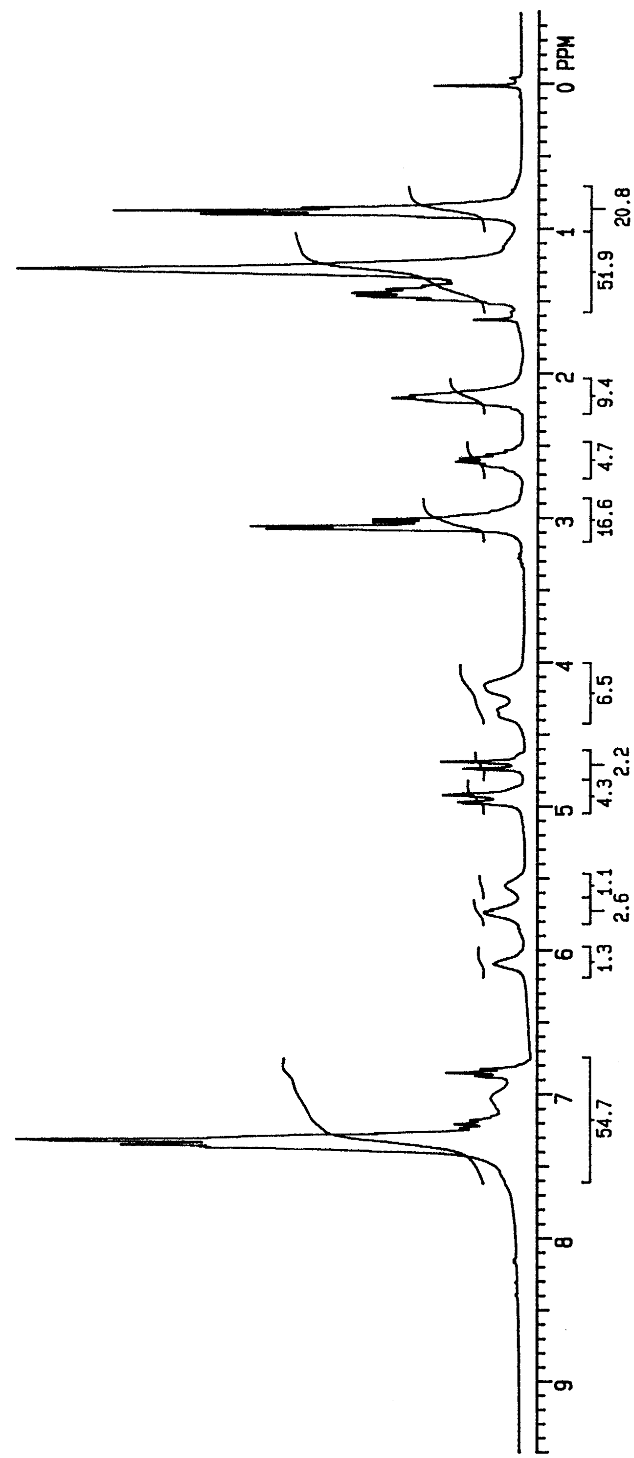
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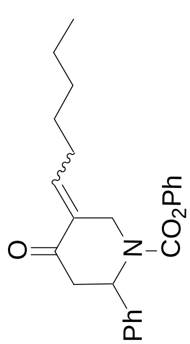

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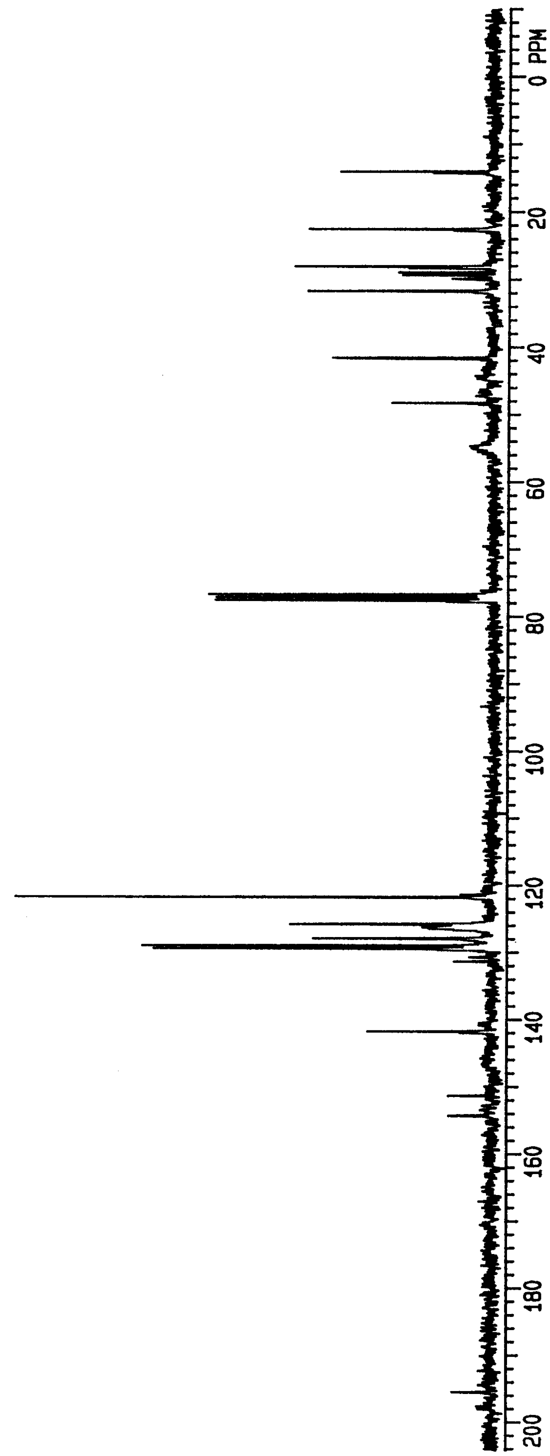

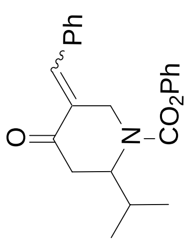

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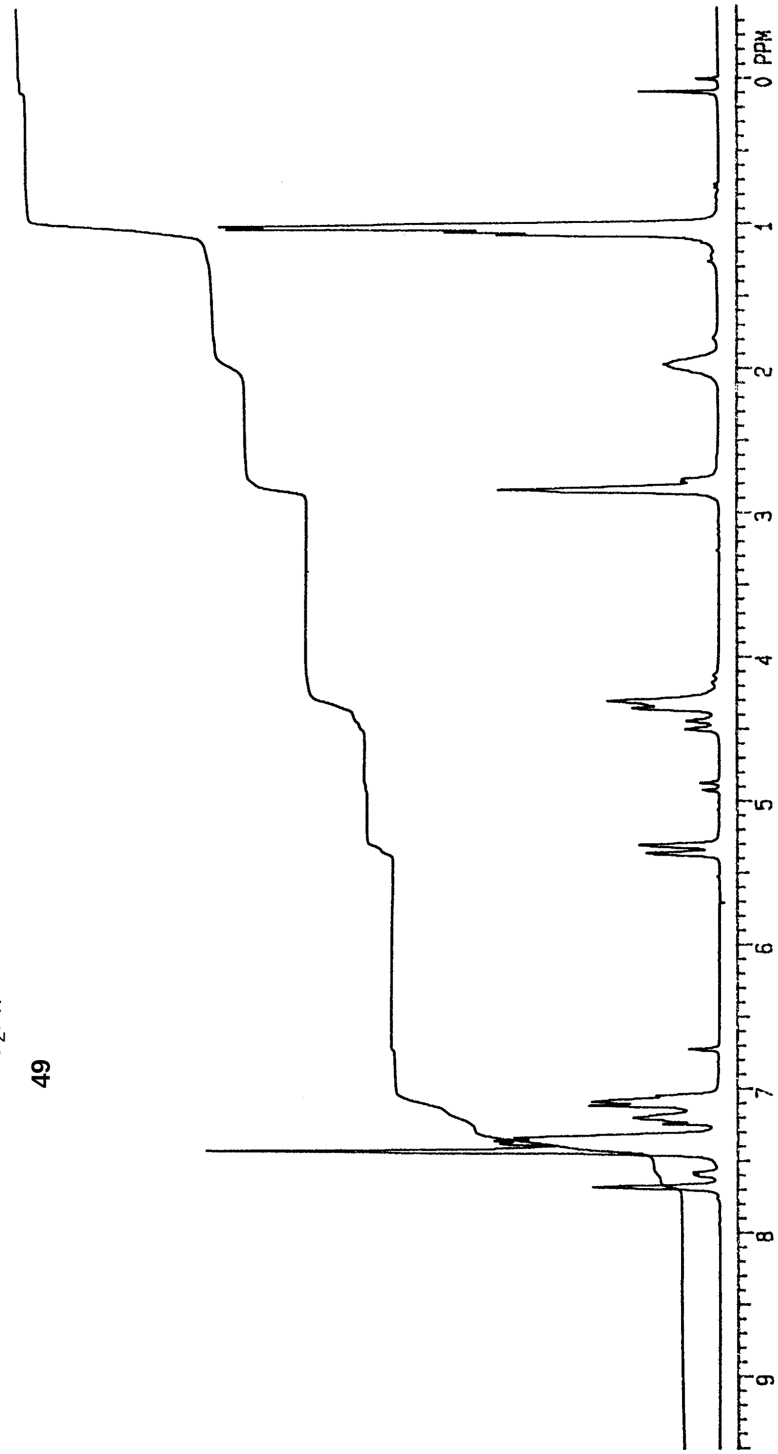

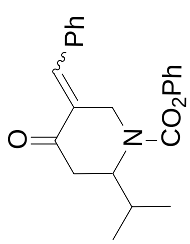

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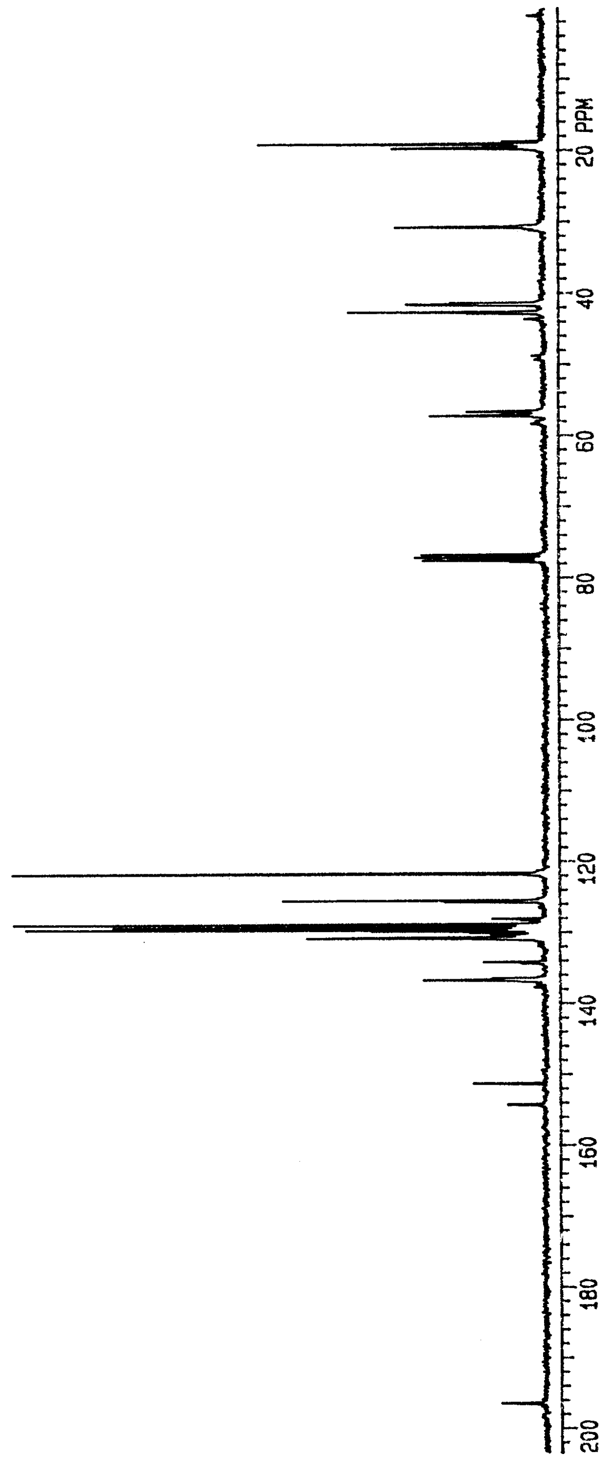

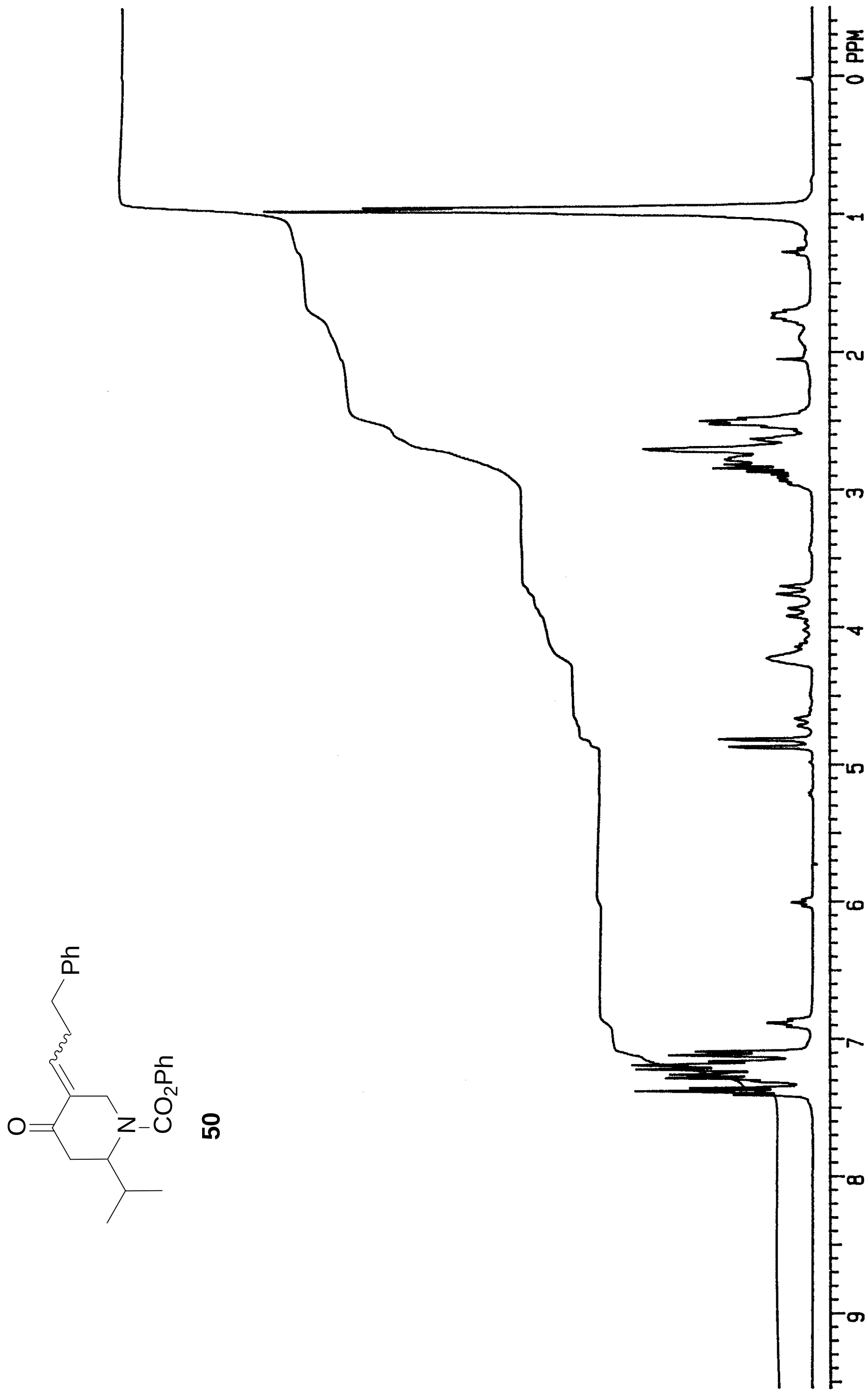

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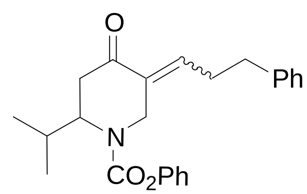

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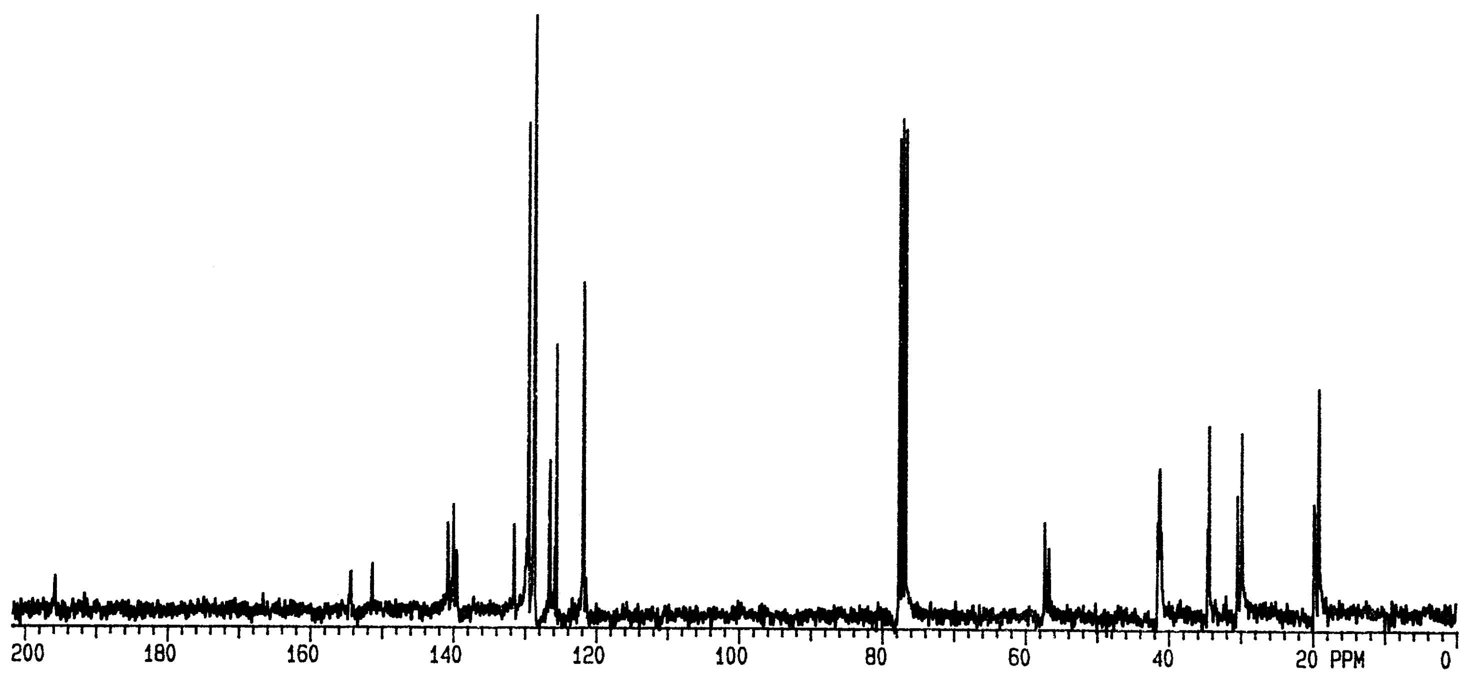

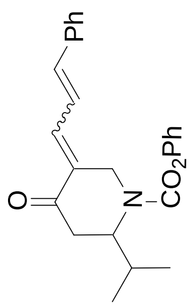

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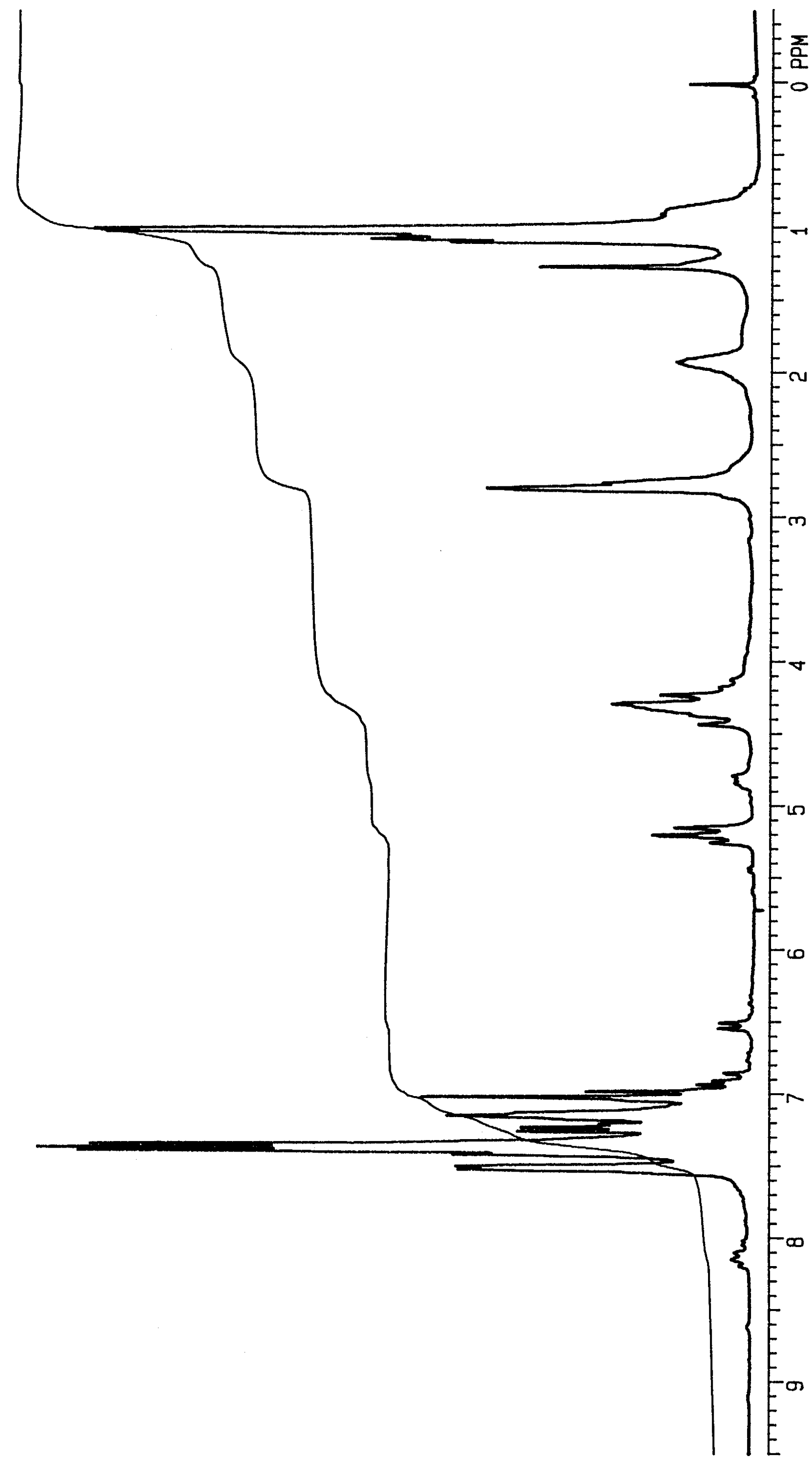

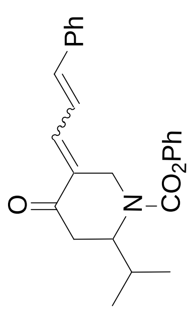

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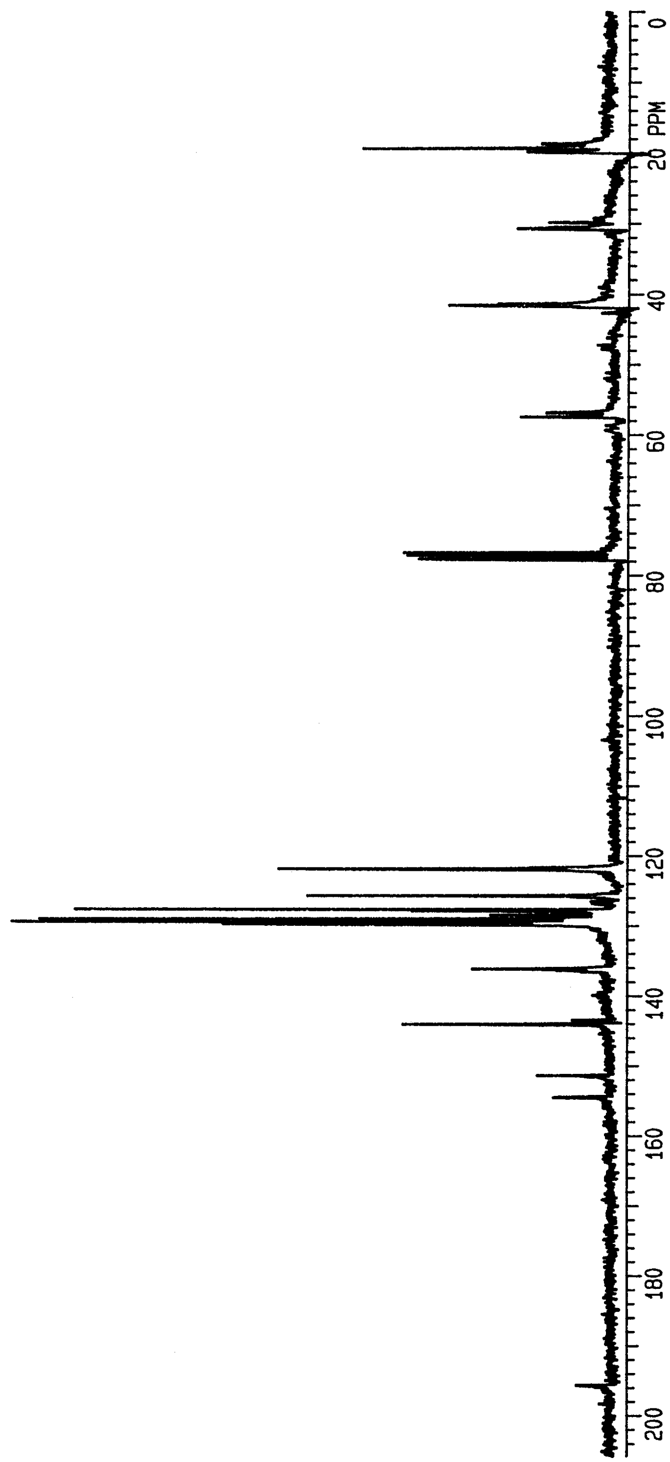

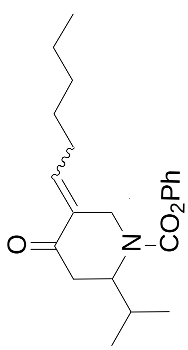

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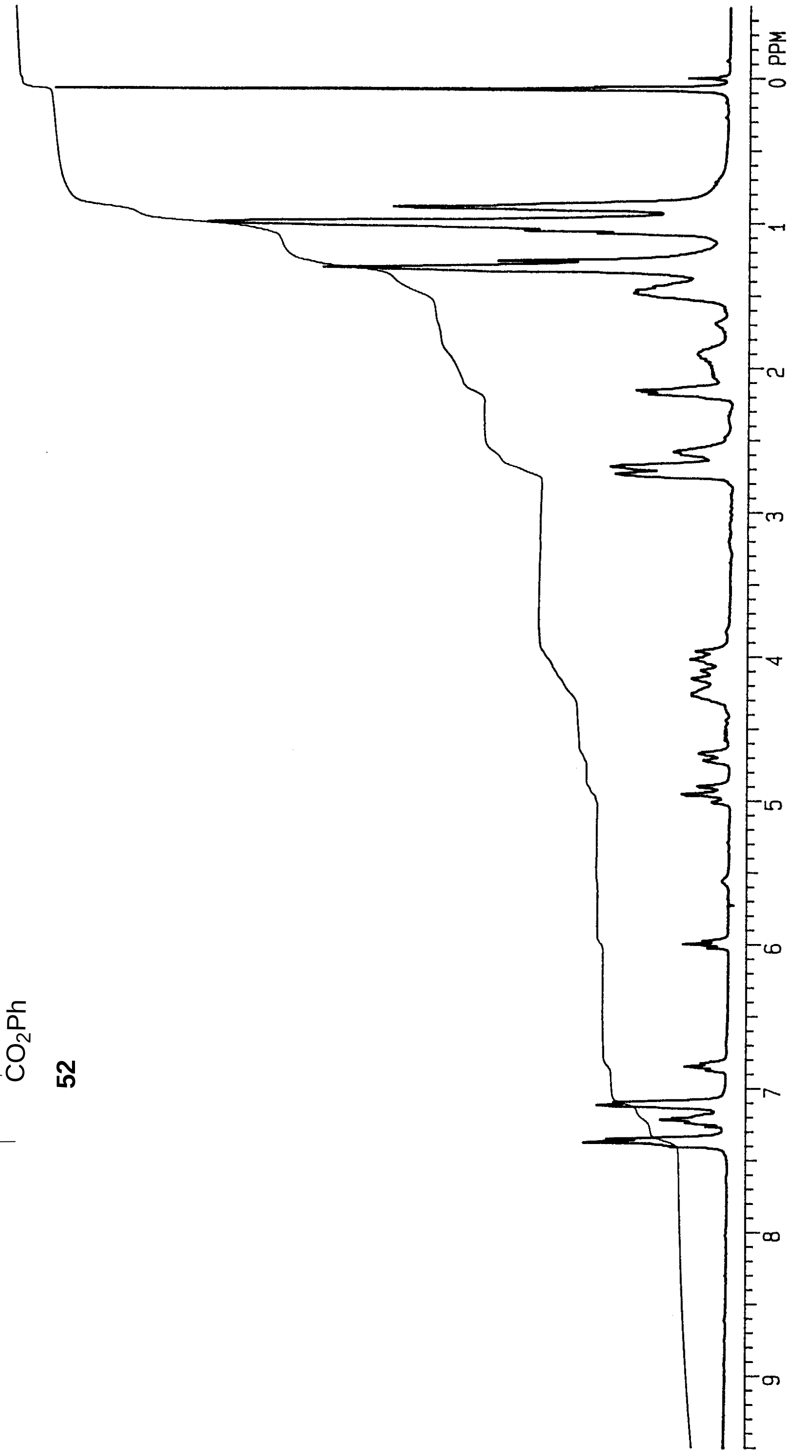

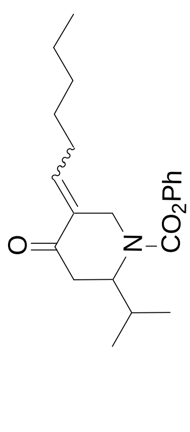

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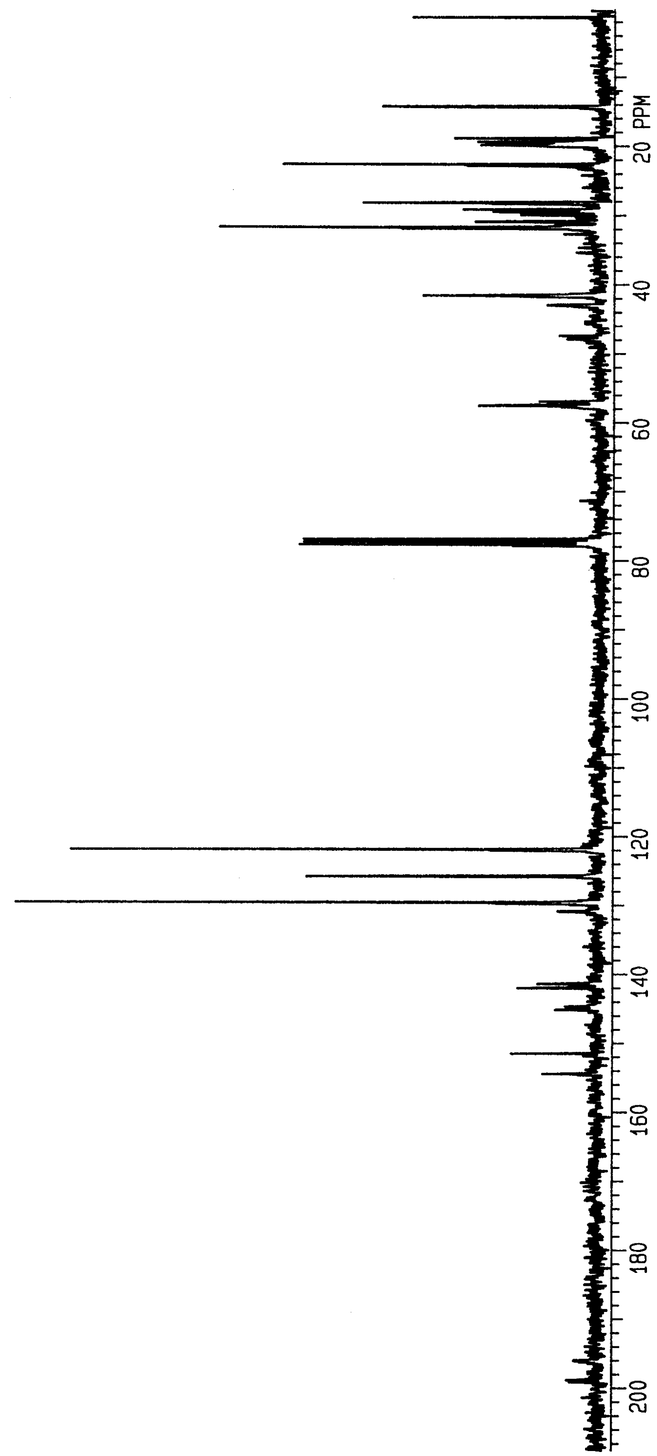

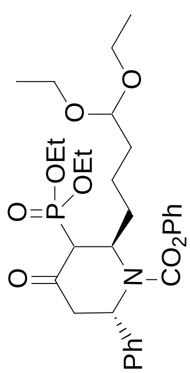

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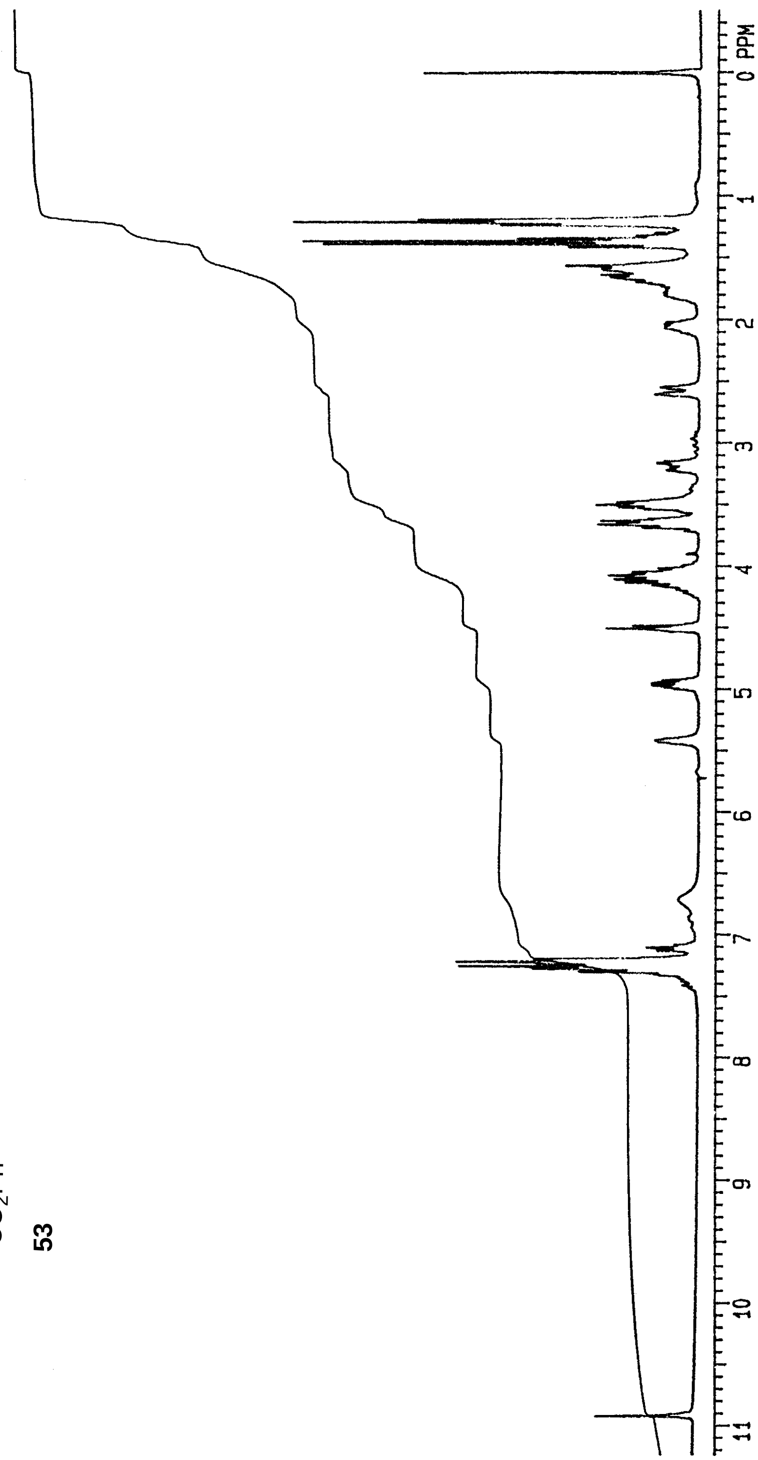

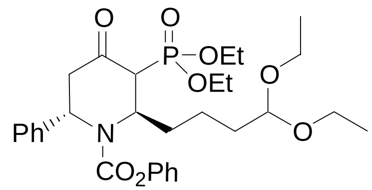

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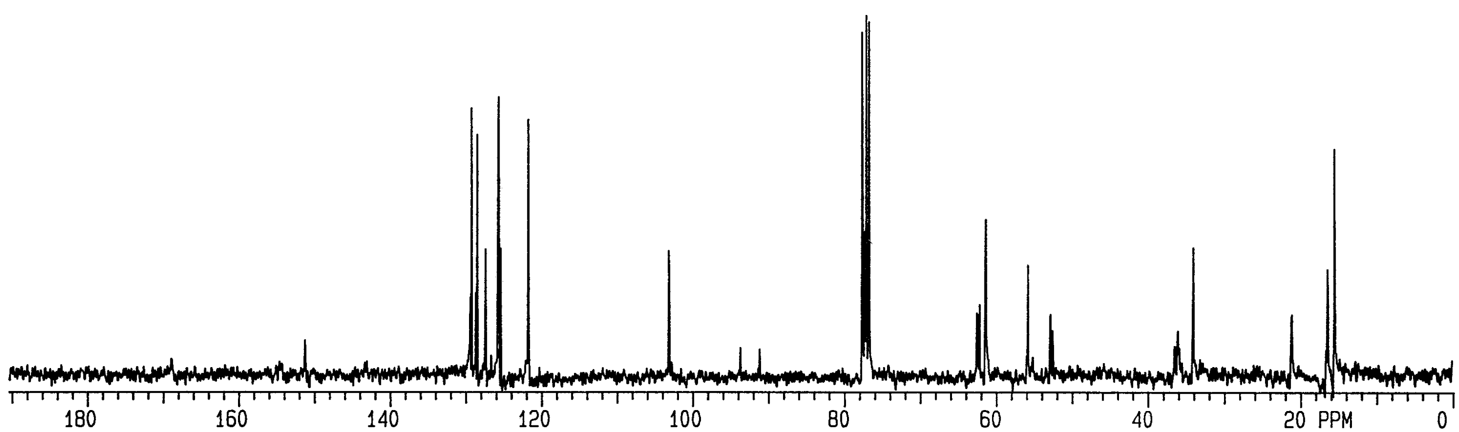
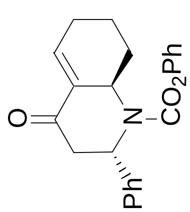
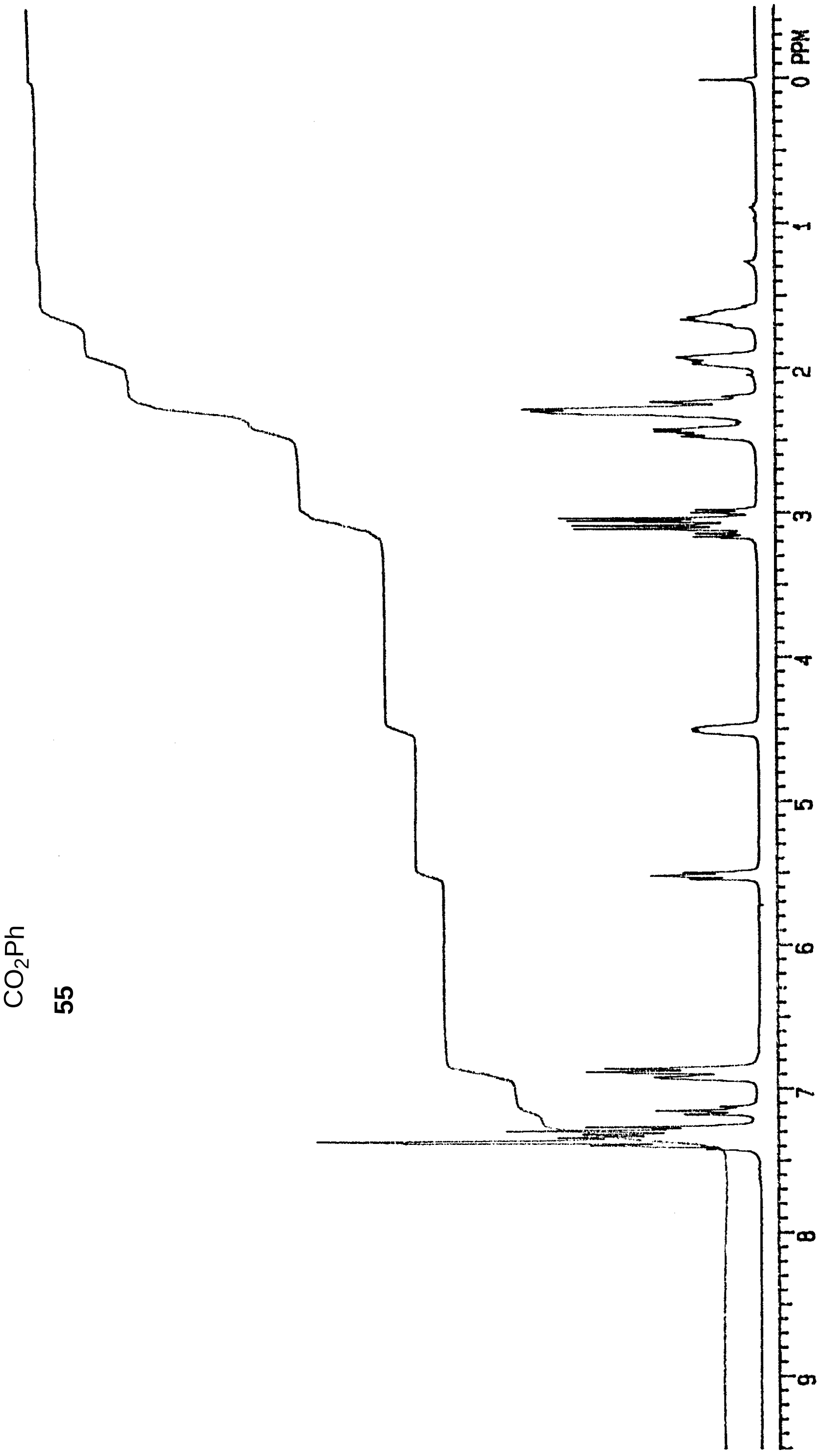
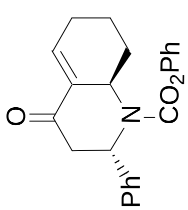


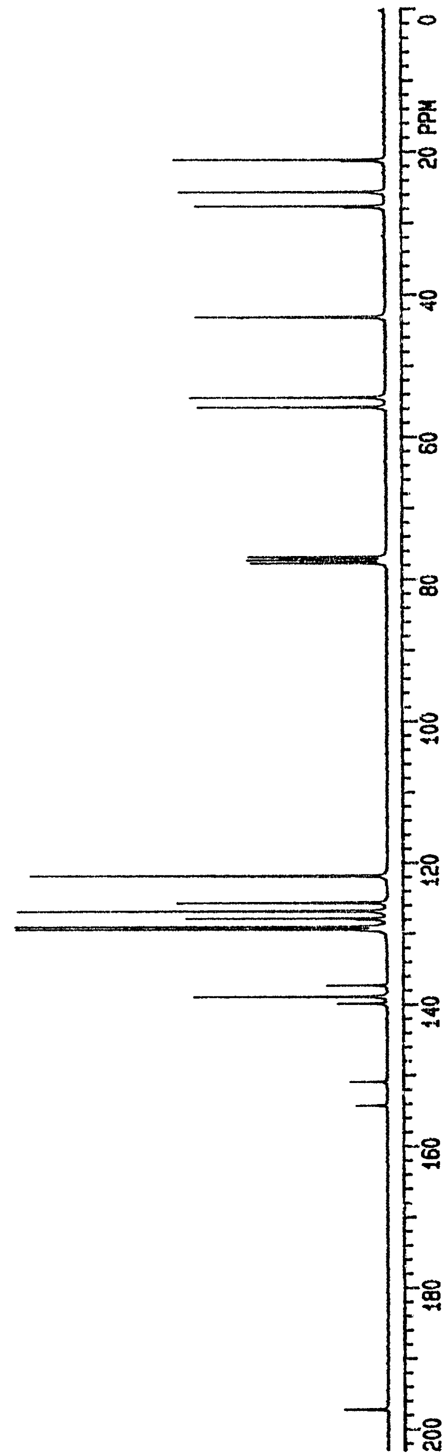


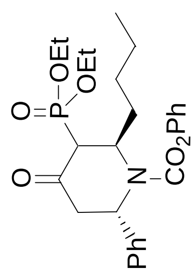
55



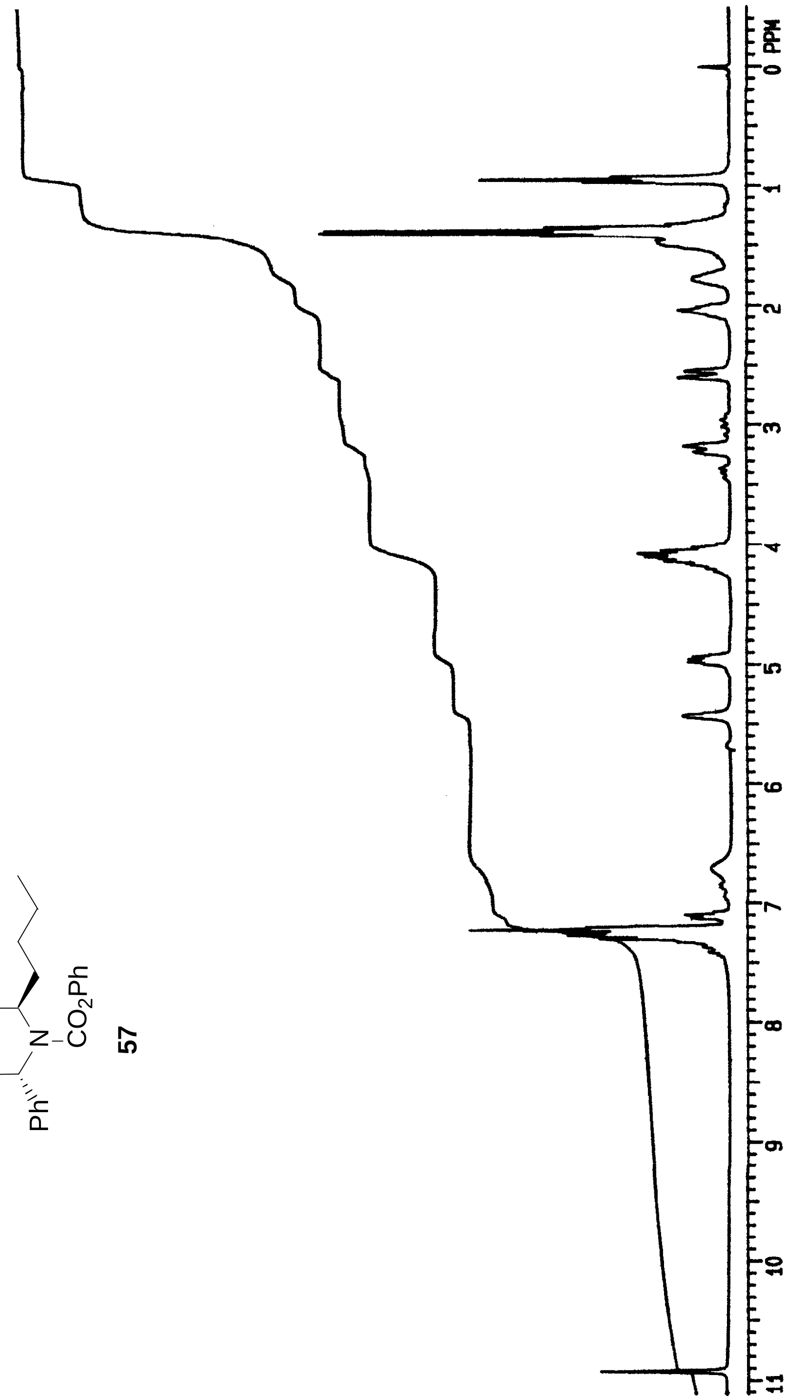


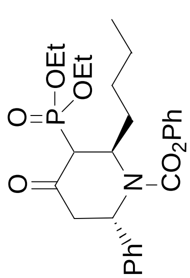
55



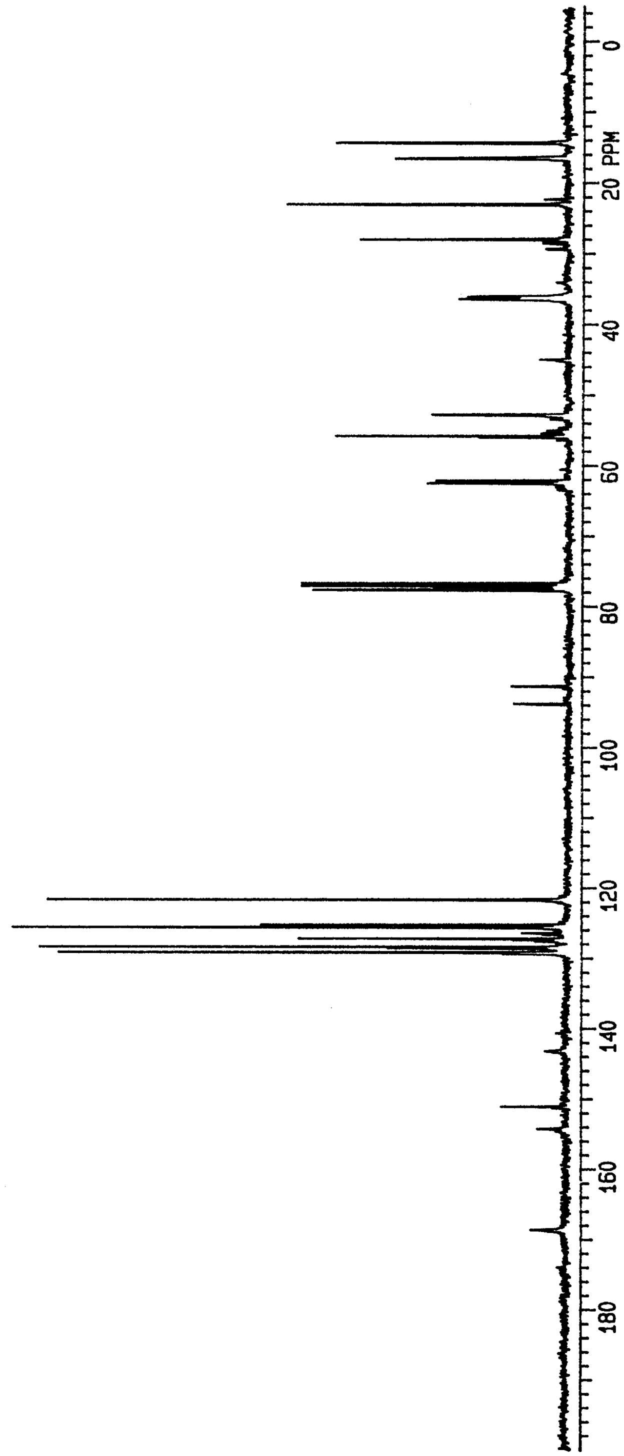


57





57



Supplemental X-Ray Information for Crystal Structure of Bicyclic Enone 55

```
   Space Group and Cell Dimensions    Monoclinic,   P 21/n
a  11.9009(6)   b   6.0534(3)   c  25.0087(9)
beta  94.523(6)
        Volume  1796.04(14)A**3

Empirical formula : C22 H21 N O3

Cell dimensions were obtained from  25 reflections with 2Theta angle
in the range  31.00 -  36.00 degrees.

Crystal dimensions :   .46  X   .28  X   .22  mm

FW =    347.41     Z =    4     F(000) =     736.44

Dcalc  1.285Mg.m-3, mu    .09mm-1, lambda  .71073A, 2Theta(max)  57.8

The intensity data were collected on a Nonius   diffractometer,
using the omega scan mode.
The h,k,l ranges used during structure solution and refinement are :--
Hmin,max  -16  16;  Kmin,max    0   8;  Lmin,max   0  33
No. of reflections measured                    4716
No. of unique reflections                      4716
No. of reflections with Inet > 1.0sigma(Inet)  3125
Merging R-value on intensities   .000
No correction was made for absorption


The last least squares cycle was calculated with
 47 atoms, 320 parameters and 3122 out of 4716 reflections.
Weights based on counting-statistics were used.
The weight modifier K in KFo**2 is  .000120

The residuals are as follows :--
For significant reflections,  RF  .041, Rw  .044 GoF  2.07
For all reflections,          RF  .041, Rw  .044.
where RF = Sum(Fo-Fc)/Sum(Fo),
      Rw = Sqrt[Sum(w(Fo-Fc)**2)/Sum(wFo**2)] and
     GoF = Sqrt[Sum(w(Fo-Fc)**2)/(No. of reflns - No. of params.)]
The maximum shift/sigma ratio was   .000.

In the last D-map, the deepest hole was   -.160e/A**3,
and the highest peak    .190e/A**3.

Secondary ext. coeff.     .4587microns     sigma     .0794
```

The following references are relevant to the NRCVAX System.

1. Full System Reference :
Gabe, E.J., Le Page, Y., Charland,.J.-P., Lee, F.L. and White, P.S. (1989) J. Appl. Cryst., 22, 384-387.

2. Scattering Factors from Int. Tab. Vol. 4 :
International Tables for X-ray Crystallography, Vol. IV, (1974) Kynoch Press, Birmingham, England.

The following references may also be relevant.

3. ORTEP Plotting :
Johnson, C.K., (1976) ORTEP - A Fortran Thermal Ellipsoid Plot Program, Technical Report ORNL-5138, Oak Ridge

4. Pluto Plotting :
S. Motherwell, University Chemical Laboratory, Cambridge, 1978

5. Missing Symmetry Treatment by MISSYM :
Le Page, Y., (1988) J. Appl. Cryst., 21, 983-984.

7. Extinction Treatment :
Larson, A.C., (1970) p.293, Crystallographic Computing, Munksgaard, Copenhagen.

Table . Atomic Parameters x,y,z and Biso.
E.S.Ds. refer to the last digit printed.

	x	y	z	Biso
O1	.18027(10)	.41676(19)	.37659(5)	4.85(5)
O2	-.24086(9)	.2048 (2)	.29089(4)	4.79(5)
O3	-.24812(8)	.30054(19)	.37797(4)	4.25(5)
N1	-.10876(9)	.08016(20)	.35477(4)	2.92(4)
C1	.10263(12)	.2851 (2)	.37473(6)	3.33(6)
C2	.08848(11)	.1091 (2)	.33303(5)	3.04(5)
C3	.18085(13)	.0146 (3)	.31642(6)	3.59(6)
C4	.18064(14)	-.1835 (3)	.28102(7)	4.24(7)
C5	.06310(14)	-.2622 (3)	.26124(7)	4.41(7)
C6	-.02370(13)	-.2219 (3)	.30177(7)	3.79(6)
C7	-.02810(11)	.0239 (3)	.31476(5)	3.07(6)
C8	-.05777(11)	.0821 (2)	.41083(5)	2.70(5)
C9	.01899(13)	.2828 (3)	.41764(6)	3.27(6)
C10	-.20230(11)	.1959 (3)	.33673(5)	3.19(5)
C11	-.34700(11)	.4238 (2)	.36792(5)	3.14(5)
C12	-.34972(14)	.6070 (3)	.33568(6)	4.02(7)
C13	-.44549(19)	.7385 (3)	.33335(8)	5.50(9)
C14	-.53545(18)	.6820 (5)	.36212(9)	6.52(11)
C15	-.53134(16)	.4968 (5)	.39289(10)	6.49(12)
C16	-.43666(14)	.3650 (3)	.39653(7)	4.70(8)
C17	-.14145(11)	.0467 (2)	.45300(5)	2.86(5)
C18	-.15415(13)	.1956 (3)	.49419(6)	3.56(6)
C19	-.23227(15)	.1574 (3)	.53155(6)	4.42(7)
C20	-.29849(16)	-.0291 (3)	.52792(7)	4.84(8)
C21	-.28512(15)	-.1812 (3)	.48773(7)	4.71(8)
C22	-.20618(13)	-.1439 (3)	.45089(6)	3.70(6)
H3	.2576 (13)	.077 (3)	.3298 (6)	4.2 (3)
H4a	.2299 (16)	-.157 (3)	.2510 (8)	6.1 (4)
H4b	.2218 (17)	-.312 (4)	.3032 (8)	6.8 (5)
H5a	.0685 (14)	-.419 (3)	.2535 (7)	5.0 (4)
H5b	.0360 (15)	-.181 (3)	.2281 (8)	6.1 (5)
H6a	-.0031 (13)	-.304 (3)	.3360 (7)	3.9 (3)
H6b	-.0982 (14)	-.266 (3)	.2881 (6)	4.3 (4)
H7	-.0561 (12)	.102 (3)	.2830 (6)	4.1 (3)
H8	-.0088 (11)	-.049 (2)	.4121 (5)	2.2 (3)
H9a	-.0263 (13)	.422 (3)	.4151 (6)	4.4 (4)
H9b	.0630 (13)	.282 (3)	.4522 (7)	4.1 (3)
H12	-.2890 (16)	.643 (3)	.3165 (7)	5.3 (4)
H13	-.4464 (16)	.871 (4)	.3122 (8)	6.5 (5)
H14	-.604 (2)	.765 (4)	.3632 (9)	8.6 (6)
H15	-.596 (2)	.458 (4)	.4131 (10)	9.3 (7)
H16	-.4289 (16)	.238 (4)	.4205 (8)	6.5 (5)
H18	-.1114 (13)	.329 (3)	.4954 (6)	4.4 (4)
H19	-.2374 (15)	.261 (3)	.5602 (8)	5.6 (4)
H20	-.3544 (16)	-.058 (3)	.5529 (8)	6.4 (5)
H21	-.3291 (15)	-.319 (3)	.4855 (7)	5.7 (4)
H22	-.1988 (12)	-.251 (3)	.4226 (6)	4.1 (3)

Biso is the Mean of the Principal Axes of the Thermal Ellipsoid

Table of u(i,j) or U values *100.
E.S.Ds. refer to the last digit printed

	u11(U)	u22	u33	u12	u13	u23
O1	5.62(7)	5.33(7)	7.56(8)	-1.47(6)	1.12(6)	-.27(6)
O2	5.44(6)	9.03(9)	3.48(5)	3.01(6)	-1.13(4)	-.83(6)
O3	4.86(6)	7.87(8)	3.34(5)	3.06(6)	-.12(4)	-.15(5)
N1	3.24(6)	5.07(7)	2.71(5)	.88(5)	-.16(4)	-.13(5)
C1	4.21(8)	3.73(8)	4.66(8)	.21(7)	.04(6)	.66(7)
C2	3.95(7)	3.96(8)	3.68(7)	.30(6)	.53(6)	.72(6)
C3	3.91(8)	4.90(9)	4.92(8)	.06(7)	.92(6)	.53(7)
C4	4.66(9)	5.70(11)	5.99(10)	.46(8)	1.79(8)	-.44(9)
C5	5.29(10)	6.68(12)	4.79(9)	1.15(9)	.49(7)	-1.55(9)
C6	3.67(8)	5.79(10)	4.90(9)	-.06(8)	.15(7)	-1.56(8)
C7	3.53(7)	5.24(9)	2.87(6)	.97(7)	.08(5)	.12(6)
C8	3.50(7)	3.86(7)	2.83(6)	.60(6)	-.32(5)	-.03(6)
C9	4.50(8)	3.95(8)	3.92(7)	.07(7)	-.03(6)	-.25(6)
C10	3.46(7)	5.25(9)	3.36(7)	.96(7)	-.08(5)	-.21(7)
C11	3.25(7)	4.91(9)	3.70(7)	1.10(6)	-.26(6)	-.32(6)
C12	4.76(9)	5.95(10)	4.43(8)	.45(8)	-.43(7)	.36(8)
C13	8.74(15)	5.89(12)	5.85(11)	2.50(11)	-2.00(10)	-.04(9)
C14	5.76(12)	10.47(19)	8.32(14)	4.39(13)	-.88(11)	-2.04(14)
C15	4.38(10)	10.97(19)	9.50(15)	1.43(12)	1.83(10)	-.36(15)
C16	5.07(10)	6.46(12)	6.47(11)	.55(9)	1.34(8)	.44(10)
C17	3.83(7)	4.13(8)	2.81(6)	.55(6)	-.29(5)	.17(6)
C18	5.28(9)	4.42(9)	3.82(7)	.16(8)	.36(6)	-.34(7)
C19	6.69(11)	6.09(11)	4.12(8)	.77(9)	1.18(8)	-.66(8)
C20	6.15(11)	7.59(13)	4.89(9)	-.34(10)	1.87(8)	.25(9)
C21	6.15(11)	6.52(11)	5.32(9)	-1.57(10)	1.02(8)	.06(9)
C22	5.32(9)	4.95(9)	3.76(7)	-.51(7)	.20(6)	-.56(7)
H3	5.4(4)					
H4a	7.8(6)					
H4b	8.6(6)					
H5a	6.3(5)					
H5b	7.7(6)					
H6a	5.0(4)					
H6b	5.4(5)					
H7	5.2(4)					
H8	2.7(3)					
H9a	5.5(5)					
H9b	5.2(4)					
H12	6.7(5)					
H13	8.2(6)					
H14	10.8(8)					
H15	11.8(8)					
H16	8.2(6)					
H18	5.5(5)					
H19	7.1(5)					
H20	8.1(6)					
H21	7.3(5)					
H22	5.1(4)					

Anisotropic Temperature Factors are of the form
Temp=-2*Pi*Pi*(h*h*u11*astar*astar+---+2*h*k*u12*astar*bstar+---)

DISANG -- The NRCVAX Distance and Angle Program

The Space Group is P 21/N Centrosymmetric
The Equivalent Positions are:
1) x y z 2) 1/2-x 1/2+y 1/2-z

The Lattice is Primitive. There are no Centring Vectors

The following Atoms are from the CD File
* Indicates that there are Symmetry Equivalents of an atom.

Name	x	y	z
O1	.18027(9)	.41676(18)	.37659(4)
O2	-.24086(8)	.20482(21)	.29089(3)
O3	-.24812(8)	.30054(19)	.37797(3)
N1	-.10876(8)	.08016(19)	.35477(3)
C1	.10263(11)	.28513(24)	.37473(5)
C2	.08848(11)	.10915(23)	.33303(5)
C3	.18085(13)	.01461(26)	.31642(5)
C4	.18064(13)	-.18353(30)	.28102(7)
C5	.06310(14)	-.26217(34)	.26124(7)
C6	-.02370(12)	-.22186(28)	.30177(6)
C7	-.02810(11)	.02395(25)	.31476(5)
C8	-.05777(10)	.08210(23)	.41083(4)
C9	.01899(12)	.28275(25)	.41764(5)
C10	-.20230(11)	.19589(25)	.33673(5)
C11	-.34700(10)	.42381(24)	.36792(5)
C12	-.34972(14)	.60696(29)	.33568(6)
C13	-.44549(18)	.73846(34)	.33335(7)
C14	-.53545(17)	.68196(45)	.36212(9)
C15	-.53134(16)	.49683(45)	.39289(9)
C16	-.43666(14)	.36505(33)	.39653(7)
C17	-.14145(11)	.04670(22)	.45300(4)
C18	-.15415(13)	.19560(27)	.49419(5)
C19	-.23227(14)	.15743(31)	.53155(6)
C20	-.29849(15)	-.02909(32)	.52792(6)
C21	-.28512(14)	-.18119(32)	.48773(6)
C22	-.20618(13)	-.14390(27)	.45089(5)
H3	.25756(128)	.07689(269)	.32980(57)
H4a	.22989(156)	-.15696(315)	.25098(79)
H4b	.22183(173)	-.31196(350)	.30319(81)
H5a	.06855(142)	-.41899(316)	.25350(66)
H5b	.03600(150)	-.18132(316)	.22811(79)
H6a	-.00306(125)	-.30414(255)	.33603(65)
H6b	-.09823(137)	-.26627(261)	.28809(62)
H7	-.05606(124)	.10154(261)	.28303(62)
H8	-.00875(105)	-.04878(222)	.41207(48)
H9a	-.02626(131)	.42201(290)	.41510(60)
H9b	.06299(130)	.28203(254)	.45216(66)
H12	-.28896(155)	.64270(300)	.31652(68)
H13	-.44637(161)	.87117(360)	.31222(79)
H14	-.60442(202)	.76515(392)	.36321(92)
H15	-.59552(201)	.45768(421)	.41314(95)

H16	-.42888(163)	.23848(353)	.42054(79)
H18	-.11137(130)	.32857(303)	.49539(63)
H19	-.23738(152)	.26105(304)	.56024(76)
H20	-.35439(163)	-.05784(324)	.55292(75)
H21	-.32911(154)	-.31928(326)	.48551(72)
H22	-.19875(124)	-.25133(269)	.42259(64)

O1-C1	1.2182(18)
O2-C10	1.2021(16)
O3-C10	1.3606(16)
O3-C11	1.3994(16)
N1-C7	1.4799(16)
N1-C8	1.4831(15)
N1-C10	1.3617(17)
C1-C2	1.491(2)
C1-C9	1.520(2)
C2-C3	1.3343(20)
C2-C7	1.5164(19)
C3-C4	1.491(2)
C3-H3	1.020(16)
C4-C5	1.523(2)
C4-H4a	1.00(2)
C4-H4b	1.05(2)
C5-C6	1.523(2)
C5-H5a	.972(19)
C5-H5b	.99(2)
C6-C7	1.525(2)
C6-H6a	1.005(17)
C6-H6b	.963(17)
C7-H7	.959(16)
C8-C9	1.521(2)
C8-C17	1.5216(18)
C8-H8	.983(14)
C9-H9a	1.000(18)
C9-H9b	.974(17)
C11-C12	1.370(2)
C11-C16	1.377(2)
C12-C13	1.388(3)
C12-H12	.924(19)
C13-C14	1.379(4)
C13-H13	.96(2)
C14-C15	1.358(4)
C14-H14	.97(2)
C15-C16	1.378(3)
C15-H15	.98(2)
C16-H16	.97(2)
C17-C18	1.3859(19)
C17-C22	1.386(2)
C18-C19	1.388(2)
C18-H18	.952(18)
C19-C20	1.376(3)
C19-H19	.959(20)
C20-C21	1.381(3)
C20-H20	.96(2)
C21-C22	1.385(2)
C21-H21	.99(2)
C22-H22	.970(17)

C10-O3-C11	119.58(10)
C7-N1-C8	113.71(10)
C7-N1-C10	116.82(10)
C8-N1-C10	124.89(11)
O1-C1-C2	122.51(13)
O1-C1-C9	121.08(13)
C2-C1-C9	116.29(12)
C1-C2-C3	118.28(13)
C1-C2-C7	120.25(11)
C3-C2-C7	121.13(13)
C2-C3-C4	124.55(15)
C2-C3-H3	118.5(9)
C4-C3-H3	116.9(9)
C3-C4-C5	113.76(13)
C3-C4-H4a	110.0(11)
C3-C4-H4b	107.6(11)
C5-C4-H4a	112.5(11)
C5-C4-H4b	108.5(11)
H4a-C4-H4b	103.8(15)
C4-C5-C6	112.80(13)
C4-C5-H5a	107.2(10)
C4-C5-H5b	110.3(11)
C6-C5-H5a	110.4(10)
C6-C5-H5b	106.5(10)
H5a-C5-H5b	109.7(15)
C5-C6-C7	109.55(14)
C5-C6-H6a	110.8(8)
C1-C9-H9a	109.1(9)
C1-C9-H9b	106.8(9)
C8-C9-H9a	110.5(9)
C8-C9-H9b	111.8(9)
H9a-C9-H9b	108.0(13)
O2-C10-O3	123.81(12)
O2-C10-N1	125.44(12)
O3-C10-N1	110.76(11)
O3-C11-C12	121.19(13)
O3-C11-C16	116.35(14)
C12-C11-C16	122.11(14)
C11-C12-C13	118.05(17)
C11-C12-H12	120.8(11)
C13-C12-H12	121.2(11)
C12-C13-C14	120.3(2)
C12-C13-H13	118.4(12)
C14-C13-H13	121.3(12)
C13-C14-C15	120.36(18)
C13-C14-H14	125.6(14)
C15-C14-H14	114.0(14)
C14-C15-C16	120.53(20)
C14-C15-H15	119.9(15)
C16-C15-H15	119.6(15)
C11-C16-C15	118.63(19)
C11-C16-H16	118.9(11)
C15-C16-H16	122.3(11)
C8-C17-C18	122.63(13)

C5-C6-H6b	111.9(9)	C8-C17-C22	118.88(12)
C7-C6-H6a	108.2(9)	C18-C17-C22	118.48(13)
C7-C6-H6b	107.6(9)	C17-C18-C19	120.66(16)
H6a-C6-H6b	108.7(13)	C17-C18-H18	119.0(9)
N1-C7-C2	110.18(11)	C19-C18-H18	120.3(9)
N1-C7-C6	113.80(12)	C18-C19-C20	120.18(16)
N1-C7-H7	104.2(9)	C18-C19-H19	118.7(11)
C2-C7-C6	110.50(12)	C20-C19-H19	121.1(11)
C2-C7-H7	109.4(9)	C19-C20-C21	119.78(16)
C6-C7-H7	108.5(9)	C19-C20-H20	121.8(12)
N1-C8-C9	108.02(11)	C21-C20-H20	118.4(12)
N1-C8-C17	114.37(10)	C20-C21-C22	119.89(17)
N1-C8-H8	102.8(7)	C20-C21-H21	121.0(11)
C9-C8-C17	117.06(11)	C22-C21-H21	119.1(11)
C9-C8-H8	106.9(7)	C17-C22-C21	120.95(15)
C17-C8-H8	106.5(7)	C17-C22-H22	120.2(9)
C1-C9-C8	110.45(12)	C21-C22-H22	118.8(9)

Torsion angles

C11	O3	C10	O2	1.20(13)	C11	O3	C10	N1	-178.2(3)
C10	O3	C11	C12	-63.4(2)	C10	O3	C11	C16	123.3(3)
C8	N1	C7	C2	-32.78(14)	C8	N1	C7	C6	92.0(2)
C10	N1	C7	C2	124.4(2)	C10	N1	C7	C6	-110.8(2)
C7	N1	C8	C9	71.17(18)	C7	N1	C8	C17	-156.6(2)
C10	N1	C8	C9	-83.9(2)	C10	N1	C8	C17	48.35(16)
C7	N1	C10	O2	21.42(14)	C7	N1	C10	O3	-159.2(3)
C8	N1	C10	O2	175.8(3)	C8	N1	C10	O3	-4.75(11)
O1	C1	C2	C3	35.68(17)	O1	C1	C2	C7	-150.9(3)
C9	C1	C2	C3	-140.3(3)	C9	C1	C2	C7	33.18(15)
O1	C1	C9	C8	-171.2(3)	C2	C1	C9	C8	4.79(12)
C1	C2	C3	C4	171.0(3)	C7	C2	C3	C4	-2.38(14)
C1	C2	C7	N1	-18.79(12)	C1	C2	C7	C6	-145.4(3)
C3	C2	C7	N1	154.5(3)	C3	C2	C7	C6	27.86(17)
C2	C3	C4	C5	4.45(16)	C3	C4	C5	C6	-32.58(17)
C4	C5	C6	C7	58.2(2)	C5	C6	C7	N1	-178.9(3)
C5	C6	C7	C2	-54.33(19)	N1	C8	C9	C1	-53.23(16)
C17	C8	C9	C1	176.0(3)	N1	C8	C17	C18	-123.6(3)
N1	C8	C17	C22	57.66(18)	C9	C8	C17	C18	4.18(14)
C9	C8	C17	C22	-174.5(3)	O3	C11	C12	C13	-171.3(3)
C16	C11	C12	C13	1.65(20)	O3	C11	C16	C15	172.5(4)
C12	C11	C16	C15	-.8(2)	C11	C12	C13	C14	-1.17(20)
C12	C13	C14	C15	-.2(2)	C13	C14	C15	C16	1.12(20)
C14	C15	C16	C11	-.66(20)	C8	C17	C18	C19	179.5(3)
C22	C17	C18	C19	-1.80(16)	C8	C17	C22	C21	-178.7(3)
C18	C17	C22	C21	2.56(17)	C17	C18	C19	C20	-.25(16)
C18	C19	C20	C21	1.58(17)	C19	C20	C21	C22	-.83(17)
C20	C21	C22	C17	-1.27(16)					

Table . Distances(A) to the least-squares planes.

Plane no. 1

Equation of the plane : 3.900(10)X + 3.271(4)Y + 18.675(15)Z = 6.898(8)

Distances(A) to the plane from the atoms in the plane.

C11	.0053(19)	C12	-.008(2)
C13	.005(3)	C14	.007(3)
C15	-.008(3)	C16	-.002(2)

Chi squared for this plane 36.395

Plane no. 2

Equation of the plane : 7.768(6)X - 2.909(4)Y + 13.316(15)Z = 4.808(8)

Distances(A) to the plane from the atoms in the plane.

C17	-.0106(17)	C18	.006(2)
C19	.008(2)	C20	-.012(2)
C21	-.001(2)	C22	.013(2)

Chi squared for this plane 123.513

Plane no. 3

Equation of the plane : 6.888(7)X + 4.857(3)Y - 4.761(19)Z = 2.048(7)

Distances(A) to the plane from the atoms in the plane.

O2	-.0008(18)	O3	-.0006(17)
N1	-.0006(17)	C10	.0031(17)

Chi squared for this plane 3.961

Plane no. 4

Equation of the plane : 6.438(8)X - 3.769(4)Y + 13.030(18)Z = 4.491(7)

Distances(A) to the plane from the atoms in the plane.

O1	.0057(16)	C1	-.0223(16)
C2	.0065(20)	C9	.007(2)

Chi squared for this plane 223.950

Plane no. 5

Equation of the plane :- .318(11)X - 4.082(4)Y + 18.449(14)Z = 5.705(5)

Distances(A) to the plane from the atoms in the plane.

C1	.012(2)	C2	-.0348(16)
C3	.015(2)	C7	.013(2)

Chi squared for this plane 589.163

Plane no. 6

Equation of the plane : 5.164(10)X + 5.450(2)Y - 1.70(2)Z = .551(8)

Distances(A) to the plane from the atoms in the plane.

C7	.000(2)	C8	.000(2)
C10	.000(2)		

Distances(A) to the plane from the atoms out of the plane.

N1	-.1784(15)

1 Dihedral angle between planes A and B

A	B	Angle(deg)
1	2	65.91(7)
1	3	59.29(7)
1	4	73.80(7)
1	5	78.54(7)
1	6	52.96(7)
2	3	95.33(6)
2	4	10.44(6)
2	5	42.37(7)
2	6	99.75(6)
3	4	105.58(6)
3	5	131.59(6)
3	6	11.89(7)
4	5	34.65(7)
4	6	110.18(7)
5	6	130.08(7)

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
-16, 0, l				5	52	45	30	4	64	58	24	1	21	23	50
2	19	0	210*	-14, 0, l				5	45	38	32	2	13	0	245*
4	4	0	751*	2	41	37	29	6	7	0	199*	3	58	55	22
6	25	8	51	4	36	32	32	7	46	44	32	4	4	0	939*
-16, 1, l				6	15	14	70	8	5	0	260*	5	62	59	22
1	24	0	177*	8	108	104	17	9	56	44	27	6	20	0	131*
2	11	0	126*	10	29	0	150*	10	39	36	35	7	4	0	977*
3	23	10	55	12	125	118	18	11	42	44	35	8	71	73	20
4	17	16	76	14	84	86	22	12	4	0	1117*	9	26	27	44
5	8	0	442*	16	46	44	33	13	4	0	885*	10	24	27	50
-15, 0, l				18	48	46	32	-14, 4, l				11	48	36	24
1	4	0	1200*	-14, 1, l				1	4	0	992*	12	52	56	26
3	18	0	191*	1	11	0	652*	2	27	33	55	13	4	0	813*
5	52	48	27	2	70	69	21	3	60	56	29	14	65	63	23
7	36	42	38	3	38	41	32	4	21	0	174*	15	4	0	878*
9	14	0	87*	4	49	53	27	5	31	39	49	16	27	11	44
11	20	0	175*	5	98	91	17	6	4	0	1030*	17	26	0	140*
13	4	0	1070*	6	4	0	1036*	-13, 0, l				18	30	32	47
15	80	76	24	7	4	0	880*	1	83	79	18	19	59	57	29
-15, 1, l				8	42	39	29	3	7	0	345*	20	19	0	211*
1	11	0	352*	9	4	0	941*	5	7	0	390*	-13, 3, l			
2	7	0	190*	10	46	38	27	7	27	29	37	1	4	0	819*
3	48	51	32	11	29	26	42	9	60	57	22	2	4	0	936*
4	7	0	180*	12	28	0	154*	11	38	42	30	3	24	0	129*
5	4	0	1080*	13	66	63	24	13	59	61	23	4	19	0	160*
6	16	0	236*	14	18	0	221*	15	65	55	22	5	21	0	153*
7	4	0	972*	15	40	33	34	17	74	72	21	6	24	17	47
8	18	0	209*	16	8	0	475*	19	4	0	1022*	7	31	33	38
9	4	0	930*	17	77	65	24	21	7	0	189*	8	41	36	32
10	31	29	45	18	69	57	25	-13, 1, l				9	9	0	116*
11	4	0	951*	-14, 2, l				1	16	0	174*	10	4	0	1068*
12	44	40	33	1	22	0	149*	2	54	54	23	11	47	43	27
13	29	0	148*	2	11	0	637*	3	93	90	17	12	44	35	32
14	4	0	876*	3	59	56	24	4	23	0	132*	13	42	39	34
-15, 2, l				4	18	0	201*	5	3	0	934*	14	78	68	22
1	39	34	38	5	15	2	73	6	57	54	21	15	47	43	32
2	25	0	132*	6	54	52	24	7	7	0	425*	16	54	57	30
3	47	46	31	7	13	0	81*	8	4	0	784*	17	40	32	38
4	12	0	108*	8	27	17	42	9	84	79	18	18	4	0	1282*
5	60	50	26	9	22	25	54	10	8	0	442*	-13, 4, l			
6	21	17	61	10	66	62	24	11	81	75	18	1	27	0	137*
7	25	0	145*	11	41	35	34	12	6	0	173*	2	11	0	324*
8	4	0	1021*	12	62	58	27	13	4	0	960*	3	11	0	673*
9	43	38	32	13	4	0	1098*	14	20	28	59	4	63	64	26
10	22	0	161*	14	49	44	29	15	16	9	70	5	4	0	1193*
11	15	0	276*	15	57	51	28	16	4	0	1082*	6	71	60	24
12	25	0	157*	16	54	51	29	17	4	0	972*	7	12	0	600*
-15, 3, l				17	37	33	41	18	18	10	70	8	4	0	902*
1	25	14	55	-14, 3, l				19	4	0	934*	9	16	0	235*
2	7	0	191*	1	11	0	643*	20	38	24	35	10	4	0	1072*
3	18	13	72	2	20	0	190*	21	53	40	27	11	4	0	944*
4	4	0	860*	3	21	0	176*	-13, 2, l				12	23	26	56

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
-13, 4, l				12	35	35	30	-12, 5, l				26	16	0	88*
13	41	40	36	13	23	0	139*	1	4	0	928*	-11, 2, l			
-12, 0, l				14	19	0	186*	2	4	0	1108*	1	10	0	263*
2	3	0	1121*	15	3	0	1135*	3	50	47	30	2	54	58	20
4	101	108	16	16	69	60	22	4	19	5	67	3	3	0	859*
6	22	0	158*	17	48	40	27	5	12	0	553*	4	89	87	16
8	146	148	14	18	24	10	45	6	82	72	21	5	67	69	18
10	55	58	22	19	40	33	35	7	20	0	185*	6	3	0	885*
12	18	6	51	20	26	22	50	8	4	0	1144*	7	23	0	126*
14	70	67	19	21	85	76	23	9	10	0	134*	8	48	52	23
16	30	41	37	22	34	28	45	10	12	0	344*	9	13	0	70*
18	4	0	928*	23	4	0	1179*	11	8	0	476*	10	38	35	26
20	86	83	20	-12, 3, l				-11, 0, l				11	41	40	26
22	83	81	22	1	3	0	1048*	1	36	45	27	12	20	0	124*
24	21	0	188*	2	38	40	29	3	426	433	35	13	79	79	18
-12, 1, l				3	48	45	25	5	3	0	1108*	14	34	37	31
1	89	89	17	4	35	33	33	7	318	321	30	15	16	0	179*
2	74	74	18	5	45	44	27	9	16	0	191*	16	37	42	30
3	46	51	25	6	37	31	29	11	128	122	14	17	74	73	20
4	33	29	31	7	19	4	53	13	31	33	32	18	4	0	881*
5	10	0	314*	8	23	0	158*	15	17	19	58	19	24	12	43
6	3	0	866*	9	3	0	1036*	17	85	78	18	20	41	39	30
7	10	0	299*	10	35	31	31	19	50	47	25	21	4	0	979*
8	10	0	301*	11	13	0	256*	21	18	18	64	22	18	12	69
9	79	73	17	12	7	0	156*	23	80	77	21	23	9	0	154*
10	76	81	19	13	18	8	55	25	79	77	23	24	14	0	253*
11	60	62	21	14	63	60	22	-11, 1, l				25	10	0	139*
12	24	0	136*	15	4	0	1098*	1	150	145	14	-11, 3, l			
13	22	16	50	16	73	68	21	2	73	77	17	1	27	28	38
14	72	71	19	17	17	12	72	3	106	102	14	2	78	78	17
15	4	0	819*	18	87	81	21	4	22	25	41	3	36	33	29
16	68	64	22	19	40	41	38	5	76	72	16	4	93	95	17
17	89	84	18	20	14	0	92*	6	33	35	31	5	106	108	16
18	4	0	722*	21	4	0	1008*	7	115	118	14	6	49	46	23
19	4	0	1025*	-12, 4, l				8	18	0	161*	7	22	26	48
20	24	0	147*	1	21	0	157*	9	230	234	22	8	64	60	19
21	18	13	70	2	3	0	1204*	10	14	0	215*	9	3	0	945*
22	4	0	1231*	3	36	35	32	11	3	0	901*	10	15	0	225*
23	20	0	220*	4	17	0	199*	12	68	71	20	11	21	16	46
24	90	77	22	5	110	108	18	13	38	38	28	12	10	0	587*
-12, 2, l				6	33	37	39	14	21	0	137*	13	31	31	35
1	13	0	74*	7	55	51	26	15	4	0	823*	14	24	0	145*
2	19	0	151*	8	4	0	811*	16	3	0	946*	15	22	0	152*
3	39	41	28	9	19	7	56	17	46	44	25	16	48	40	25
4	3	0	935*	10	4	0	931*	18	34	34	33	17	4	0	1005*
5	37	41	30	11	67	66	23	19	49	45	26	18	27	0	148*
6	16	0	174*	12	13	0	92*	20	10	0	118*	19	50	52	30
7	20	0	147*	13	35	35	39	21	19	0	170*	20	11	0	331*
8	62	61	20	14	39	33	35	22	8	0	446*	21	4	0	960*
9	48	38	23	15	12	0	758*	23	23	0	145*	22	14	0	320*
10	4	0	840*	16	4	0	964*	24	20	10	60	23	28	0	146*
11	38	38	29	17	4	0	1036*	25	7	0	183*	-11, 4, l			

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
-11, 4, l				20	53	46	23	19	31	34	34	18	42	39	34
1	30	22	35	22	4	0	868*	20	51	47	25	19	18	0	225*
2	4	0	920*	24	11	0	668*	21	4	0	857*	20	85	81	22
3	38	39	30	26	4	0	1172*	22	4	0	1049*	21	4	0	1183*
4	119	118	16	28	22	0	166*	23	26	16	42	22	34	31	42
5	43	43	27	-10, 1, l				24	11	0	656*	-10, 5, l			
6	34	37	35	1	144	148	14	25	73	59	23	1	44	37	28
7	24	28	47	2	41	42	24	26	26	0	147*	2	13	0	79*
8	34	34	33	3	260	263	28	-10, 3, l				3	4	0	995*
9	42	45	30	4	80	81	15	1	34	36	29	4	13	0	206*
10	8	0	442*	5	5	0	195*	2	22	0	142*	5	24	0	132*
11	28	28	40	6	156	156	16	3	75	77	16	6	4	0	926*
12	22	0	176*	7	360	365	34	4	3	0	683*	7	40	41	30
13	7	0	164*	8	113	113	14	5	3	0	714*	8	15	10	72
14	21	0	168*	9	113	112	13	6	33	31	30	9	72	70	20
15	27	25	43	10	83	84	16	7	57	58	21	10	11	0	707*
16	23	30	55	11	148	148	14	8	48	48	24	11	32	39	40
17	4	0	840*	12	46	44	21	9	115	116	15	12	44	50	32
18	47	43	30	13	3	0	887*	10	42	38	26	13	31	24	42
19	25	0	158*	14	153	162	16	11	66	66	19	14	23	0	150*
20	21	0	193*	15	54	61	22	12	25	0	137*	15	59	56	26
-11, 5, l				16	3	0	1044*	13	3	0	905*	16	28	19	45
1	4	0	800*	17	22	0	127*	14	44	39	25	17	4	0	812*
2	42	39	29	18	3	0	300*	15	19	8	53	18	19	0	195*
3	7	0	174*	19	33	33	32	16	17	0	186*	-10, 6, l			
4	11	0	325*	20	17	0	206*	17	4	0	817*	1	42	36	33
5	63	66	23	21	68	71	20	18	4	0	963*	2	16	0	205*
6	43	50	32	22	10	0	105*	19	64	60	23	3	21	0	57
7	70	63	23	23	4	0	905*	20	64	68	23	4	23	0	160*
8	11	0	370*	24	59	55	25	21	4	0	1015*	5	4	0	872*
9	14	0	265*	25	44	42	31	22	4	0	916*	6	4	0	1054*
10	12	0	100*	26	12	0	696*	23	40	40	36	7	4	0	836*
11	4	0	1160*	27	4	0	958*	24	4	0	1306*	8	57	46	28
12	25	0	128*	-10, 2, l				25	48	47	31	9	12	0	274*
13	4	0	1074*	1	10	0	79*	-10, 4, l				10	38	29	36
14	12	0	560*	2	68	71	16	1	71	70	19	11	17	13	73
15	4	0	979*	3	16	0	172*	2	10	0	278*	-9, 0, l			
-11, 6, l				4	138	138	14	3	3	0	844*	1	413	417	30
1	25	0	145*	5	201	206	21	4	110	113	16	3	357	365	31
2	17	0	85*	6	26	22	35	5	92	100	17	5	196	203	21
3	51	44	31	7	64	68	17	6	69	68	20	7	245	254	23
4	27	32	54	8	124	128	13	7	44	40	25	9	128	124	12
-10, 0, l				9	180	182	19	8	3	0	951*	11	52	57	18
2	300	306	27	10	14	21	64	9	89	88	18	13	124	135	13
4	225	228	24	11	30	35	32	10	3	0	1064*	15	115	127	13
6	127	130	13	12	96	100	16	11	48	48	26	17	3	0	687*
8	117	114	13	13	3	0	925*	12	41	42	28	19	111	126	15
10	46	47	21	14	23	0	130*	13	33	28	33	21	108	106	15
12	62	54	17	15	19	18	50	14	27	33	43	23	42	46	28
14	103	109	14	16	17	0	164*	15	47	47	26	25	44	48	29
16	148	158	14	17	23	0	150*	16	11	0	619*	27	16	0	235*
18	15	9	64	18	51	48	23	17	40	37	31	29	30	25	47

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-9, 1, l			22	13	0	243*	18	25	0	130*	18	7	0	353*
1	32	32	25	23	26	0	130*	19	22	0	199*	20	3	0	1254*
2	34	38	24	24	18	12	59	20	4	0	781*	22	53	52	23
3	76	77	14	25	24	0	158*	21	11	0	700*	24	61	55	21
4	43	40	20	26	22	0	151*	22	4	0	1057*	26	72	58	21
5	2	0	1064*	27	7	0	190*	23	74	74	24	28	49	48	31
6	196	207	20	28	4	0	961*	24	39	39	38	30	17	0	203*
7	277	290	27		-9, 3, l				-9, 5, l				-8, 1, l		
8	35	39	27	1	124	129	13	1	3	0	979*	1	90	92	12
9	53	44	18	2	125	134	13	2	39	40	31	2	46	43	17
10	194	202	20	3	25	28	35	3	4	0	811*	3	16	0	146*
11	137	140	14	4	3	0	892*	4	3	0	981*	4	97	98	12
12	93	89	14	5	145	157	15	5	4	0	921*	5	114	122	13
13	89	92	14	6	14	0	187*	6	3	0	1002*	6	79	81	13
14	41	44	23	7	91	92	15	7	4	0	903*	7	168	174	17
15	41	52	25	8	272	273	30	8	88	77	18	8	87	85	13
16	51	60	20	9	111	116	14	9	59	59	22	9	23	28	33
17	58	60	20	10	78	81	16	10	22	0	159*	10	185	189	19
18	3	0	811*	11	20	0	133*	11	11	0	578*	11	155	154	15
19	47	55	24	12	102	102	15	12	33	33	36	12	184	190	18
20	11	0	86*	13	19	0	147*	13	4	0	927*	13	9	0	525*
21	36	34	31	14	30	33	36	14	60	58	24	14	12	0	68*
22	4	0	815*	15	21	22	47	15	4	0	907*	15	20	11	38
23	49	48	25	16	84	83	17	16	28	15	41	16	5	0	155*
24	4	0	1046*	17	4	0	854*	17	11	0	596*	17	25	39	40
25	18	6	61	18	91	93	17	18	25	14	49	18	97	103	15
26	29	35	41	19	36	29	32	19	32	31	42	19	95	104	15
27	18	0	217*	20	38	32	31	20	4	0	1112*	20	22	0	127*
28	43	38	35	21	23	28	51		-9, 6, l			21	3	0	917*
29	4	0	862*	22	6	0	181*	1	11	0	333*	22	66	68	20
	-9, 2, l			23	24	20	50	2	52	48	26	23	48	51	24
1	45	48	21	24	4	0	1134*	3	4	0	809*	24	17	0	191*
2	94	93	14	25	70	61	25	4	39	40	34	25	80	80	20
3	172	171	17	26	4	0	973*	5	53	38	27	26	44	36	29
4	319	318	27		-9, 4, l			6	28	0	155*	27	65	64	23
5	9	0	665*	1	25	30	39	7	4	0	885*	28	49	46	30
6	79	83	16	2	85	80	16	8	24	15	52	29	24	17	57
7	271	271	28	3	14	0	216*	9	35	31	35	30	41	33	35
8	261	268	28	4	37	41	28	10	22	0	161*		-8, 2, l		
9	80	81	16	5	49	52	24	11	4	0	870*	1	10	0	214*
10	8	0	106*	6	106	107	15	12	12	0	323*	2	63	61	15
11	187	192	17	7	31	26	35	13	4	0	883*	3	19	17	40
12	200	204	20	8	75	75	18	14	21	0	183*	4	117	122	12
13	187	196	18	9	35	37	30		-8, 0, l			5	85	89	13
14	108	113	14	10	85	88	17	2	50	51	15	6	101	100	12
15	16	6	59	11	107	107	16	4	17	0	120*	7	34	39	26
16	109	101	15	12	3	0	854*	6	147	144	15	8	152	153	15
17	40	41	27	13	4	0	853*	8	27	35	28	9	7	0	125*
18	34	39	29	14	4	0	760*	10	17	0	120*	10	355	365	32
19	3	0	923*	15	7	0	440*	12	52	56	17	11	83	81	15
20	19	16	51	16	13	0	223*	14	96	99	13	12	104	105	13
21	44	35	26	17	20	17	53	16	3	0	873*	13	71	69	16

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-8,	2, l		6	122	124	14	9	4	0	843*	18	21	0	127*
14	59	64	18	7	3	0	1080*	10	48	48	27	19	84	83	15
15	85	88	16	8	60	59	19	11	4	0	1017*	20	3	0	1028*
16	193	198	20	9	120	123	15	12	26	0	135*	21	43	42	24
17	150	155	14	10	94	104	16	13	58	53	26	22	17	25	57
18	17	0	167*	11	3	0	797*	14	14	0	263*	23	16	0	174*
19	3	0	879*	12	53	57	21	15	7	0	184*	24	30	24	36
20	101	99	16	13	52	52	22	16	45	40	29	25	17	0	186*
21	36	33	31	14	54	60	22	17	4	0	810*	26	83	77	19
22	7	0	424*	15	64	63	20		-8,	7, l		27	4	0	821*
23	18	0	215*	16	4	0	836*	1	8	0	461*	28	11	0	657*
24	4	0	835*	17	11	0	575*	2	20	13	71	29	38	37	37
25	4	0	856*	18	56	51	23	3	21	0	178*	30	23	28	59
26	17	9	65	19	40	34	28	4	17	0	239*	31	42	34	34
27	4	0	867*	20	4	0	755*	5	71	70	26		-7,	2, l	
28	44	45	32	21	11	0	640*	6	25	0	160*	1	20	0	104*
29	79	76	24	22	78	71	23	7	8	0	454*	2	60	55	13
	-8,	3, l		23	34	35	43		-7,	0, l		3	82	85	12
1	40	44	24	24	48	37	30	1	178	180	19	4	182	183	19
2	91	95	14	25	39	38	38	3	305	293	26	5	180	185	19
3	34	35	26		-8,	5, l		5	108	105	11	6	87	84	12
4	109	107	13	1	100	104	17	7	344	348	27	7	172	178	17
5	118	116	13	2	14	0	233*	9	16	18	36	8	90	97	13
6	55	60	19	3	42	42	27	11	21	19	31	9	169	173	17
7	40	37	26	4	7	0	438*	13	184	186	20	10	105	109	12
8	376	385	34	5	115	120	16	15	50	42	18	11	227	230	23
9	25	0	132*	6	6	0	175*	17	57	56	17	12	193	192	20
10	90	89	15	7	30	30	37	19	30	13	28	13	161	158	16
11	136	139	13	8	18	0	154*	21	92	93	15	14	187	190	18
12	184	179	19	9	44	46	26	23	48	57	24	15	11	0	239*
13	79	81	16	10	3	0	930*	25	4	0	812*	16	3	0	802*
14	3	0	870*	11	50	47	24	27	148	141	16	17	3	0	940*
15	61	61	19	12	4	0	793*	29	113	107	20	18	98	94	14
16	104	100	16	13	50	49	25	31	8	0	448*	19	3	0	990*
17	37	33	30	14	55	52	23		-7,	1, l		20	3	0	868*
18	97	99	16	15	26	18	40	1	78	80	11	21	51	55	22
19	23	13	46	16	4	0	1023*	2	173	175	18	22	23	0	120*
20	23	0	175*	17	16	21	71	3	33	30	19	23	40	40	31
21	63	65	21	18	31	22	40	4	165	177	17	24	51	52	24
22	98	97	18	19	27	18	44	5	292	291	27	25	47	50	28
23	100	100	18	20	16	0	217*	6	39	28	17	26	42	41	30
24	4	0	1126*	21	66	59	25	7	172	171	17	27	4	0	792*
25	45	42	32	22	22	0	157*	8	13	9	47	28	85	81	21
26	42	39	35		-8,	6, l		9	48	48	16	29	40	36	38
27	4	0	985*	1	4	0	1042*	10	52	52	16	30	4	0	1065*
28	56	53	29	2	19	0	181*	11	46	46	18		-7,	3, l	
	-8,	4, l		3	63	65	22	12	2	0	956*	1	37	33	22
1	17	4	51	4	4	0	927*	13	111	113	12	2	30	33	27
2	126	130	13	5	21	0	177*	14	20	0	113*	3	18	27	44
3	142	149	14	6	62	60	22	15	19	0	137*	4	109	112	12
4	91	95	15	7	32	28	37	16	3	0	1001*	5	17	0	134*
5	136	138	13	8	36	41	37	17	88	101	14	6	13	0	218*

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-7,	3, l		1	3	0	914*	9	17	0	244*	32	30	32	48
7	366	374	32	2	67	67	19	10	55	52	28		-6,	2, l	
8	281	285	27	3	50	58	24	11	12	0	339*	1	263	270	22
9	2	0	1286*	4	81	79	17		-6,	0, l		2	73	80	11
10	115	112	13	5	3	0	852*	2	327	323	23	3	138	138	14
11	107	110	13	6	39	40	27	4	88	87	10	4	2	0	789*
12	56	50	18	7	3	0	832*	6	95	92	9	5	69	68	11
13	8	0	106*	8	52	54	23	8	180	177	19	6	63	65	12
14	117	123	13	9	64	56	19	10	12	18	42	7	44	45	16
15	41	39	24	10	44	44	25	12	120	127	11	8	21	18	29
16	131	135	13	11	16	0	180*	14	51	50	16	9	12	0	171*
17	93	89	15	12	4	0	877*	16	102	100	12	10	17	10	40
18	57	52	21	13	21	0	170*	18	58	53	16	11	128	129	12
19	14	0	186*	14	22	0	134*	20	22	0	113*	12	71	71	14
20	64	69	20	15	16	10	61	22	44	42	22	13	84	87	13
21	82	87	18	16	41	41	28	24	55	56	22	14	37	37	24
22	15	0	222*	17	21	18	51	26	85	83	18	15	20	0	136*
23	31	33	38	18	4	0	801*	28	70	63	22	16	144	141	13
24	42	34	29	19	25	32	46	30	134	124	18	17	43	45	21
25	55	52	24	20	15	0	79*	32	22	0	179*	18	86	94	15
26	43	45	34	21	8	0	470*		-6,	1, l		19	54	58	19
27	31	27	42	22	78	78	24	1	150	147	15	20	101	110	14
28	4	0	943*	23	15	0	94*	2	95	90	10	21	141	140	14
29	27	24	52		-7,	6, l		3	176	175	18	22	3	0	986*
	-7,	4, l		1	6	0	160*	4	96	101	10	23	3	0	900*
1	107	109	13	2	24	0	153*	5	306	296	25	24	4	0	781*
2	77	73	15	3	17	0	205*	6	14	19	37	25	49	45	26
3	61	60	17	4	4	0	992*	7	164	159	17	26	60	53	23
4	22	0	120*	5	8	0	436*	8	82	82	10	27	36	35	33
5	55	56	19	6	24	16	45	9	180	173	18	28	4	0	985*
6	65	67	17	7	18	18	58	10	96	95	10	29	4	0	879*
7	18	0	148*	8	48	50	27	11	122	115	12	30	17	0	224*
8	73	71	16	9	89	89	19	12	69	67	13	31	48	51	33
9	90	89	15	10	4	0	955*	13	20	21	36		-6,	3, l	
10	24	16	35	11	22	12	49	14	185	187	18	1	51	50	16
11	3	0	706*	12	21	0	164*	15	20	0	106*	2	27	24	26
12	51	44	21	13	23	25	50	16	78	79	14	3	73	69	13
13	110	110	15	14	4	0	850*	17	92	94	13	4	53	52	16
14	22	0	146*	15	4	0	892*	18	56	57	17	5	150	156	15
15	42	38	25	16	20	18	62	19	11	0	72*	6	42	42	20
16	3	0	1037*	17	3	0	446*	20	9	0	1628*	7	57	58	16
17	53	48	23	18	4	0	1140*	21	46	43	22	8	15	0	168*
18	35	35	33	19	22	0	167*	22	129	131	14	9	97	99	13
19	11	0	276*		-7,	7, l		23	69	64	18	10	69	72	16
20	21	15	50	1	4	0	935*	24	46	42	25	11	182	188	17
21	19	0	149*	2	4	0	1046*	25	45	43	26	12	22	0	135*
22	40	35	31	3	40	33	35	26	37	34	33	13	3	0	1051*
23	79	75	21	4	23	22	58	27	78	75	20	14	326	324	28
24	23	0	154*	5	12	0	99*	28	34	41	37	15	22	11	41
25	23	19	57	6	27	29	50	29	93	91	19	16	40	36	24
26	25	20	53	7	30	20	45	30	4	0	937*	17	63	57	18
	-7,	5, l		8	19	0	223*	31	62	59	27	18	176	183	17

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-6,	3, l		12	19	0	168*	7	679	658	23	5	101	98	10
19	18	0	143*	13	15	18	68	9	705	677	24	6	146	147	15
20	3	0	827*	14	30	27	35	11	598	579	26	7	124	126	12
21	53	49	23	15	38	32	28	13	233	232	23	8	155	156	16
22	84	81	18	16	52	54	24	15	26	21	29	9	40	45	15
23	10	0	560*	17	66	63	22	17	74	78	14	10	155	154	15
24	18	15	60	18	3	0	1332*	19	105	104	13	11	126	126	13
25	55	47	23	19	45	46	29	21	14	0	171*	12	81	83	12
26	35	29	35	20	4	0	927*	23	51	51	21	13	105	107	12
27	27	31	47	21	21	0	158*	25	41	44	26	14	13	0	191*
28	4	0	1065*	22	16	0	203*	27	44	45	27	15	124	128	12
29	22	0	155*	23	4	0	925*	29	50	46	27	16	110	116	13
	-6,	4, l		24	19	0	219*	31	95	89	22	17	348	344	33
1	3	0	1050*		-6,	6, l		33	4	0	1075*	18	273	279	28
2	41	40	22	1	24	0	129*		-5,	1, l		19	157	161	16
3	201	202	20	2	4	0	872*	1	175	170	17	20	73	76	17
4	58	61	18	3	15	0	194*	2	4	0	381*	21	18	13	51
5	183	184	19	4	33	25	32	3	286	289	23	22	20	17	47
6	36	39	28	5	61	61	21	4	119	123	12	23	87	90	17
7	178	176	17	6	27	17	40	5	263	249	23	24	3	0	855*
8	158	156	16	7	55	59	24	6	12	0	142*	25	94	93	17
9	240	246	23	8	6	0	175*	7	110	107	11	26	39	40	31
10	129	131	13	9	78	79	19	8	440	433	24	27	4	0	964*
11	95	97	15	10	4	0	867*	9	287	281	25	28	16	0	212*
12	156	155	15	11	4	0	842*	10	42	41	15	29	4	0	838*
13	51	52	21	12	26	21	43	11	145	143	14	30	4	0	907*
14	80	85	16	13	19	0	187*	12	39	49	17	31	4	0	903*
15	28	28	35	14	11	0	625*	13	347	341	28		-5,	3, l	
16	3	0	917*	15	56	50	24	14	38	32	21	1	137	136	14
17	131	134	15	16	50	47	27	15	153	155	16	2	25	27	25
18	55	49	22	17	28	0	165*	16	196	196	20	3	2	0	815*
19	4	0	875*	18	8	0	464*	17	84	87	14	4	59	49	13
20	37	39	31	19	58	54	27	18	94	98	14	5	77	81	11
21	13	0	235*	20	19	0	195*	19	119	120	13	6	174	170	17
22	19	0	227*		-6,	7, l		20	39	43	23	7	112	108	12
23	15	16	72	1	4	0	1108*	21	110	111	14	8	13	0	176*
24	69	67	23	2	33	32	42	22	72	69	17	9	53	52	16
25	60	56	26	3	4	0	1026*	23	36	41	30	10	139	140	14
26	65	58	25	4	25	0	138*	24	38	31	28	11	90	82	14
27	7	0	184*	5	44	39	31	25	16	0	206*	12	359	370	32
	-6,	5, l		6	4	0	1007*	26	3	0	951*	13	118	120	12
1	160	160	15	7	26	0	132*	27	64	57	21	14	47	57	21
2	89	90	16	8	46	44	31	28	23	0	151*	15	106	106	13
3	200	198	19	9	42	45	37	29	4	0	876*	16	22	26	42
4	18	0	173*	10	55	52	30	30	54	53	27	17	123	124	13
5	3	0	1051*	11	27	24	50	31	4	0	1090*	18	70	76	16
6	35	33	30	12	8	0	483*	32	8	0	470*	19	48	46	21
7	180	183	18	13	29	24	46		-5,	2, l		20	10	0	310*
8	3	0	976*		-5,	0, l		1	311	309	25	21	3	0	840*
9	20	0	147*	1	352	355	21	2	374	362	24	22	39	36	28
10	20	0	158*	3	393	399	21	3	85	76	10	23	15	0	198*
11	101	96	16	5	100	99	9	4	211	215	21	24	80	73	19

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-5, 3, l			16	18	0	155*	8	586	575	22	5	201	198	20
25	3	0	1190*	17	70	75	20	10	697	685	23	6	271	276	24
26	53	51	26	18	4	0	970*	12	30	34	19	7	2	0	894*
27	16	0	191*	19	20	0	181*	14	366	351	27	8	2	0	803*
28	24	0	149*	20	4	0	1036*	16	247	242	24	9	48	41	13
29	4	0	1300*	21	21	0	176*	18	35	38	23	10	86	84	10
30	57	52	29	22	8	0	511*	20	213	214	22	11	11	0	148*
	-5, 4, l			23	4	0	1241*	22	113	113	13	12	194	185	20
1	46	47	19	24	18	0	180*	24	109	110	15	13	46	50	17
2	110	111	12	25	4	0	987*	26	91	94	17	14	83	84	13
3	221	221	23		-5, 6, l			28	51	53	26	15	15	0	157*
4	29	34	29	1	75	67	19	30	75	74	21	16	65	65	15
5	12	0	222*	2	102	106	16	32	12	0	682*	17	140	140	14
6	128	131	14	3	95	98	17		-4, 1, l			18	156	162	14
7	153	151	15	4	23	0	127*	1	33	35	15	19	3	0	1020*
8	267	277	28	5	3	0	974*	2	29	32	16	20	75	83	17
9	116	117	13	6	27	29	39	3	336	335	20	21	167	171	16
10	179	176	18	7	7	0	421*	4	40	40	12	22	42	37	25
11	182	185	19	8	115	115	16	5	356	366	21	23	3	0	854*
12	3	0	833*	9	10	0	380*	6	307	304	22	24	69	66	18
13	77	81	16	10	35	23	31	7	130	126	13	25	45	41	27
14	95	104	15	11	50	51	25	8	462	457	23	26	75	76	20
15	9	0	91*	12	35	42	34	9	385	385	24	27	55	66	25
16	95	99	16	13	19	0	167*	10	150	150	15	28	50	47	24
17	3	0	1030*	14	13	0	229*	11	90	80	9	29	4	0	900*
18	3	0	848*	15	57	57	24	12	18	0	105*	30	14	0	276*
19	34	37	32	16	22	0	151*	13	41	41	15	31	34	30	39
20	24	0	139*	17	4	0	1042*	14	159	158	16	32	44	40	34
21	31	31	38	18	48	40	29	15	86	88	12		-4, 3, l		
22	70	70	20	19	29	19	47	16	152	161	16	1	37	28	17
23	35	37	35	20	50	47	30	17	48	46	19	2	244	237	22
24	25	24	47	21	27	0	152*	18	57	53	19	3	190	187	20
25	29	20	42		-5, 7, l			19	195	197	20	4	99	104	10
26	41	36	35	1	36	42	35	20	61	59	18	5	333	334	27
27	18	0	206*	2	21	0	179*	21	101	102	14	6	117	112	11
28	40	35	34	3	46	44	27	22	205	208	20	7	337	332	27
	-5, 5, l			4	16	0	225*	23	132	130	14	8	67	68	13
1	89	94	15	5	68	63	22	24	7	0	453*	9	60	57	15
2	101	113	14	6	25	29	48	25	70	70	19	10	253	247	25
3	58	56	20	7	24	0	155*	26	3	0	1035*	11	127	126	12
4	211	212	21	8	11	0	682*	27	40	47	31	12	280	293	25
5	38	46	27	9	4	0	956*	28	20	0	164*	13	105	104	12
6	195	198	20	10	26	0	147*	29	72	74	22	14	113	118	12
7	66	67	18	11	43	42	32	30	4	0	940*	15	60	60	18
8	52	49	21	12	19	18	68	31	4	0	983*	16	79	77	15
9	96	99	15	13	69	60	25	32	43	38	34	17	112	111	13
10	23	22	43	14	35	36	40	33	31	26	45	18	154	162	16
11	76	78	17	15	4	0	1024*		-4, 2, l			19	99	106	15
12	14	0	203*		-4, 0, l			1	57	58	11	20	29	26	32
13	47	43	23	2	215	213	20	2	598	599	23	21	19	0	141*
14	25	27	40	4	218	220	20	3	173	174	17	22	3	0	969*
15	39	38	29	6	554	546	21	4	34	40	15	23	54	57	22

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-4,	3, l		13	66	66	19	15	54	50	29	32	28	17	46
24	54	52	23	14	85	79	17	16	22	0	176*	33	4	0	816*
25	15	0	218*	15	68	71	19		-3,	0, l			-3,	2, l	
26	4	0	955*	16	10	0	598*	1	604	617	16	1	544	558	21
27	67	65	22	17	4	0	879*	3	450	443	17	2	172	173	18
28	14	0	267*	18	13	0	80*	5	154	157	16	3	156	152	15
29	4	0	962*	19	89	87	18	7	157	161	15	4	112	111	12
30	4	0	802*	20	24	0	153*	9	329	335	21	5	185	175	19
31	15	0	84*	21	62	57	23	11	371	365	23	6	118	124	12
	-4,	4, l		22	40	33	32	13	368	360	25	7	70	71	10
1	42	40	19	23	53	51	28	15	144	139	13	8	186	192	20
2	157	152	15	24	18	0	227*	17	253	258	25	9	207	203	21
3	71	74	14	25	12	0	675*	19	525	530	31	10	156	151	15
4	155	156	15	26	4	0	1106*	21	94	91	14	11	69	61	11
5	167	164	16		-4,	6, l		23	65	66	17	12	54	58	13
6	101	102	12	1	97	102	17	25	150	146	15	13	9	0	197*
7	2	0	1013*	2	39	39	29	27	157	160	16	14	109	110	11
8	76	76	14	3	20	0	147*	29	19	0	170*	15	53	53	16
9	19	22	43	4	132	131	15	31	4	0	1064*	16	265	274	25
10	66	64	15	5	22	0	162*	33	26	0	143*	17	90	87	13
11	3	0	1043*	6	137	139	15		-3,	1, l		18	28	34	31
12	179	183	18	7	30	26	34	1	48	41	10	19	3	0	1091*
13	81	80	15	8	41	42	28	2	453	463	18	20	23	0	125*
14	80	83	15	9	51	51	24	3	453	457	18	21	151	151	14
15	45	42	22	10	87	85	18	4	24	25	19	22	157	164	14
16	83	83	16	11	54	52	24	5	628	564	19	23	40	43	26
17	160	160	16	12	30	29	37	6	261	242	21	24	14	0	203*
18	32	23	32	13	4	0	942*	7	224	216	21	25	3	0	969*
19	7	0	431*	14	4	0	973*	8	286	300	22	26	55	55	24
20	21	0	138*	15	43	35	29	9	191	188	19	27	35	36	33
21	55	53	24	16	31	29	37	10	542	520	23	28	38	38	32
22	3	0	1100*	17	28	30	41	11	178	175	18	29	44	44	31
23	8	0	137*	18	28	26	46	12	33	37	17	30	50	50	29
24	3	0	1123*	19	86	77	21	13	375	367	26	31	25	0	172*
25	8	0	387*	20	12	0	336*	14	133	131	13	32	36	29	40
26	28	0	147*	21	7	0	193*	15	88	91	11		-3,	3, l	
27	16	0	233*	22	15	0	243*	16	125	118	13	1	434	418	25
28	34	29	41		-4,	7, l		17	46	50	18	2	165	154	17
29	5	0	271*	1	36	33	35	18	73	79	15	3	120	125	12
	-4,	5, l		2	50	42	25	19	40	40	24	4	40	40	15
1	75	72	16	3	19	0	178*	20	270	270	28	5	276	264	26
2	24	28	35	4	33	24	35	21	504	503	34	6	183	182	17
3	73	78	16	5	43	38	28	22	72	75	19	7	49	51	13
4	118	115	13	6	34	33	35	23	92	97	15	8	11	3	50
5	52	52	19	7	4	0	964*	24	71	70	19	9	120	121	13
6	40	39	23	8	48	46	28	25	89	93	18	10	159	160	16
7	18	27	48	9	27	20	43	26	71	68	20	11	201	200	19
8	110	111	14	10	26	22	51	27	104	106	17	12	186	186	18
9	34	38	27	11	21	0	176*	28	52	53	25	13	127	124	13
10	59	56	18	12	4	0	1007*	29	26	24	47	14	66	67	15
11	22	18	41	13	44	34	31	30	11	0	328*	15	107	100	13
12	12	0	83*	14	22	0	192*	31	9	0	140*	16	169	165	16

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-3, 3, l			6	7	0	117*	8	48	47	25	19	213	215	21
17	191	194	18	7	13	0	209*	9	51	48	26	20	189	188	17
18	33	31	29	8	5	0	150*	10	4	0	928*	21	14	0	205*
19	38	38	24	9	3	0	797*	11	11	0	335*	22	35	32	28
20	48	50	21	10	18	17	46	12	46	49	32	23	76	80	17
21	19	0	142*	11	19	0	120*	13	4	0	1054*	24	97	100	15
22	67	63	19	12	7	0	421*	14	14	0	275*	25	76	72	18
23	29	31	37	13	69	66	18	15	13	0	91*	26	110	108	16
24	76	72	19	14	94	92	16	16	17	0	209*	27	3	0	1187*
25	3	0	1061*	15	115	115	15	17	4	0	1007*	28	40	43	32
26	41	40	29	16	65	60	20		-3, 8, l			29	49	44	28
27	4	0	824*	17	81	87	18	1	4	0	990*	30	14	0	85*
28	14	0	234*	18	57	57	22	2	4	0	1179*	31	11	0	603*
29	4	0	949*	19	75	78	20	3	22	0	162*	32	4	0	887*
30	29	0	157*	20	4	0	977*	4	31	34	45	33	7	0	197*
31	19	0	200*	21	26	26	46	5	4	0	861*		-2, 2, l		
	-3, 4, l			22	84	78	20		-2, 0, l			1	331	338	20
1	48	46	17	23	16	0	222*	2	988	1004	61	2	246	245	21
2	163	166	17	24	50	49	30	4	114	115	11	3	339	352	20
3	2	0	1240*	25	29	19	45	6	191	211	18	4	343	339	21
4	102	99	12	26	8	0	483*	8	105	106	11	5	270	265	21
5	62	67	15		-3, 6, l			10	90	88	9	6	114	108	12
6	56	55	16	1	82	77	17	12	545	547	23	7	260	271	22
7	202	199	20	2	141	143	16	14	84	81	10	8	47	45	12
8	151	150	15	3	49	44	24	16	152	158	16	9	2	0	903*
9	31	29	25	4	50	48	23	18	495	500	30	10	392	376	24
10	68	67	15	5	38	44	29	20	64	53	18	11	280	274	26
11	136	133	14	6	43	45	26	22	325	320	28	12	165	165	16
12	113	112	13	7	56	49	22	24	88	84	16	13	47	51	14
13	36	31	24	8	89	88	17	26	25	0	131*	14	112	117	11
14	75	72	14	9	6	0	180*	28	104	102	17	15	253	255	24
15	3	0	1079*	10	32	35	34	30	51	48	26	16	90	90	12
16	244	251	23	11	16	0	178*	32	93	95	22	17	149	149	14
17	48	49	22	12	21	0	129*		-2, 1, l			18	100	97	13
18	112	116	15	13	27	15	36	1	92	92	9	19	99	98	13
19	24	0	141*	14	8	0	499*	2	187	186	17	20	56	55	19
20	24	0	130*	15	22	30	53	3	177	177	15	21	207	205	19
21	33	40	34	16	38	30	31	4	217	220	18	22	18	16	51
22	10	0	294*	17	13	0	83*	5	274	288	18	23	3	0	845*
23	16	15	65	18	34	32	36	6	12	0	108*	24	3	0	1023*
24	8	0	146*	19	28	30	48	7	83	83	8	25	62	64	21
25	24	27	49	20	16	0	227*	8	1	0	978*	26	39	28	29
26	58	59	25	21	30	31	47	9	371	367	21	27	21	0	162*
27	4	0	1195*	22	21	0	197*	10	190	195	19	28	38	37	33
28	36	33	39		-3, 7, l			11	107	108	11	29	4	0	1128*
29	17	0	223*	1	12	0	90*	12	306	297	24	30	3	0	1307*
	-3, 5, l			2	38	38	32	13	127	129	13	31	28	0	165*
1	48	40	19	3	25	31	47	14	193	192	19	32	4	0	943*
2	76	75	15	4	29	35	40	15	168	165	17		-2, 3, l		
3	3	0	964*	5	60	60	23	16	168	165	17	1	184	174	18
4	53	52	18	6	22	0	169*	17	58	64	15	2	87	85	9
5	105	100	13	7	4	0	1056*	18	146	141	15	3	19	0	97*

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-2, 3, l			23	4	0	935*	18	70	71	22	3	1064	1071	67
4	251	252	21	24	23	3	47	19	4	0	812*	4	256	267	16
5	343	335	25	25	64	61	23	20	22	18	55	5	886	888	74
6	192	183	19	26	44	46	31	21	4	0	1108*	6	73	70	8
7	2	0	760*	27	25	0	142*	22	50	43	30	7	412	399	19
8	137	133	12	28	26	0	182*	23	4	0	817*	8	371	368	20
9	91	94	10	29	4	0	979*		-2, 7, l			9	309	297	21
10	2	0	835*		-2, 5, l			1	106	105	18	10	127	127	13
11	4	0	140*	1	238	237	25	2	57	59	24	11	2	0	807*
12	56	56	15	2	96	97	13	3	51	54	26	12	101	98	11
13	56	60	16	3	30	32	28	4	30	35	41	13	60	57	11
14	3	0	885*	4	32	27	25	5	74	74	21	14	254	247	21
15	40	42	21	5	9	0	270*	6	22	0	143*	15	92	92	10
16	22	24	37	6	58	57	17	7	22	14	51	16	30	17	22
17	54	65	19	7	34	34	25	8	4	0	967*	17	140	135	13
18	13	0	223*	8	3	0	944*	9	46	50	28	18	89	92	13
19	4	0	192*	9	33	28	26	10	4	0	969*	19	17	0	165*
20	42	43	22	10	21	0	111*	11	16	0	216*	20	127	125	12
21	3	0	822*	11	35	37	27	12	37	34	36	21	39	36	24
22	3	0	989*	12	40	47	24	13	81	76	22	22	250	255	25
23	54	49	22	13	67	64	18	14	26	0	137*	23	72	70	18
24	14	0	228*	14	57	58	20	15	23	0	140*	24	41	41	27
25	3	0	954*	15	34	29	30	16	7	0	188*	25	74	71	18
26	3	0	1020*	16	121	121	15	17	34	27	40	26	63	64	22
27	32	28	36	17	37	34	29		-2, 8, l			27	22	0	160*
28	31	26	36	18	3	0	919*	1	37	26	36	28	95	94	18
29	4	0	1104*	19	48	50	26	2	35	36	41	29	11	0	721*
30	59	59	27	20	89	93	19	3	4	0	863*	30	32	28	38
31	21	0	201*	21	51	54	26	4	12	0	617*	31	34	28	38
	-2, 4, l			22	22	0	163*	5	12	0	320*	32	35	34	42
1	40	45	19	23	96	89	20	6	26	16	51	33	63	63	27
2	43	44	17	24	4	0	986*		-1, 0, l				-1, 2, l		
3	9	0	211*	25	15	0	84*	1	1026	1021	45	1	362	366	19
4	11	0	184*	26	4	0	1021*	3	116	118	12	2	431	423	19
5	63	63	14		-2, 6, l			5	179	181	16	3	242	238	20
6	3	0	267*	1	141	144	14	7	341	339	17	4	91	89	9
7	32	37	24	2	3	0	1112*	9	159	155	17	5	233	221	21
8	8	0	287*	3	3	0	1064*	11	6	0	256*	6	127	126	12
9	151	155	14	4	80	80	17	13	114	116	11	7	23	33	21
10	54	49	17	5	78	83	18	15	274	269	26	8	27	25	18
11	120	123	14	6	71	68	19	17	106	109	11	9	297	278	23
12	72	67	15	7	113	110	16	19	120	116	12	10	113	106	11
13	65	72	16	8	93	90	16	21	379	376	33	11	204	196	21
14	3	0	929*	9	60	63	22	23	138	139	14	12	44	51	15
15	9	0	293*	10	21	21	49	25	343	335	31	13	175	170	18
16	24	20	34	11	45	47	26	27	141	140	15	14	60	56	13
17	100	98	14	12	52	57	24	29	46	45	29	15	45	46	18
18	67	67	17	13	4	0	839*	31	24	0	153*	16	138	134	14
19	3	0	1082*	14	18	0	171*	33	47	43	34	17	8	0	684*
20	87	92	18	15	22	28	50		-1, 1, l			18	50	49	19
21	85	81	18	16	7	0	153*	1	56	54	8	19	150	154	16
22	23	13	43	17	21	0	131*	2	161	168	13	20	48	53	21

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	-1, 2, l			7	133	132	14	2	60	65	21	8	274	269	19
21	16	0	171*	8	85	82	13	3	31	28	33	10	117	118	12
22	122	119	13	9	71	67	14	4	3	0	927*	12	246	237	23
23	124	124	14	10	15	0	159*	5	79	83	18	14	100	103	9
24	17	13	59	11	74	77	14	6	56	55	21	16	19	0	107*
25	139	139	15	12	10	0	237*	7	10	0	103*	18	34	31	25
26	46	47	27	13	91	93	13	8	74	80	19	20	85	82	15
27	18	0	181*	14	3	0	844*	9	26	28	42	22	83	77	15
28	35	41	35	15	98	100	14	10	3	0	1048*	24	251	255	24
29	31	28	37	16	3	0	1020*	11	18	8	51	26	38	29	31
30	22	0	155*	17	13	0	200*	12	10	0	535*	28	98	95	18
31	24	0	164*	18	42	40	23	13	13	0	247*	30	16	22	76
32	30	21	46	19	39	45	29	14	44	41	26	32	4	0	1143*
	-1, 3, l			20	162	164	17	15	31	30	38		0, 1, l		
1	355	351	24	21	68	63	20	16	4	0	1031*	1	504	521	13
2	2	0	971*	22	66	71	22	17	45	47	29	2	767	780	61
3	325	323	24	23	70	77	21	18	46	44	29	3	382	387	15
4	7	0	260*	24	74	74	21	19	47	55	30	4	292	304	16
5	181	168	18	25	8	0	446*	20	54	56	28	5	1265	1273	74
6	294	296	25	26	4	0	1038*	21	4	0	986*	6	930	946	78
7	95	95	10	27	7	0	198*	22	34	24	39	7	404	414	19
8	184	176	19	28	53	53	29	23	22	0	167*	8	392	373	20
9	14	0	126*	29	4	0	1029*		-1, 7, l			9	672	663	21
10	138	133	15		-1, 5, l			1	49	49	25	10	111	108	12
11	27	21	23	1	50	57	23	2	56	47	23	11	143	146	14
12	49	48	16	2	38	43	24	3	27	23	41	12	92	92	9
13	4	0	155*	3	90	88	13	4	8	0	407*	13	28	23	20
14	99	95	12	4	46	52	20	5	4	0	855*	14	174	167	17
15	83	86	13	5	18	0	131*	6	31	37	38	15	143	143	15
16	3	0	929*	6	103	104	13	7	7	0	169*	16	102	101	11
17	99	95	13	7	50	46	19	8	4	0	957*	17	195	187	20
18	84	91	14	8	3	0	1133*	9	16	0	222*	18	33	33	26
19	18	0	145*	9	3	0	924*	10	4	0	966*	19	22	0	116*
20	78	82	16	10	3	0	873*	11	40	42	32	20	79	79	15
21	125	126	13	11	38	44	24	12	30	29	40	21	164	169	16
22	3	0	880*	12	70	70	16	13	4	0	1018*	22	172	175	17
23	37	31	29	13	24	0	123*	14	52	50	30	23	23	0	149*
24	77	78	18	14	57	53	20	15	27	32	49	24	23	0	150*
25	36	34	33	15	33	37	31	16	4	0	832*	25	52	59	24
26	4	0	949*	16	78	77	18	17	27	16	48	26	92	90	17
27	22	15	50	17	56	62	23		-1, 8, l			27	71	66	21
28	5	0	211*	18	3	0	911*	1	29	0	153*	28	38	36	34
29	4	0	877*	19	63	64	22	2	32	22	42	29	42	41	32
30	96	94	20	20	112	111	17	3	4	0	1053*	30	29	15	41
31	7	0	203*	21	104	109	18	4	32	23	42	31	64	67	24
	-1, 4, l			22	4	0	1107*	5	12	0	335*	32	31	32	46
1	10	0	62*	23	9	0	131*	6	10	0	140*	33	42	37	37
2	144	140	14	24	27	28	48	7	12	0	115*		0, 2, l		
3	98	94	11	25	4	0	1131*		0, 0, l			0	663	670	19
4	157	159	16	26	93	85	21	2	226	217	10	1	163	167	16
5	38	40	19		-1, 6, l			4	979	985	58	2	285	279	19
6	90	91	12	1	83	81	17	6	800	799	69	3	427	431	19

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	0,	2, l		22	94	95	17	11	45	52	22	11	48	47	28
4	339	337	20	23	3	0	1051*	12	13	0	70*	12	8	0	460*
5	401	397	20	24	42	43	30	13	54	62	21	13	53	46	28
6	2	0	986*	25	27	28	44	14	69	77	19	14	18	25	77
7	581	579	22	26	15	0	259*	15	41	45	29	15	4	0	1087*
8	256	247	23	27	35	43	36	16	26	36	41	16	38	39	41
9	183	178	17	28	22	0	178*	17	25	25	43	17	54	47	31
10	139	132	15	29	69	65	24	18	3	0	997*		0,	8, l	
11	540	530	25	30	4	0	932*	19	43	46	29	0	43	35	33
12	2	0	933*	31	4	0	1014*	20	82	84	19	1	25	0	150*
13	217	218	23		0,	4, l		21	19	15	64	2	35	32	39
14	314	312	28	0	111	113	11	22	44	43	30	3	4	0	1002*
15	237	232	24	1	155	160	16	23	4	0	989*	4	12	0	106*
16	2	0	1032*	2	135	130	13	24	72	77	24	5	4	0	904*
17	144	144	13	3	170	169	17	25	13	0	103*	6	15	0	256*
18	116	120	13	4	104	107	11	26	4	0	889*	7	4	0	958*
19	68	71	17	5	183	179	18		0,	6, l			1,	0, l	
20	3	0	1170*	6	2	0	1054*	0	58	57	20	1	213	231	11
21	69	72	17	7	106	109	11	1	3	0	909*	3	666	682	56
22	164	169	16	8	98	95	12	2	57	57	20	5	876	882	67
23	3	0	1079*	9	16	0	133*	3	14	0	227*	7	813	795	18
24	189	191	18	10	117	120	13	4	151	151	15	9	3	0	187*
25	25	0	132*	11	54	53	17	5	75	78	18	11	156	153	16
26	19	0	175*	12	3	0	968*	6	32	41	34	13	302	292	25
27	6	0	195*	13	69	66	15	7	107	106	16	15	9	0	66*
28	62	62	23	14	3	0	1090*	8	35	34	30	17	72	77	14
29	22	0	156*	15	31	27	28	9	14	0	228*	19	136	138	14
30	43	52	34	16	111	118	13	10	54	62	23	21	55	47	19
31	12	0	362*	17	16	22	57	11	3	0	876*	23	48	50	22
32	25	0	172*	18	75	80	17	12	42	44	27	25	119	123	15
	0,	3, l		19	60	62	21	13	20	24	53	27	110	110	17
1	44	46	13	20	10	0	344*	14	37	34	31	29	135	134	17
2	483	477	23	21	68	69	21	15	43	44	29	31	99	105	20
3	295	288	24	22	13	0	284*	16	11	0	621*	33	51	56	33
4	158	157	16	23	3	0	1102*	17	46	52	28		1,	1, l	
5	355	352	25	24	4	0	1028*	18	40	39	33	0	1323	1334	63
6	59	58	11	25	4	0	1024*	19	39	32	36	1	531	540	14
7	76	73	10	26	14	0	282*	20	4	0	940*	2	235	230	15
8	108	108	11	27	40	36	34	21	4	0	1185*	3	379	386	15
9	108	98	10	28	4	0	1228*	22	4	0	1107*	4	851	867	71
10	47	44	15	29	49	43	31	23	66	66	28	5	498	510	17
11	29	34	23		0,	5, l			0,	7, l		6	337	339	18
12	15	0	137*	1	2	0	1249*	1	24	33	47	7	117	112	11
13	175	169	18	2	182	182	18	2	11	0	316*	8	26	19	19
14	37	31	23	3	13	14	61	3	30	27	37	9	573	568	21
15	2	0	1095*	4	123	123	13	4	23	25	52	10	34	22	16
16	8	0	583*	5	272	276	28	5	41	34	28	11	486	465	23
17	111	104	13	6	65	65	17	6	46	41	25	12	317	305	25
18	11	0	240*	7	3	0	860*	7	24	0	135*	13	108	101	11
19	230	234	23	8	99	102	14	8	38	36	31	14	50	52	14
20	152	153	15	9	40	44	23	9	4	0	884*	15	234	225	23
21	52	55	22	10	94	98	14	10	42	35	30	16	113	115	11

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	1, 1, l				1, 3, l			18	45	42	26	11	3	0	1058*
17	101	99	12	0	10	0	160*	19	120	120	15	12	4	0	845*
18	123	120	13	1	141	138	14	20	53	54	24	13	10	0	309*
19	138	142	13	2	61	61	11	21	52	59	24	14	17	11	63
20	67	70	17	3	15	0	110*	22	3	0	1020*	15	41	43	30
21	134	140	13	4	154	155	15	23	34	36	33	16	48	44	27
22	23	0	117*	5	177	168	18	24	4	0	1028*	17	43	38	31
23	13	0	215*	6	75	76	10	25	16	9	74	18	60	65	24
24	78	79	18	7	109	98	10	26	26	0	171*	19	12	0	110*
25	138	138	15	8	11	0	161*	27	4	0	1023*	20	4	0	982*
26	109	109	17	9	111	110	12	28	19	18	72	21	68	63	25
27	69	70	21	10	19	0	104*	29	22	0	171*	22	68	56	27
28	76	74	21	11	81	78	12		1, 5, l				1, 7, l		
29	66	66	23	12	51	46	17	0	64	64	16	0	6	0	172*
30	65	71	25	13	56	53	16	1	73	74	15	1	21	0	151*
31	4	0	1010*	14	59	60	17	2	61	56	17	2	61	54	22
32	20	0	221*	15	12	0	228*	3	152	146	16	3	52	47	25
33	62	58	27	16	197	200	20	4	175	176	17	4	38	35	30
	1, 2, l			17	141	143	13	5	33	32	28	5	24	0	172*
0	338	344	19	18	47	46	21	6	36	36	25	6	43	37	28
1	212	208	20	19	146	152	14	7	5	0	171*	7	4	0	677*
2	48	52	11	20	3	0	1006*	8	81	87	15	8	4	0	881*
3	150	147	14	21	157	163	15	9	102	104	14	9	4	0	322*
4	28	26	17	22	133	136	15	10	118	121	13	10	20	17	55
5	110	107	11	23	43	43	28	11	60	59	18	11	19	0	178*
6	43	43	12	24	3	0	1171*	12	57	61	19	12	4	0	995*
7	387	382	22	25	62	67	23	13	39	37	27	13	4	0	912*
8	12	0	136*	26	77	77	21	14	85	90	17	14	4	0	1015*
9	454	452	23	27	47	49	29	15	22	0	134*	15	55	56	30
10	97	96	9	28	6	0	186*	16	63	69	20	16	25	0	175*
11	325	322	25	29	8	0	433*	17	26	28	40	17	54	52	33
12	59	55	13	30	33	32	46	18	37	34	30		1, 8, l		
13	106	101	11	31	29	25	51	19	44	40	27	0	41	34	33
14	235	235	24		1, 4, l			20	3	0	1267*	1	12	0	363*
15	50	51	18	0	42	44	16	21	32	34	38	2	4	0	928*
16	214	212	22	1	67	65	13	22	8	0	461*	3	4	0	1087*
17	45	39	21	2	394	386	28	23	53	47	26	4	4	0	1012*
18	52	52	20	3	2	0	900*	24	57	51	26	5	8	0	171*
19	151	160	15	4	194	194	19	25	12	0	684*	6	15	0	91*
20	14	0	211*	5	73	74	13	26	53	49	31	7	4	0	1012*
21	136	137	15	6	103	96	12		1, 6, l				2, 0, l		
22	22	0	148*	7	87	88	12	0	71	75	18	0	342	334	13
23	53	52	22	8	58	63	16	1	23	0	142*	2	128	137	13
24	101	104	16	9	76	70	14	2	49	52	23	4	1414	1407	72
25	64	66	23	10	90	97	14	3	10	0	297*	6	1023	1024	79
26	10	0	318*	11	14	0	159*	4	3	0	955*	8	381	374	20
27	63	63	22	12	88	87	14	5	73	71	19	10	217	216	19
28	43	40	30	13	41	47	24	6	48	47	24	12	780	733	24
29	37	30	35	14	92	97	14	7	48	46	24	14	33	29	19
30	4	0	1074*	15	42	49	25	8	10	0	341*	16	240	234	24
31	25	27	57	16	140	143	14	9	50	48	22	18	108	96	12
32	28	0	157*	17	63	61	18	10	20	0	159*	20	202	208	20

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	2,	0, l		9	743	737	24	27	19	0	210*	17	52	46	24
22	66	67	18	10	137	141	14	28	30	34	45	18	41	40	29
24	166	169	16	11	148	140	15	29	4	0	1197*	19	58	57	23
26	76	74	19	12	31	35	21	30	61	65	28	20	59	54	24
28	113	113	17	13	105	110	10		2,	4, l		21	11	0	629*
30	33	43	42	14	15	0	171*	0	156	153	14	22	4	0	1021*
32	4	0	1268*	15	87	75	13	1	164	162	16	23	25	0	148*
	2,	1, l		16	34	34	26	2	67	69	13	24	14	0	280*
0	327	348	16	17	25	27	35	3	56	52	16	25	21	30	71
1	839	865	70	18	190	186	19	4	42	36	19	26	4	0	1055*
2	403	403	16	19	24	0	123*	5	237	235	25		2,	6, l	
3	1110	1113	75	20	87	85	15	6	2	0	1011*	0	123	122	15
4	1728	1693	80	21	3	0	1088*	7	53	60	17	1	95	89	16
5	118	119	12	22	36	32	29	8	84	85	13	2	60	60	20
6	598	613	19	23	87	93	17	9	253	258	26	3	49	46	24
7	132	121	13	24	23	0	144*	10	41	46	23	4	35	35	30
8	26	32	20	25	14	0	219*	11	192	190	19	5	25	22	40
9	400	394	22	26	20	22	58	12	69	71	16	6	30	34	35
10	260	261	23	27	63	65	23	13	87	88	14	7	11	0	84*
11	92	86	9	28	19	0	197*	14	68	72	17	8	20	9	48
12	453	445	25	29	19	14	64	15	85	85	16	9	16	0	179*
13	212	199	22	30	28	0	159*	16	86	88	16	10	10	0	586*
14	345	327	27	31	46	51	35	17	51	55	23	11	16	0	169*
15	149	147	14	32	95	89	23	18	3	0	1100*	12	34	38	34
16	175	164	18		2,	3, l		19	102	102	16	13	11	0	730*
17	74	68	14	0	8	0	70*	20	66	71	21	14	42	34	29
18	50	48	19	1	286	291	24	21	118	119	16	15	22	23	53
19	8	0	675*	2	345	340	25	22	42	38	28	16	88	84	18
20	42	45	24	3	115	109	12	23	30	23	38	17	43	41	31
21	37	39	27	4	341	347	25	24	4	0	982*	18	28	0	169*
22	23	29	40	5	84	86	10	25	21	19	58	19	67	55	24
23	3	0	1060*	6	15	0	122*	26	11	0	796*	20	23	0	178*
24	77	78	18	7	121	129	13	27	4	0	932*	21	4	0	1075*
25	24	0	164*	8	145	150	15	28	19	0	192*	22	38	31	41
26	62	63	22	9	102	103	10		2,	5, l			2,	7, l	
27	62	60	23	10	139	142	15	0	58	61	19	0	49	49	27
28	101	102	19	11	16	0	151*	1	111	111	13	1	55	52	24
29	40	45	35	12	12	0	63*	2	37	40	25	2	20	0	158*
30	25	0	129*	13	196	194	20	3	111	108	13	3	47	48	29
31	28	22	53	14	100	101	13	4	9	0	344*	4	19	0	167*
32	4	0	975*	15	68	72	17	5	73	70	16	5	34	30	32
33	38	38	41	16	163	161	16	6	119	124	13	6	35	40	35
	2,	2, l		17	59	58	19	7	94	91	14	7	25	0	133*
0	131	134	14	18	190	191	19	8	148	148	15	8	23	0	133*
1	678	673	20	19	70	70	17	9	19	0	136*	9	31	25	37
2	112	115	12	20	108	113	15	10	163	168	17	10	4	0	1089*
3	205	196	17	21	72	71	19	11	3	0	905*	11	28	30	43
4	245	247	21	22	64	68	21	12	3	0	986*	12	4	0	915*
5	273	273	22	23	45	42	27	13	153	150	14	13	38	38	36
6	108	105	11	24	49	44	26	14	43	47	26	14	18	0	210*
7	122	121	12	25	21	0	146*	15	78	81	19	15	4	0	1108*
8	96	101	10	26	89	87	18	16	3	0	983*	16	21	0	188*

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	2,	8, l		26	72	74	21	11	31	29	25	1	21	0	131*
0	43	33	33	27	21	0	149*	12	39	32	22	2	142	141	15
1	8	0	177*	28	55	55	26	13	87	92	14	3	3	0	1220*
2	23	14	52	29	25	0	160*	14	82	81	15	4	44	46	22
3	12	0	616*	30	4	0	1208*	15	98	96	14	5	11	0	251*
4	19	0	189*	31	98	99	21	16	45	44	22	6	126	129	13
5	60	56	27	32	103	103	21	17	3	0	974*	7	42	40	23
6	36	32	40		3,	2, l		18	156	154	17	8	3	0	899*
	3,	0, l		0	2	0	1045*	19	22	20	44	9	57	59	19
1	212	218	18	1	438	430	21	20	95	98	16	10	109	114	14
3	65	66	9	2	375	377	22	21	30	23	36	11	48	43	22
5	10	3	45	3	183	183	19	22	63	68	21	12	103	102	15
7	668	664	20	4	67	63	10	23	92	101	19	13	101	103	16
9	80	77	9	5	259	252	23	24	10	0	679*	14	48	53	25
11	463	449	24	6	21	13	26	25	3	0	1060*	15	3	0	990*
13	146	143	16	7	287	288	24	26	17	16	74	16	93	90	18
15	125	123	12	8	51	55	13	27	4	0	1096*	17	3	0	1091*
17	145	145	15	9	192	193	19	28	16	0	240*	18	129	120	16
19	26	13	35	10	2	0	991*	29	94	92	22	19	20	0	161*
21	48	53	22	11	150	153	15	30	25	27	62	20	8	0	153*
23	216	225	22	12	71	72	12		3,	4, l		21	11	0	348*
25	156	153	16	13	253	254	24	0	112	112	11	22	24	0	151*
27	17	0	185*	14	53	55	18	1	95	96	12	23	34	33	41
29	41	36	31	15	30	29	28	2	17	0	126*	24	4	0	1095*
31	44	53	35	16	79	79	14	3	156	151	15	25	4	0	834*
	3,	1, l		17	106	108	13	4	19	23	42		3,	6, l	
0	96	93	10	18	45	47	23	5	11	0	217*	0	110	106	15
1	249	259	19	19	38	37	26	6	22	19	36	1	3	0	888*
2	183	198	16	20	61	65	19	7	332	329	32	2	39	41	27
3	819	820	19	21	31	27	32	8	133	138	13	3	89	90	17
4	288	293	20	22	35	48	32	9	104	109	13	4	88	93	17
5	17	31	29	23	70	68	20	10	125	120	12	5	63	59	20
6	241	239	21	24	42	44	29	11	41	43	25	6	21	0	140*
7	17	0	91*	25	90	92	18	12	135	134	13	7	3	0	899*
8	219	222	19	26	4	0	943*	13	89	90	15	8	3	0	891*
9	71	65	10	27	60	60	23	14	136	133	13	9	23	0	121*
10	237	232	20	28	37	40	35	15	82	81	16	10	64	56	21
11	57	55	12	29	18	0	200*	16	99	96	15	11	3	0	1102*
12	101	110	11	30	140	144	19	17	3	0	986*	12	3	0	981*
13	94	90	10	31	115	113	22	18	22	0	151*	13	27	0	145*
14	83	81	12		3,	3, l		19	92	92	18	14	60	57	23
15	41	37	20	0	207	208	21	20	131	134	16	15	40	44	32
16	62	57	16	1	312	300	25	21	15	0	253*	16	30	29	41
17	15	11	55	2	7	0	299*	22	3	0	1034*	17	11	0	294*
18	89	82	14	3	448	450	25	23	57	52	24	18	28	35	50
19	135	136	14	4	204	192	22	24	71	72	22	19	4	0	1161*
20	119	121	13	5	47	41	14	25	4	0	1081*	20	44	48	36
21	138	146	14	6	187	184	20	26	24	0	167*	21	12	0	358*
22	97	99	15	7	37	34	17	27	8	0	507*		3,	7, l	
23	53	50	22	8	80	76	11	28	4	0	1381*	0	4	0	983*
24	60	61	22	9	42	40	18		3,	5, l		1	32	29	39
25	126	128	15	10	175	170	18	0	42	40	23	2	80	76	20

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	3, 7, l			12	2	0	921*	31	49	50	35	19	78	84	19
3	42	33	28	13	204	205	21		4, 3, l			20	16	0	188*
4	44	43	30	14	83	85	13	0	265	277	22	21	15	0	232*
5	37	30	32	15	75	70	14	1	85	83	10	22	84	89	20
6	37	38	34	16	68	64	16	2	147	147	14	23	4	0	1005*
7	26	0	132*	17	98	95	14	3	16	0	103*	24	20	15	66
8	8	0	434*	18	95	95	14	4	271	263	23	25	18	0	220*
9	48	47	28	19	3	0	1132*	5	72	77	12	26	54	53	28
10	4	0	1063*	20	91	100	15	6	12	0	169*	27	44	32	35
11	28	0	143*	21	35	43	30	7	121	121	11		4, 5, l		
12	4	0	1001*	22	56	58	21	8	74	73	13	0	155	153	16
13	4	0	1207*	23	97	99	17	9	107	105	12	1	145	153	14
14	4	0	1255*	24	85	83	18	10	51	51	19	2	129	125	14
15	4	0	1124*	25	59	63	24	11	278	277	26	3	14	0	174*
16	21	0	189*	26	25	14	45	12	21	0	130*	4	64	62	17
	3, 8, l			27	8	0	520*	13	44	47	23	5	152	155	16
0	8	0	451*	28	11	0	700*	14	203	208	20	6	94	92	14
1	4	0	850*	29	36	40	39	15	111	118	13	7	60	61	18
2	4	0	945*	30	46	44	35	16	42	47	25	8	67	67	17
3	10	0	137*	31	107	113	22	17	69	65	17	9	6	0	142*
4	26	24	54		4, 2, l			18	36	43	30	10	22	5	45
	4, 0, l			0	527	519	23	19	100	100	16	11	47	45	25
0	525	541	19	1	104	103	11	20	28	40	42	12	129	132	15
2	423	414	19	2	421	411	23	21	3	0	1169*	13	3	0	956*
4	531	517	20	3	272	263	24	22	120	122	16	14	40	38	29
6	11	4	43	4	292	291	24	23	3	0	1018*	15	86	84	18
8	9	0	57*	5	374	359	24	24	4	0	947*	16	3	0	1037*
10	285	266	25	6	189	192	18	25	35	33	35	17	90	85	18
12	279	286	27	7	340	331	25	26	4	0	966*	18	15	17	76
14	88	88	11	8	146	149	15	27	18	0	223*	19	48	47	29
16	327	321	31	9	238	247	22	28	8	0	503*	20	26	0	150*
18	218	205	22	10	286	289	27	29	42	32	37	21	40	37	33
20	91	92	15	11	166	165	15		4, 4, l			22	19	18	69
22	138	150	14	12	138	139	13	0	149	148	15	23	27	0	154*
24	43	49	29	13	118	121	12	1	68	70	15	24	12	0	579*
26	134	137	16	14	165	165	17	2	73	75	14		4, 6, l		
28	57	61	26	15	18	23	51	3	58	58	17	0	110	105	16
30	4	0	1183*	16	155	149	15	4	68	71	16	1	7	0	449*
32	58	62	30	17	12	0	248*	5	265	275	26	2	35	30	31
	4, 1, l			18	55	54	20	6	2	0	1112*	3	69	59	19
0	476	474	20	19	14	0	199*	7	124	130	12	4	26	25	41
1	1	0	980*	20	17	0	165*	8	3	0	1073*	5	117	112	16
2	166	163	15	21	88	92	17	9	22	0	122*	6	32	26	35
3	292	294	21	22	57	57	22	10	139	138	14	7	23	28	48
4	164	169	16	23	3	0	1194*	11	263	267	25	8	3	0	1015*
5	106	113	11	24	111	110	16	12	31	25	32	9	14	9	69
6	321	321	22	25	11	0	365*	13	219	215	21	10	28	32	40
7	14	0	121*	26	44	46	30	14	126	129	14	11	17	17	62
8	427	419	24	27	37	40	36	15	82	78	16	12	47	50	26
9	419	405	25	28	8	0	497*	16	35	34	31	13	4	0	315*
10	311	306	26	29	70	68	25	17	88	87	17	14	40	31	32
11	90	88	10	30	30	0	152*	18	17	0	187*	15	16	0	223*

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	4,	6, l		11	231	230	24	30	12	0	700*	20	67	72	23
16	22	17	54	12	66	62	14		5,	3, l		21	22	0	156*
17	27	0	142*	13	16	0	152*	0	92	94	11	22	53	53	27
18	36	41	39	14	150	145	16	1	44	41	16	23	33	35	40
19	28	25	51	15	46	45	21	2	89	87	11	24	4	0	947*
20	15	0	89*	16	2	0	1116*	3	49	42	15	25	4	0	1030*
	4,	7, l		17	17	0	148*	4	151	150	15	26	4	0	1248*
0	51	54	25	18	13	0	219*	5	52	42	16		5,	5, l	
1	25	28	48	19	14	9	61	6	155	154	16	0	222	226	23
2	21	0	153*	20	61	62	19	7	143	148	14	1	55	56	21
3	53	55	26	21	43	46	27	8	47	44	19	2	138	139	13
4	69	68	21	22	55	53	22	9	89	89	13	3	28	31	32
5	29	0	148*	23	50	50	26	10	157	159	17	4	51	53	20
6	4	0	830*	24	56	57	25	11	452	460	32	5	70	73	17
7	27	23	46	25	29	29	41	12	56	53	20	6	9	0	97*
8	65	52	23	26	23	0	156*	13	26	25	38	7	18	0	153*
9	55	49	27	27	23	25	55	14	3	0	1314*	8	71	74	18
10	24	0	157*	28	14	0	325*	15	234	240	23	9	10	0	616*
11	4	0	859*	29	4	0	1237*	16	155	152	15	10	12	0	270*
12	30	10	45	30	34	27	45	17	40	45	28	11	3	0	966*
13	26	19	52	31	24	0	161*	18	146	146	15	12	33	41	32
14	4	0	1212*		5,	2, l		19	70	74	20	13	25	33	43
15	4	0	979*	0	271	268	25	20	2	0	464*	14	3	0	1097*
	5,	0, l		1	12	0	148*	21	30	37	41	15	10	0	295*
1	222	215	22	2	402	389	25	22	15	0	246*	16	4	0	886*
3	111	120	11	3	39	37	15	23	3	0	1069*	17	11	0	603*
5	264	262	23	4	61	61	12	24	34	32	37	18	44	49	30
7	414	408	24	5	208	201	22	25	4	0	1011*	19	18	0	218*
9	468	453	25	6	131	134	13	26	54	55	28	20	4	0	941*
11	201	204	18	7	160	165	17	27	4	0	1010*	21	19	8	71
13	96	89	12	8	199	194	20	28	24	0	189*	22	12	0	372*
15	47	46	20	9	148	148	14		5,	4, l		23	24	0	159*
17	23	0	133*	10	2	0	1109*	0	89	89	13		5,	6, l	
19	32	31	29	11	164	162	16	1	120	125	12	0	18	16	59
21	39	42	26	12	81	83	14	2	83	86	14	1	16	10	61
23	100	97	17	13	284	286	26	3	41	44	21	2	20	0	133*
25	13	0	283*	14	52	52	21	4	50	49	20	3	3	0	987*
27	22	21	55	15	66	65	18	5	307	309	27	4	104	102	16
29	30	37	51	16	56	54	20	6	128	134	13	5	39	45	29
31	113	109	21	17	40	39	26	7	229	232	25	6	35	31	31
	5,	1, l		18	73	72	17	8	34	35	29	7	4	0	926*
0	106	99	11	19	49	47	23	9	109	109	13	8	20	0	165*
1	30	36	17	20	5	0	209*	10	72	68	16	9	61	66	22
2	413	402	22	21	106	105	16	11	165	166	18	10	17	0	202*
3	263	262	23	22	39	38	31	12	29	32	35	11	22	15	50
4	446	436	23	23	99	99	18	13	79	82	16	12	70	65	21
5	274	283	24	24	10	0	703*	14	3	0	960*	13	21	0	201*
6	85	86	10	25	8	0	543*	15	27	23	40	14	22	0	200*
7	222	205	21	26	29	38	44	16	80	87	18	15	4	0	1059*
8	65	66	12	27	3	0	1232*	17	50	57	25	16	82	82	22
9	137	135	14	28	8	0	455*	18	32	34	36	17	4	0	1143*
10	256	245	22	29	38	42	40	19	53	49	25	18	4	0	1078*

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	5, 6, l			17	95	101	15	6	134	134	13	2	52	49	21
19	29	14	47	18	147	147	15	7	84	88	14	3	78	78	17
	5, 7, l			19	5	0	187*	8	89	92	14	4	41	42	25
0	18	0	187*	20	219	218	23	9	316	313	27	5	58	69	20
1	14	0	227*	21	95	94	18	10	422	422	33	6	3	0	887*
2	41	38	32	22	19	0	168*	11	38	43	29	7	7	0	424*
3	54	50	26	23	3	0	1077*	12	57	63	20	8	10	0	291*
4	23	26	54	24	56	57	24	13	3	0	1126*	9	10	0	324*
5	56	51	26	25	4	0	1014*	14	288	291	30	10	17	0	167*
6	4	0	970*	26	53	61	28	15	113	115	15	11	59	54	22
7	52	56	29	27	48	54	31	16	55	62	22	12	10	0	291*
8	83	78	22	28	4	0	1216*	17	208	208	22	13	3	0	1182*
9	39	32	36	29	4	0	1137*	18	73	66	20	14	25	0	129*
10	24	12	54	30	53	56	32	19	47	54	27	15	4	0	919*
11	4	0	1020*		6, 2, l			20	56	63	23	16	48	47	29
12	4	0	1116*	0	345	340	26	21	51	46	25	17	89	85	20
13	4	0	1065*	1	67	64	12	22	17	0	202*	18	25	30	53
	6, 0, l			2	102	96	10	23	4	0	1059*	19	4	0	1196*
0	15	0	119*	3	32	25	19	24	4	0	1185*	20	4	0	814*
2	100	101	10	4	250	250	23	25	4	0	1163*	21	12	0	345*
4	200	183	21	5	66	65	13	26	4	0	939*	22	25	0	159*
6	193	197	19	6	17	0	126*	27	35	32	44		6, 6, l		
8	118	114	11	7	93	100	12		6, 4, l			0	3	0	998*
10	110	105	11	8	172	165	17	0	92	96	14	1	6	0	174*
12	2	0	1099*	9	140	138	14	1	96	94	14	2	55	57	23
14	126	127	13	10	2	0	1057*	2	9	0	649*	3	45	44	27
16	98	103	14	11	205	209	21	3	181	176	18	4	38	39	31
18	24	20	38	12	360	365	32	4	36	38	26	5	42	38	27
20	18	12	56	13	186	189	18	5	161	162	16	6	44	39	27
22	190	196	19	14	318	314	28	6	49	50	21	7	11	0	640*
24	37	39	33	15	237	236	23	7	60	54	18	8	83	76	19
26	4	0	1045*	16	141	142	14	8	69	73	17	9	36	34	35
28	51	53	32	17	80	71	17	9	119	125	14	10	4	0	980*
30	89	78	23	18	60	65	20	10	30	30	31	11	16	0	227*
	6, 1, l			19	10	0	748*	11	42	49	25	12	104	97	19
0	117	115	12	20	45	52	28	12	41	34	26	13	39	33	35
1	67	64	11	21	62	61	22	13	80	73	17	14	27	0	145*
2	498	479	24	22	3	0	1088*	14	3	0	1226*	15	58	57	27
3	261	247	25	23	6	0	197*	15	24	18	45	16	25	0	159*
4	413	402	25	24	39	34	32	16	82	89	18	17	14	0	273*
5	12	10	51	25	27	0	153*	17	26	29	43	18	4	0	904*
6	156	154	15	26	4	0	1073*	18	43	40	28		6, 7, l		
7	213	206	23	27	4	0	1064*	19	76	70	21	0	18	0	219*
8	215	212	20	28	18	0	221*	20	4	0	1062*	1	35	30	38
9	206	197	21	29	27	0	137*	21	22	18	55	2	28	22	45
10	92	96	12		6, 3, l			22	27	0	140*	3	47	44	31
11	54	52	17	0	129	130	11	23	4	0	1020*	4	16	0	231*
12	94	93	13	1	59	54	15	24	23	0	172*	5	23	0	170*
13	251	250	27	2	140	139	15	25	12	0	679*	6	46	37	30
14	81	76	15	3	47	45	18		6, 5, l			7	12	0	328*
15	22	0	140*	4	31	27	27	0	3	0	1191*	8	49	51	31
16	94	92	14	5	166	169	17	1	92	88	15	9	51	54	31

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	6, 7, l			1	2	0	990*	23	4	0	1051*	21	4	0	1087*
10	20	21	66	2	92	83	12	24	21	0	180*		7, 6, l		
11	4	0	1049*	3	112	113	11	25	21	17	67	0	11	0	96*
	7, 0, l			4	68	63	14	26	4	0	1276*	1	49	48	25
1	253	247	22	5	48	51	18		7, 4, l			2	37	40	32
3	365	359	26	6	37	36	22	0	3	0	983*	3	39	32	30
5	44	37	16	7	29	26	28	1	125	133	13	4	24	12	45
7	303	296	29	8	96	97	13	2	5	0	171*	5	15	0	223*
9	229	229	21	9	18	0	150*	3	11	0	208*	6	15	0	209*
11	341	339	31	10	263	261	27	4	21	0	135*	7	13	0	259*
13	301	306	26	11	201	200	20	5	96	98	14	8	22	0	179*
15	163	170	18	12	77	77	16	6	3	0	1011*	9	4	0	874*
17	43	41	24	13	15	14	66	7	23	29	41	10	11	0	338*
19	23	0	140*	14	297	290	29	8	16	0	169*	11	23	0	156*
21	80	78	20	15	121	118	14	9	138	143	15	12	43	48	35
23	35	40	37	16	25	23	41	10	82	88	17	13	43	45	36
25	71	67	22	17	25	20	45	11	3	0	1143*	14	18	10	74
27	52	55	31	18	141	143	15	12	99	102	16	15	8	0	409*
29	75	69	25	19	24	0	143*	13	55	58	23	16	25	0	170*
	7, 1, l			20	158	156	16	14	66	75	21		7, 7, l		
0	106	102	11	21	67	65	22	15	15	0	212*	0	35	29	39
1	191	192	19	22	3	0	1011*	16	3	0	1143*	1	20	18	64
2	74	74	11	23	29	22	45	17	30	36	41	2	4	0	1044*
3	133	139	13	24	90	92	20	18	23	23	51	3	20	0	184*
4	34	35	20	25	25	32	52	19	22	0	175*	4	22	0	176*
5	249	237	23	26	4	0	1203*	20	24	0	122*	5	8	0	520*
6	224	215	24	27	4	0	1121*	21	14	0	291*	6	26	0	164*
7	4	0	194*	28	28	21	53	22	4	0	983*	7	22	0	206*
8	159	156	15		7, 3, l			23	31	25	46	8	53	56	31
9	52	53	18	0	118	125	12	24	15	0	282*		8, 0, l		
10	48	46	19	1	22	15	34		7, 5, l			0	133	134	13
11	16	0	168*	2	94	87	13	0	41	41	27	2	73	67	13
12	214	212	21	3	51	52	18	1	25	17	39	4	74	75	13
13	145	144	15	4	2	0	1075*	2	32	44	33	6	35	36	24
14	34	35	28	5	39	33	22	3	35	41	30	8	51	53	18
15	126	125	13	6	80	73	15	4	29	36	35	10	18	0	166*
16	157	159	15	7	20	0	143*	5	21	0	139*	12	416	413	33
17	70	71	17	8	9	0	320*	6	30	28	36	14	207	211	21
18	53	58	22	9	19	22	49	7	17	4	56	16	77	83	18
19	74	73	20	10	107	106	14	8	3	0	976*	18	128	130	15
20	14	0	242*	11	160	164	16	9	46	44	27	20	63	55	23
21	37	31	32	12	49	49	22	10	68	64	20	22	105	107	18
22	41	45	32	13	34	34	29	11	10	0	309*	24	26	17	48
23	69	73	22	14	74	75	18	12	22	34	55	26	4	0	1087*
24	4	0	1011*	15	60	67	21	13	29	22	40	28	4	0	1232*
25	91	90	19	16	71	67	20	14	16	3	70		8, 1, l		
26	40	38	35	17	136	138	15	15	19	0	205*	0	105	107	11
27	24	0	168*	18	27	33	44	16	3	0	1281*	1	93	90	12
28	19	0	207*	19	3	0	1247*	17	40	31	32	2	134	136	12
29	48	50	35	20	14	0	89*	18	11	0	671*	3	2	0	1016*
	7, 2, l			21	42	42	30	19	4	0	952*	4	47	49	19
0	29	29	23	22	6	0	193*	20	59	56	28	5	24	28	33

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	8,	1, l		0	50	59	20	0	17	20	65	19	3	0	1062*
6	50	52	18	1	9	0	280*	1	12	0	239*	21	36	39	37
7	51	46	18	2	176	185	18	2	18	0	182*	23	34	37	41
8	72	77	15	3	3	0	1093*	3	72	73	19	25	25	0	200*
9	58	57	18	4	56	57	18	4	3	0	910*		9,	1, l	
10	91	101	14	5	34	33	27	5	54	51	23	0	109	110	12
11	117	119	13	6	53	49	18	6	16	2	63	1	257	256	26
12	49	46	21	7	44	38	21	7	78	84	19	2	74	81	15
13	70	72	17	8	83	85	15	8	32	31	36	3	17	0	155*
14	157	158	16	9	44	42	22	9	13	0	301*	4	64	64	16
15	109	108	14	10	20	0	136*	10	14	0	76*	5	49	51	19
16	62	57	20	11	3	0	1129*	11	11	0	511*	6	148	143	14
17	46	43	26	12	25	0	143*	12	29	25	40	7	16	0	158*
18	190	191	19	13	140	150	15	13	4	0	851*	8	36	35	26
19	24	0	146*	14	116	122	15	14	25	29	51	9	123	125	13
20	25	0	145*	15	85	87	18	15	23	0	177*	10	57	55	19
21	94	85	18	16	41	44	31	16	26	14	52	11	99	100	15
22	6	0	191*	17	62	67	23	17	84	76	22	12	132	135	13
23	14	0	90*	18	3	0	976*	18	8	0	529*	13	170	176	16
24	43	43	32	19	41	46	33	19	59	59	29	14	40	43	30
25	64	71	28	20	28	28	44		8,	6, l		15	128	131	15
26	33	41	46	21	6	0	196*	0	3	0	1136*	16	3	0	1174*
27	4	0	1054*	22	53	53	28	1	4	0	963*	17	92	92	18
	8,	2, l		23	8	0	458*	2	4	0	1011*	18	31	26	40
0	97	99	12	24	48	44	31	3	23	0	133*	19	25	0	134*
1	117	113	11	25	22	0	192*	4	4	0	924*	20	70	68	22
2	26	30	29		8,	4, l		5	14	0	232*	21	3	0	1095*
3	23	0	117*	0	40	43	24	6	11	0	316*	22	74	72	22
4	53	59	18	1	71	80	17	7	4	0	971*	23	43	56	37
5	63	62	16	2	13	0	192*	8	10	0	127*	24	56	56	30
6	89	92	14	3	17	23	52	9	24	0	149*	25	14	0	284*
7	2	0	1161*	4	53	57	20	10	29	8	42	26	57	54	30
8	80	79	15	5	62	69	19	11	25	0	144*		9,	2, l	
9	14	0	221*	6	36	39	28	12	42	34	33	0	53	59	18
10	9	0	303*	7	13	0	71*	13	4	0	1385*	1	78	83	15
11	3	0	1019*	8	22	0	132*	14	15	0	270*	2	60	54	16
12	99	102	14	9	88	88	17		8,	7, l		3	13	2	61
13	127	139	14	10	22	23	45	0	22	0	183*	4	14	20	60
14	67	69	18	11	42	46	27	1	23	16	57	5	27	28	31
15	39	45	31	12	55	50	23	2	22	0	161*	6	25	30	35
16	49	49	25	13	105	107	17	3	4	0	1142*	7	102	103	14
17	10	0	357*	14	126	126	16	4	27	0	166*	8	51	44	19
18	151	153	15	15	73	71	22		9,	0, l		9	38	36	25
19	38	35	33	16	15	0	245*	1	181	185	19	10	33	30	29
20	45	44	31	17	50	48	27	3	117	110	12	11	72	73	17
21	118	116	17	18	26	20	50	5	17	0	143*	12	10	0	619*
22	40	39	35	19	4	0	1020*	7	3	0	868*	13	23	0	134*
23	116	122	19	20	40	39	36	9	133	131	13	14	3	0	970*
24	28	0	158*	21	36	33	42	11	299	289	29	15	60	51	21
25	4	0	1284*	22	26	0	173*	13	68	67	18	16	39	43	32
26	4	0	1047*	23	15	0	264*	15	137	144	15	17	36	33	34
	8,	3, l			8,	5, l		17	119	117	16	18	45	46	29

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	9,	2, l		18	54	54	28	1	23	25	39	2	70	73	17
19	15	0	244*	19	51	42	30	2	93	95	14	3	44	44	24
20	98	100	19	20	28	0	153*	3	20	0	138*	4	162	174	17
21	124	124	18	21	36	33	41	4	119	124	13	5	43	43	25
22	13	0	111*		9,	5, l		5	14	0	190*	6	46	48	24
23	90	84	22	0	63	63	21	6	12	0	68*	7	80	84	18
24	16	0	92*	1	6	0	179*	7	23	0	139*	8	67	69	20
25	8	0	204*	2	72	70	20	8	171	165	16	9	50	54	24
	9,	3, l		3	36	38	32	9	3	0	1042*	10	3	0	1061*
0	34	46	28	4	24	0	144*	10	109	111	14	11	24	0	128*
1	3	0	1105*	5	13	0	263*	11	2	0	466*	12	6	0	185*
2	68	75	16	6	89	99	19	12	93	89	17	13	21	0	189*
3	37	40	25	7	23	0	166*	13	25	32	44	14	3	0	1062*
4	80	79	15	8	22	0	134*	14	73	71	19	15	113	108	18
5	52	60	20	9	45	36	27	15	20	22	56	16	67	69	24
6	3	0	859*	10	26	0	134*	16	70	70	21	17	85	88	21
7	3	0	733*	11	29	33	43	17	3	0	980*	18	4	0	1249*
8	20	0	130*	12	4	0	1086*	18	58	60	24	19	4	0	1278*
9	3	0	1075*	13	83	84	23	19	4	0	1003*	20	18	0	262*
10	71	70	18	14	4	0	1258*	20	14	0	91*	21	4	0	1022*
11	10	0	627*	15	4	0	1154*	21	4	0	1230*		10,	4, l	
12	53	41	22	16	117	109	20	22	39	46	40	0	19	0	169*
13	58	53	23	17	44	47	36	23	64	59	28	1	18	0	186*
14	106	101	17		9,	6, l		24	61	61	30	2	16	0	62
15	24	26	50	0	4	0	792*		10,	2, l		3	63	62	21
16	137	134	16	1	7	0	186*	0	16	0	53	4	36	35	31
17	22	18	54	2	20	0	179*	1	3	0	1004*	5	62	59	21
18	35	27	37	3	57	55	27	2	119	127	13	6	84	91	19
19	20	30	65	4	47	41	31	3	3	0	1049*	7	3	0	1048*
20	16	0	232*	5	71	74	24	4	3	0	1019*	8	18	0	204*
21	50	58	31	6	20	8	64	5	58	58	18	9	25	0	133*
22	4	0	1189*	7	4	0	1101*	6	79	88	16	10	17	0	234*
23	46	47	35	8	38	32	36	7	21	0	154*	11	8	0	480*
	9,	4, l		9	68	66	25	8	19	0	128*	12	20	0	190*
0	63	69	19	10	14	0	99*	9	25	0	131*	13	92	84	20
1	59	67	21	11	28	0	142*	10	70	78	19	14	19	0	228*
2	33	41	32		10,	0, l		11	3	0	1095*	15	34	34	42
3	32	40	32	0	106	110	13	12	62	64	21	16	4	0	1294*
4	3	0	1076*	2	272	278	28	13	46	37	26	17	85	87	23
5	84	84	17	4	246	248	24	14	38	30	32	18	26	0	154*
6	3	0	1029*	6	63	60	17	15	21	0	187*	19	31	26	46
7	101	106	16	8	62	59	18	16	33	29	36		10,	5, l	
8	19	0	163*	10	129	128	13	17	17	0	216*	0	20	0	151*
9	34	34	32	12	39	45	29	18	67	61	22	1	71	71	20
10	18	0	167*	14	121	123	15	19	4	0	1049*	2	11	0	307*
11	40	34	29	16	34	34	34	20	49	41	30	3	32	34	35
12	43	40	30	18	13	0	246*	21	11	0	293*	4	8	0	426*
13	17	0	237*	20	3	0	1167*	22	33	42	48	5	34	35	36
14	108	104	18	22	77	72	23	23	32	14	44	6	49	47	27
15	81	84	21	24	80	81	25		10,	3, l		7	36	28	34
16	19	0	192*		10,	1, l		0	181	192	20	8	18	0	221*
17	69	72	23	0	12	0	213*	1	43	41	24	9	85	82	21

Columns are 10Fo   10Fc   100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
	10, 5, l			21	36	37	44	5	4	0	820*	11	21	0	157*
10	27	18	47	22	39	46	44	6	98	99	18	12	17	0	207*
11	11	0	645*		11, 2, l			7	4	0	894*	13	51	46	26
12	39	34	37	0	10	0	305*	8	26	0	143*	14	62	64	24
13	26	13	51	1	47	46	24	9	60	61	23	15	20	22	62
14	12	0	695*	2	62	63	20	10	11	0	315*	16	4	0	1013*
15	37	37	42	3	66	64	19	11	18	0	226*	17	52	48	30
	10, 6, l			4	211	218	21	12	52	45	31	18	38	42	40
0	8	0	452*	5	100	110	16	13	48	46	33	19	34	21	41
1	4	0	1156*	6	3	0	982*	14	98	88	21	20	22	18	65
2	4	0	1169*	7	3	0	1169*	15	4	0	1032*		12, 2, l		
3	4	0	1055*	8	176	171	16	16	46	43	34	0	32	32	34
4	4	0	1058*	9	47	36	25		11, 5, l			1	53	55	23
5	46	37	31	10	90	98	18	0	24	0	132*	2	3	0	1234*
6	7	0	206*	11	10	0	670*	1	18	0	199*	3	120	123	16
7	21	0	157*	12	26	18	42	2	21	0	185*	4	3	0	1097*
8	4	0	1013*	13	32	31	36	3	8	0	406*	5	39	43	30
	11, 0, l			14	28	23	42	4	4	0	872*	6	95	92	17
1	105	103	14	15	4	0	997*	5	23	24	59	7	99	100	18
3	22	0	123*	16	4	0	1017*	6	16	0	203*	8	15	0	231*
5	63	54	18	17	52	55	28	7	24	0	168*	9	50	56	26
7	20	0	157*	18	18	0	215*	8	44	46	32	10	78	73	20
9	3	0	931*	19	26	0	166*	9	8	0	477*	11	45	36	28
11	113	114	15	20	4	0	1062*	10	27	32	52	12	56	51	24
13	24	0	128*	21	4	0	1235*	11	19	0	205*	13	41	44	32
15	31	37	40		11, 3, l			12	37	35	43	14	12	0	98*
17	52	61	27	0	21	0	133*		11, 6, l			15	4	0	1174*
19	24	0	173*	1	49	52	24	0	4	0	926*	16	72	65	25
21	122	119	20	2	88	88	17		12, 0, l			17	26	0	165*
23	72	63	27	3	88	99	18	0	3	0	917*	18	4	0	1163*
	11, 1, l			4	25	0	147*	2	104	112	16	19	4	0	1140*
0	87	87	16	5	23	0	124*	4	76	79	18		12, 3, l		
1	142	145	14	6	23	0	131*	6	19	0	164*	0	78	76	19
2	240	242	24	7	24	0	123*	8	63	61	21	1	10	0	629*
3	3	0	1072*	8	47	49	26	10	47	48	27	2	47	50	26
4	124	130	14	9	43	36	28	12	28	31	42	3	81	75	19
5	18	0	173*	10	27	34	43	14	4	0	838*	4	25	0	129*
6	139	137	14	11	23	0	150*	16	27	0	158*	5	39	33	30
7	68	69	19	12	4	0	1035*	18	4	0	1358*	6	15	0	206*
8	97	98	16	13	4	0	944*	20	4	0	1398*	7	4	0	1036*
9	131	129	15	14	4	0	897*		12, 1, l			8	60	67	24
10	3	0	1036*	15	57	58	27	0	165	163	16	9	13	0	81*
11	3	0	953*	16	29	21	46	1	18	22	57	10	17	5	69
12	67	68	21	17	4	0	1154*	2	49	54	25	11	28	31	45
13	94	85	17	18	12	0	382*	3	3	0	966*	12	48	49	31
14	57	51	24	19	21	0	210*	4	24	0	127*	13	15	0	93*
15	139	138	16		11, 4, l			5	50	53	25	14	67	67	26
16	24	0	156*	0	4	0	921*	6	3	0	1107*	15	57	52	30
17	4	0	944*	1	31	26	34	7	3	0	1058*	16	4	0	1112*
18	11	0	748*	2	57	55	22	8	23	0	140*	17	31	0	163*
19	25	0	157*	3	96	96	18	9	3	0	984*		12, 4, l		
20	12	0	375*	4	23	0	136*	10	26	17	43	0	22	4	53

Columns are 10Fo 10Fc 100Sig, * for Insignificant

l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig	l	kFo	Fc	Sig
12, 4, l				12	25	23	51	6	4	0	1165*	1	50	45	28
1	4	0	814*	13	18	11	73	7	20	17	65	2	38	28	36
2	17	0	218*	14	4	0	1151*	8	32	28	44	3	4	0	1011*
3	18	0	196*	15	48	36	32	9	39	25	35	4	31	36	46
4	33	25	37	16	53	44	30	14, 0, l				5	18	0	196*
5	16	26	78	17	4	0	1242*	0	71	63	21	6	4	0	998*
6	24	0	170*	13, 2, l				2	160	157	16	7	73	69	25
7	25	24	53	0	34	31	32	4	147	138	16	8	44	38	35
8	16	0	213*	1	13	0	251*	6	65	67	23	9	27	6	52
9	4	0	322*	2	32	27	34	8	50	41	28	14, 4, l			
10	4	0	1147*	3	6	0	185*	10	93	82	21	0	52	45	30
11	25	15	52	4	31	35	37	12	43	33	36	1	46	45	34
12	48	46	32	5	20	0	166*	14	36	25	40	15, 0, l			
13	28	0	145*	6	20	0	171*	14, 1, l				1	45	47	33
12, 5, l				7	42	38	30	0	56	55	24	3	73	67	24
0	41	40	36	8	19	17	65	1	35	32	34	5	55	50	28
1	4	0	981*	9	32	31	39	2	11	0	113*	7	61	63	28
2	4	0	1022*	10	44	46	31	3	122	119	17	9	56	52	30
3	27	25	54	11	21	0	200*	4	26	0	163*	15, 1, l			
4	4	0	939*	12	57	54	27	5	29	30	44	0	23	0	173*
5	8	0	448*	13	31	32	47	6	11	0	711*	1	65	57	25
6	27	0	155*	14	22	0	184*	7	27	16	44	2	72	63	23
7	30	24	48	15	85	80	22	8	21	0	182*	3	28	22	48
13, 0, l				16	27	27	55	9	4	0	1207*	4	23	0	168*
1	44	44	27	13, 3, l				10	17	14	77	5	4	0	996*
3	279	271	30	0	4	0	989*	11	37	33	38	6	118	113	20
5	3	0	1023*	1	39	42	30	12	51	39	31	7	78	75	24
7	27	39	41	2	23	0	133*	13	19	0	214*	8	7	0	219*
9	33	30	36	3	17	0	195*	14	4	0	1302*	9	42	37	36
11	56	54	27	4	24	0	130*	14, 2, l				15, 2, l			
13	66	60	24	5	43	49	31	0	7	0	167*	0	47	43	31
15	22	0	201*	6	19	0	164*	1	4	0	947*	1	25	0	167*
17	79	77	25	7	16	0	217*	2	29	26	40	2	37	40	40
13, 1, l				8	27	0	136*	3	54	49	26	3	4	0	1168*
0	4	0	884*	9	4	0	1109*	4	21	0	196*	4	29	35	49
1	42	42	29	10	12	0	433*	5	20	0	208*	5	4	0	1013*
2	57	50	23	11	47	43	34	6	31	23	43	6	12	0	715*
3	17	0	202*	12	4	0	1123*	7	24	0	167*	7	38	34	39
4	47	45	26	13	54	56	31	8	24	0	171*	15, 3, l			
5	98	98	17	13, 4, l				9	4	0	939*	0	17	0	247*
6	39	39	31	0	39	33	35	10	27	0	164*	16, 0, l			
7	61	67	24	1	14	0	96*	11	29	26	50	0	28	33	51
8	19	0	216*	2	40	35	35	12	79	75	24	2	4	0	1047*
9	50	49	26	3	12	0	370*	14, 3, l				16, 1, l			
10	55	52	27	4	4	0	869*	0	4	0	1003*	0	30	28	49
11	23	24	54	5	40	36	36								

ERRANL -- The NRCVAX Error Analysis Routine

Crystal Data File x01001.cd

Reflection File x01001.re

ERRANL -- Agreement Tables Routine

Analysis as a function of F and Sint
Counting Statistics weights were used, with .0001200*Fobs**2
4716 Reflections Processed, of which 3125 are Observed.
The Max. Values of Fobs & Sin(theta)/lambda are 172.82 & .67936
The Maximum h,k,l values are 16 8 33

Results in terms of F

Interval	Sum Fobs	Sum Fc	Sum DF	Sum Deltasq	Sum wDeltasq	No. Refl.	Sum Fc/ Sum Fo
2.13	223.4	186.0	73.8	65.17	225.61	127.	.83
2.61	310.2	283.1	76.2	65.85	351.11	130.	.91
3.01	363.4	339.8	67.4	57.83	352.72	129.	.93
3.40	406.5	393.1	54.7	36.99	352.22	127.	.97
3.70	449.2	442.3	47.4	27.92	266.15	127.	.98
3.99	481.9	465.3	51.7	32.93	412.02	125.	.97
4.27	508.0	498.2	43.1	23.50	275.40	123.	.98
4.63	563.2	548.0	47.7	27.66	354.39	127.	.97
4.93	597.1	581.7	44.3	25.41	459.51	125.	.97
5.29	649.2	646.2	45.6	24.86	430.23	127.	1.00
5.70	708.4	693.0	49.3	30.53	550.74	129.	.98
6.22	750.8	738.5	41.6	21.52	478.62	126.	.98
6.79	820.9	810.5	40.6	22.14	473.32	126.	.99
7.35	877.1	863.3	48.4	29.73	696.91	124.	.98
8.14	983.9	978.4	40.6	20.47	526.78	127.	.99
8.94	1094.4	1090.5	44.0	24.73	680.73	128.	1.00
9.73	1172.8	1170.2	46.8	27.83	796.25	126.	1.00
10.82	1279.9	1283.6	39.3	20.39	614.94	125.	1.00
11.98	1433.8	1439.1	44.0	24.91	730.81	126.	1.00
13.93	1622.1	1631.6	40.5	22.02	539.25	125.	1.01
15.99	1903.9	1909.5	39.8	20.77	402.53	127.	1.00
19.55	2241.8	2245.8	53.6	37.38	512.49	126.	1.00
26.51	2883.9	2874.7	65.3	56.76	505.11	126.	1.00
39.18	4027.2	4011.4	79.5	75.04	396.30	126.	1.00
172.84	5589.9	5558.1	95.5	181.63	326.83	91.	.99

Interval	Av.of Fc/Fo	R Value	Av. Fo	Av. Fc	Av. Deltasq	Av. wDeltasq	Av.Fc/ Av.Fo
2.13	.828	.3302	1.8	1.5	.5	1.776	.833
2.61	.912	.2457	2.4	2.2	.5	2.701	.913
3.01	.934	.1856	2.8	2.6	.4	2.734	.935
3.40	.967	.1347	3.2	3.1	.3	2.773	.967
3.70	.985	.1056	3.5	3.5	.2	2.096	.985
3.99	.966	.1073	3.9	3.7	.3	3.296	.966
4.27	.981	.0848	4.1	4.1	.2	2.239	.981
4.63	.973	.0847	4.4	4.3	.2	2.791	.973
4.93	.974	.0742	4.8	4.7	.2	3.676	.974
5.29	.995	.0703	5.1	5.1	.2	3.388	.995
5.70	.978	.0696	5.5	5.4	.2	4.269	.978
6.22	.983	.0554	6.0	5.9	.2	3.799	.984
6.79	.987	.0494	6.5	6.4	.2	3.757	.987
7.35	.984	.0552	7.1	7.0	.2	5.620	.984
8.14	.994	.0413	7.7	7.7	.2	4.148	.994

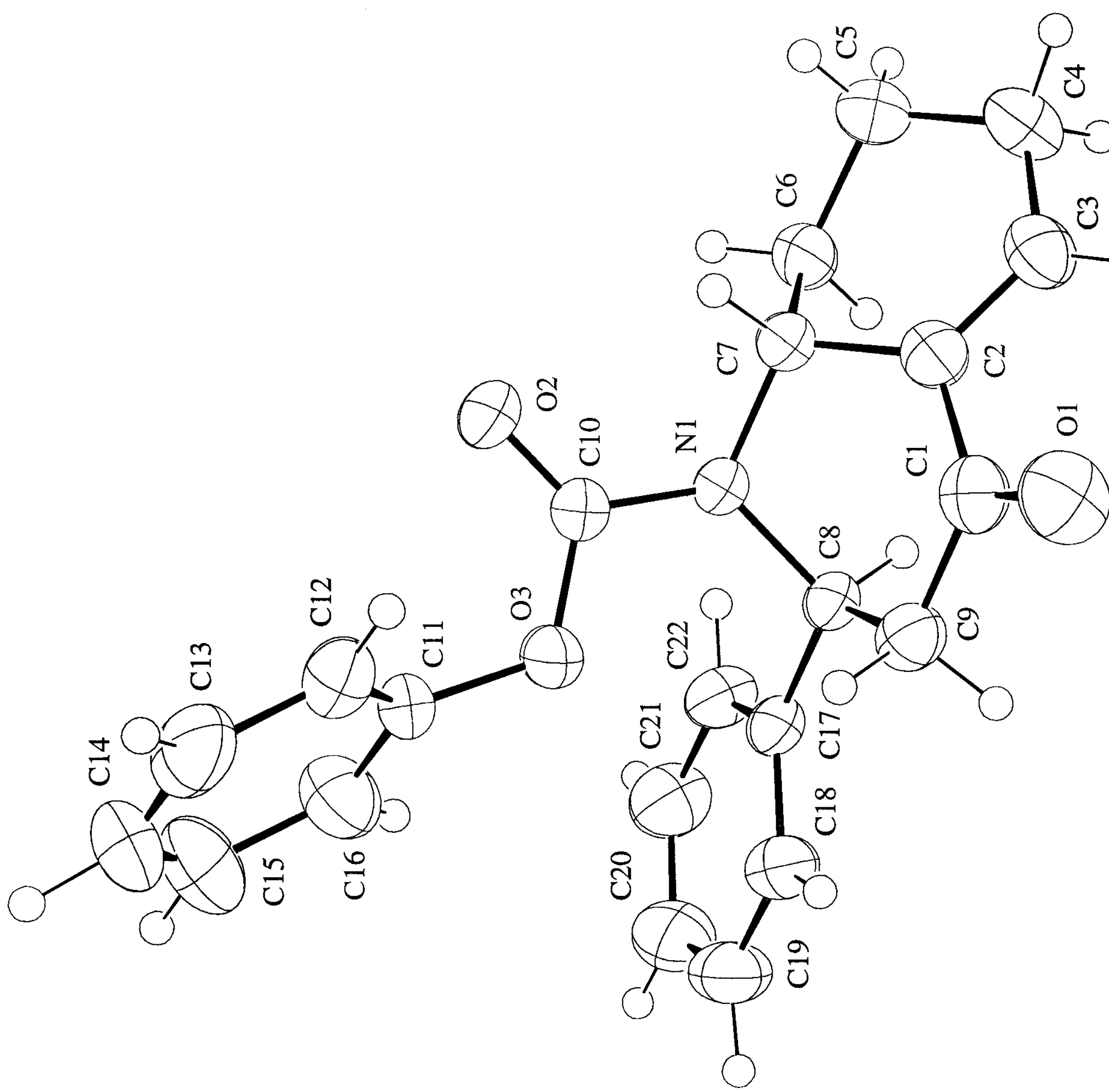
8.94	.996	.0402	8.5	8.5	.2	5.318	.996
9.73	.998	.0399	9.3	9.3	.2	6.319	.998
10.82	1.003	.0307	10.2	10.3	.2	4.920	1.003
11.98	1.003	.0307	11.4	11.4	.2	5.800	1.004
13.93	1.006	.0249	13.0	13.1	.2	4.314	1.006
15.99	1.003	.0209	15.0	15.0	.2	3.169	1.003
19.55	1.002	.0239	17.8	17.8	.3	4.067	1.002
26.51	.997	.0227	22.9	22.8	.5	4.009	.997
39.18	.996	.0197	32.0	31.8	.6	3.145	.996
172.84	.993	.0171	61.4	61.1	2.0	3.592	.994

Results in terms of Sin(theta)

Interval	Sum Fobs	Sum Fc	Sum DF	Sum Deltasq	Sum wDeltasq	No. Refl.	Sum Fc/ Sum Fo
.16520	6039.8	6047.4	120.3	167.15	971.64	179.	1.00
.20814	4114.2	4030.9	113.0	138.12	1054.17	175.	.98
.23826	2171.8	2149.6	66.5	45.80	894.80	158.	.99
.26223	1852.4	1838.7	55.0	27.76	681.11	173.	.99
.28248	1853.4	1865.9	52.4	27.09	541.74	163.	1.01
.30018	1931.7	1941.0	55.5	33.27	607.51	153.	1.00
.31601	1654.4	1667.9	47.0	24.48	504.75	148.	1.01
.33039	1520.2	1540.7	57.6	35.68	634.58	150.	1.01
.34362	1278.4	1301.8	53.6	31.83	596.59	145.	1.02
.35591	1115.8	1130.3	49.6	29.71	507.81	136.	1.01
.36740	1098.2	1114.3	52.6	32.41	582.00	143.	1.01
.37821	886.7	892.1	45.0	24.52	397.12	129.	1.01
.38843	788.1	789.7	47.8	29.01	387.05	128.	1.00
.39815	589.4	574.7	37.4	22.10	281.55	103.	.98
.40741	606.7	592.2	40.9	25.20	284.22	115.	.98
.41627	582.7	572.9	43.0	27.96	307.76	111.	.98
.42477	522.1	505.7	37.3	22.01	266.71	102.	.97
.43294	552.5	529.3	49.4	33.92	322.46	119.	.96
.44081	414.8	399.1	41.2	27.23	213.12	91.	.96
.44841	401.1	386.5	38.5	26.47	253.79	87.	.96
.45577	470.2	438.8	49.2	36.89	316.17	96.	.93
.46289	370.2	343.1	38.1	30.08	272.23	76.	.93
.46980	372.6	340.1	44.2	35.91	274.60	83.	.91
.47651	385.6	351.0	43.5	35.16	301.18	79.	.91
.48304	369.8	338.2	42.2	34.22	256.31	83.	.91

Interval	Av.of Fc/Fo	R Value	Av. Fo	Av. Fc	Av. Deltasq	Av. wDeltasq	Av.Fc/ Av.Fo
.16520	.999	.0199	33.7	33.8	.9	5.428	1.001
.20814	.984	.0275	23.5	23.0	.8	6.024	.980
.23826	.982	.0306	13.7	13.6	.3	5.663	.990
.26223	.991	.0297	10.7	10.6	.2	3.937	.993
.28248	1.006	.0283	11.4	11.4	.2	3.324	1.007
.30018	1.002	.0288	12.6	12.7	.2	3.971	1.005
.31601	1.009	.0284	11.2	11.3	.2	3.410	1.008
.33039	1.004	.0379	10.1	10.3	.2	4.231	1.013
.34362	1.027	.0419	8.8	9.0	.2	4.114	1.018
.35591	.999	.0445	8.2	8.3	.2	3.734	1.013
.36740	1.014	.0479	7.7	7.8	.2	4.070	1.015
.37821	1.011	.0507	6.9	6.9	.2	3.078	1.006
.38843	.986	.0606	6.2	6.2	.2	3.024	1.002
.39815	.953	.0634	5.7	5.6	.2	2.733	.975
.40741	.966	.0674	5.3	5.1	.2	2.472	.976
.41627	.977	.0739	5.2	5.2	.3	2.773	.983
.42477	.963	.0714	5.1	5.0	.2	2.615	.969

.43294	.944	.0894	4.6	4.4	.3	2.710	.958
.44081	.944	.0993	4.6	4.4	.3	2.342	.962
.44841	.955	.0960	4.6	4.4	.3	2.917	.963
.45577	.907	.1046	4.9	4.6	.4	3.293	.933
.46289	.911	.1031	4.9	4.5	.4	3.582	.927
.46980	.896	.1187	4.5	4.1	.4	3.308	.913
.47651	.891	.1128	4.9	4.4	.4	3.812	.910
.48304	.898	.1142	4.5	4.1	.4	3.088	.915


C5
C4
C6
C3
C7
C2
O2
O1
C10
N1
C1
C8
C12
C11
O3
C9
C13
C22
C14
C21
C17
C18
C15
C16
C20
C19