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Verification of CORINAIR 90 emission inventory by comparison with ambient air measurements

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Contents

Summary

This study aims at a validation of the CORINAIR 90 emission inventory by a comparison with measured air quality in the Netherlands and in the Slovak Republic. The study has been performed by TNO and SHMU and has been funded by the European Environment Agency as part of a series of contracts aimed at the verification of the CORINAIR 90 emission inventory.

The CORINAIR 90 has been used to calculate air quality on a $\pm$ 60 x 60 km^2 grid over Europe, using the LOTOS model. The calculations have been performed by using the 1990 meteorological situation. Application of the model results in calculated wind direction dependent concentrations of NO_x, SO_2 and CO, which are statistically compared with measurements of national air quality monitoring networks.

Due to characteristics of the monitoring networks in the Netherlands and in the Slovak Republic, the results of this comparison are more conclusive for the Netherlands as compared to the Slovak Republic. Slovak measuring sites seem to be influenced by local effects to such an extent that comparison with the relatively course spatial resolution of the model does not yield clear results. Drawing conclusions from this comparison therefore is not possible.

Within the Netherlands a fair agreement between calculated and measured air pollutant concentrations is observed.

For SO_2 no clear pattern is recognized in the comparison. Differences between measurements and calculations are mainly attributed to local influences.

NO_x and CO concentrations appear to be underestimated in South-Easterly directions. At many grid cells calculated concentrations are lower than the measured ones. The difference seems to be larger for NO_x than for CO.

Biogenic NMVOC emissions stored in the CORINAIR 90 emission inventory have been compared with the LOTOS and GEIA estimates. The following conclusions are drawn:

- The LOTOS and GEIA calculation methods apply for all countries the same estimation procedure. Therefore, for both methods, the results for the different countries can be regarded as mutually comparable. The CORINAIR data show clear inconsistencies, which have to be subject to further examination.
- The NMVOC emissions for forests might well be underestimated by a factor of 1.5 - 1.9 on an average. The overall biogenic NMVOC emissions might well be underestimated by a factor of 3 - 4 on an average.
- The NMVOC data for CORINAIR 90 lack detailed information about speciation. Therefore, the applicability for studies on e.g. ozone formation will be hampered, as these will require information on the reactivity of the species.

We conclude that the CORINAIR system now seems to work at the regional scale (NUTS-3, same order of magnitude as the 60 km grid of LOTOS) and produces reliable emission estimates. For air quality studies at the local level a spatially more detailed inventory and modelling system is needed. The CORINAIR system will (at the EU level) not be extended to the scale that is needed for such analyses of local air quality. However the CORINAIR methodology can be used at higher spatial resolutions (down to 1 x 1 km). The topic centre and EEA should help countries by providing emission factors and inventory compilation procedures, to produce these kind of inventories.

1. Introduction

1.1 Background

Within Europe much effort has been put into the collection of a complete, consistent and transparent emissions inventory within the programme of CORINAIR. In this programme 29 countries co-operate and compile data within this common framework. This project has been initiated and facilitated by the European Commission in collaboration with participating countries and the CORINE work programme. The databases have been taken over by the European Environment Agency (EEA). The common framework has been developed in close cooperation with the UN-ECE/ EMEP Task Force on Emission Inventories and its expert panels. The inventory compilation methodology therefor is applied in both the EEA's emission inventory CORINAIR and in the emission databases used in EMEP's modelling efforts.

Recently the 1990 database has become available . This database contains the emissions of NO_x, SO_2, NMVOC, CH_4, CO, CO_2, N_2O and NH_3 as caused by human activities and by natural processes.

Pollutants emitted in the atmosphere are subjected to a number of meteorological, physical and chemical processes that result in ambient air concentration fields and deposition on the surface. Due to the lifetime of the pollutants in the atmosphere, emissions in one part of Europe will influence deposition and ambient air quality in other parts of Europe that might be quite distant from the origin of the emission.

This report describes a short study, commissioned by the European Environment Agency as part of its 1994 work programme (project SA2) to verify the database by comparison with measured air quality data and with an independent emission database. The project is a close cooperation between TNO (the Netherlands) and SHMU (Slovak Republic). It aims at two more or less independent verification activities:

a) A ground truth validation technique to study completeness of the database: the TNO LOTOS model will be used to calculate averaged concentrations and wind direction dependent concentrations at different locations using CORINAIR90 emissions as input and compare these with measured concentrations in two countries (Slovak Republic and the Netherlands).

This comparison can rather easily be done for NO_x, SO_2 and CO. Comparison of measured and calculated concentrations might indicate different qualities of the input data for different wind directions (= countries).

b) A check of CORINAIR90 data against an independent database:
the validation of biogenic NMVOC emission data on a European scale. Within the LOTOS model biogenic NMVOC emissions are calculated on the basis of meteorological data (temperature), forest type, leaf biomass and a temperature re-

lated emission algorithm. The availability of the data enables emission calculation of biogenic NMVOC by country and by 0.25° x 0.5° grid cells for a year or an episode within the year. These results will be compared with CORINAIR90 data and differences will be analyzed.

Both activities use emission data of all countries for the components NO_x, SO_2, CO (first activity) and biogenic NMVOC (second activity).

1.2 Uncertainties in measurements, inventories and models

Both measurements, inventories and models are in a number of respects simplifications of reality. They contain in many cases a broad range of assumptions that will influence the accuracy with which these measurements, inventories and models reflect real values. Sources of these kind of uncertainties might have different origins:

a) Assuming averaged values in time, space or activity definitions:
   - measurements, made at a certain location, are assumed to produce a representative concentration for a larger area;
   - inventories aggregate emissions over different technologies, a range of pollutants (NMVOC) and a administrative geographical unit (NUTS-3);
   - models like LOTOS use distinct time steps and calculate grid cell averaged concentrations and produce time resolved emissions for main sectors.

   This will induce uncertainties because of (stochastic) deviations of averaged values within the real world.

b) Parametrizations might contain simplifications that induce uncertainties and errors:
   - in inventories the relation between activity rates and emissions might be more complicated than just multiplying it with an emission factor
   - parametrization of atmospheric processes in many cases neglect higher order effects.

   This type of uncertainties may result in systematic errors, which in some cases can be estimated but in many cases are fully unknown.

c) Errors might exist in measuring results or values of parameters or inputs used:
   - every measurement has a limited accuracy: measurement errors might be present;
   - activity rates and emission factors contain uncertainties or might not be known;
   - both parameters and input data might contain errors and uncertainties.

   This type of uncertainties can in principle be dealt with using error propagation techniques. However this is only seldom done.

Given all these uncertainties, simplifications and possible errors, modellers are reasonably satisfied if their models predict actual concentrations within a factor of 2. In

such a case it might be concluded that the model, including the inputs (emission inventory, meteorological data) is not in conflict with the measurements. It however cannot be regarded as proof that the model and input data are correct. If differences between calculated and measured values are much larger, it must be concluded that something is wrong:

d) the model might be wrong
e) the input data might contain errors or not be complete
f) the measurement might not be as representative for the area as the spatial averaged value obtained from the model.

The above considerations will hamper comparisons between measurements and calculations less if not absolute values but structures and dependencies are studied. Since wind direction is - apart from the emissions - the most important variable to determine concentrations, this study will concentrate on comparing wind direction dependencies as measured and as calculated. It is expected that this comparison will be more conclusive than comparison of absolute values only.

From this it will be clear this study can not proof the correctness of either the model, the inventory or the measurements. It might however produce support for the confidence users might have in both the model and the input data. This study concentrates on verifying the emissions input data as provided by CORINAIR 90.

1.3 Structure of the project

The study in this report was performed in a co-operation between TNO MEP in Delft, the Netherlands, and the Slovak Hydrometeorological Institute (SHMU) in Bratislava, Slovak Republic. The LOTOS model runs were performed at TNO while the statistical analyses were mainly done at SHMU.

Chapter 2 presents the methods used and describes the basics of the emission data and measurements used in the study. A short description of the statistical analyses is given. Chapter 3 describes the results of both the model runs and of the comparisons between calculated and measured concentrations. In chapter 4 the results are discussed and conclusions are drawn.

2. Methods

2.1 Emissions database

The CORINAIR 90 emission database contains emissions of 8 components for 29 European countries. The database has been compiled from national emission inventories, collected by national experts within the CORINAIR framework. The data can be regarded as the "official national emissions" for the year 1990.

Table 1 presents an overview of the European totals stored within this emission database (from [1]). This table shows that anthropogenic emissions in Europe are at least one order of magnitude higher that the biogenic ones, except for nitrous oxide (N_2O), methane (CH_4) and non methane volatile organic compounds (NMVOC).

Table 1. Total emissions in Europe as stored in the CORINAIR 90 emission inventory [1]

	CORINAIR[1] emissions for Europe 1990 (Mg)			LOTOS[2] estimate	difference
pollutant	anthropogenic	biogenic	total	(Mg)	(%)
SO_2	27,478	573	28,051	25,458	10.2
NO_x	17,956	50	18,006	14,989	20.1
NMVOC [3]	17,423	4,347	21,770	15,885	9.7
NH_3	5,586	115	5,701	6,050	-5.8
CO	68,354	1,358	69,712	63,347	10.0
CH_4	35,009	10,406	45,415	37,984	19.6
N_2O	1,327	553	1,880	212	786.8
CO_2	4,472,980	291,779	4,764,759		

1) The CORINAIR 90 emission inventory contains the emissions in the following countries:

a) European Union: Belgium, Denmark, France, the former West Germany, Greece, Ireland, Italy, Luxembourg, Netherlands, Portugal, Spain and United Kingdom

b) EFTA countries: Austria, Finland, Norway, Sweden, Switzerland

c) PHARE countries: Bulgaria, Czech Republic, Hungary, Poland, Rumania, Slovak Republic, Estonia, Latvia, Lithuania, Slovenia

d) Croatia, Malta and the former East Germany

2) The LOTOS database includes emissions in all European countries. The totals presented here are excluding the former Soviet Union

3) LOTOS NMVOC emission estimate for anthropogenic emissions only

Table 1 presents as a comparison also the total emissions as stored in the LOTOS emission database. With the exception of N_2O a fair agreement between the two databases is observed: differences are 20 % or less.

The components relevant for this study are total (anthropogenic + biogenic) emissions of NO_x, SO_2 and CO in the comparison with measured air quality and of biogenic NMVOC at the national total levels.
The emissions stored in the database are emitted by a broad range of activities or may be due to various natural causes. In the CORINAIR system these activities and natural causes are grouped in 11 main sectors. Table 2 presents an overview of the main sector split of the CORINAIR90 emissions. The most important source for NO_x and CO is road traffic. About half of the SO_2 emissions is originating from public power and energy stations. NMVOC's are mainly emitted by road transport, solvent use activities and natural causes.

Table 2. European 1990 emissions split over the 11 main economic sectors as used in the CORINAIR system.

Source sector	Emission (Mg)			
	NO_x	SO_2	CO	NMVOC
Public power, cogeneration and district heating	3,768	15,017	807	55
Commercial, institutional and residential combustion	759	3,065	9,947	989
Industrial combustion	2,457	7,041	8,200	154
Production processes	393	924	3,188	1,220
Extraction and distribution of fossil fuels	82	45	63	1,376
Solvent use	1	0	1	4,920
Road transport	7,874	725	38,919	6,766
Other mobile sources	2,331	571	2,223	677
Waste treatment and disposal	241	89	4,427	506
Agriculture	50	1	579	759
Nature	50	573	1,358	4,347
Total	**18,006**	**28,051**	**69,712**	**21,769**

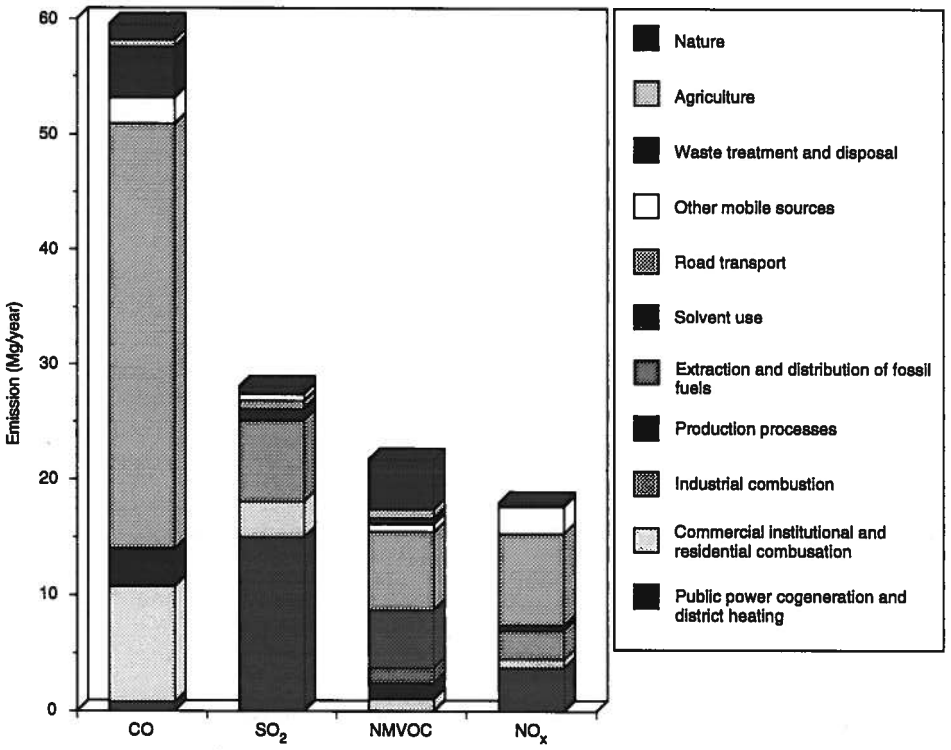


Figure 1. European 1990 emissions split over the 11 main economic sectors as used in the CORINAIR system

The information in table 2 is graphically represented in figure 1. Road traffic is by far the largest contributor to the CO emissions in Europe. The same sector is responsible for more than 40 % of the emissions of NO_x. Public power generation and industrial combustion are the main contributors to the SO_2 emissions in Europe and both sectors also are important sources for NO_x. About 20 % of the emissions of NMVOC in Europe are biogenic.

2.2 LOTOS model

The LOTOS (LOng Term Ozone Simulation) model uses a three dimensional time dependent atmospheric continuity equation for multiple chemical species, to calculate concentrations on an Eulerian grid system over Europe. Grid size is 0.5° latitude x 1.0° longitude (about 60 x 60 km^2). Vertically the model uses 4 dynamic layers to an altitude of about 2.5 km:

a) the surface layer, immediately above ground level;
b) the mixed layer extending to the base of the elevated inversion;
c) the lower inversion layer to keep track of emissions injected into the stable layer during the night; and
d) the upper inversion layer which acts as a reservoir for emissions penetrating the upper part of the whole inversion layer and for aged pollutants entrained into this layer during multiday episodes.

The LOTOS model uses a chemical kinetics package called CBM-IV (Carbon Bond Mechanism - IV) to model atmospheric chemistry. This chemistry package requires speciated VOC's and time resolved emissions (hourly averages) per sector, which are not available in CORINAIR. Since this comparison only aims at relatively inert chemical species the chemical aspect of LOTOS is not needed explicitly. The chemical part of LOTOS has not been switched off however and the LOTOS parametrization of both speciation and time resolution have been used. Calculated concentrations of NO_x, SO_2 and CO are expected to be not very sensitive to these assumptions.

The LOTOS-model contains per emission source category the relevant time and temperature dependencies. Internally, the model calculates the hourly emissions which are used in the dispersion calculation.

The model uses, in this project, a time step of six hours. The input of the model consists of

a) a database of annual total emissions of NO_x, SO_2, CO and a number of speciated NMVOC's and of
b) meteorological data:
   - layer averaged gridded wind fields, temperature and humidity
   - gridded mixing height fields
   - gridded fields of surface wind speeds, precipitation amounts, fractions of cloud cover, and ground level temperature.

Meteorological data for 1990 were obtained from LAM-150 (Limited Area Model - 150 km^2 grid resolution) of the Norwegian Meteorological Institute in Oslo. It is the same database which is used in the EMEP model.

The model output (at six hour time step) consists of

a) every sixth hour the hourly averaged concentrations of NO_x, SO_2, CO and O_3 at every grid cell within Europe
b) an hourly value of biogenic NMVOC emission per grid cell.

2.3 Measurements

2.3.1 Slovak Republic

In the Slovak Republic the Slovak Hydrometeorological Institute carries out the air quality measurements. The first measurements of air pollutants in Slovakia were started in 1970. At the beginning SO_2 an particulate matter were measured, gradually both the number of stations was extended and measurements of NO_2, As, F and heavy metals were included. The data were obtained by manual methods discontinuously as 24-hour daily concentrations. The last 20 years the aim was to evaluate the local air pollution level in a broader scale and to estimate the most polluted regions

which were monitored. The manual stations have been gradually replaced by new automatic monitoring stations. Every year two mobile automatic monitoring stations are for one year placed in new locations to estimate the air pollution level in other regions. The measurements from the regional stations provide daily averages of NO_x and SO_2 concentrations.

The automatic monitoring stations now are placed in so called 'heavy polluted areas'. The heavy polluted areas include areas of at least 50 km^2 if the

- annual air quality standard for a pollutant is exceeded
- short term air quality standards of two pollutants for SO_2, NO_2 , PM, CO are exceeded in more than 5% of the time.

The heavy polluted territory is 2978 km^2, i.e. 5.7% of the total Slovak Republic. The number of inhabitants living in the heavy polluted areas is 1.27 million, i.e. 24% of the total number of inhabitants in the Slovak Republic.

The air quality monitoring network consist of

a) about 25 measuring sites for SO_2
b) about 25 measuring sites for NO_x
c) about 10 measuring sites for CO

Figure 2 (right column) shows measured wind direction dependencies at a grid cell within the Slovak Republic. SO_2 concentrations show a pronounced wind direction dependency. Also the CO concentrations vary with wind direction, although the concentrations are high at all wind directions. For NO_x only minor differences are observed at different wind directions which might be insignificant.

Measuring sites in the Slovak Republic are to a large extent influenced by the orography of the region. Due to the many hills, mountains and valleys wind directions at ground level might deviate quite far from the wind direction at transport height of distant plumes. At ground level the hills and valleys will determine wind direction (and wind velocity), whereas at transport height wind direction and wind velocity are mainly determined by large scale general circulation patterns. Large scale atmospheric transport is a result of these circulation patterns. This effects can not be expressed fully into the model because of the relatively coarse spatial resolution ($\pm$ 60 km). This complicates the analyses for the measuring sites within the Slovak Republic.

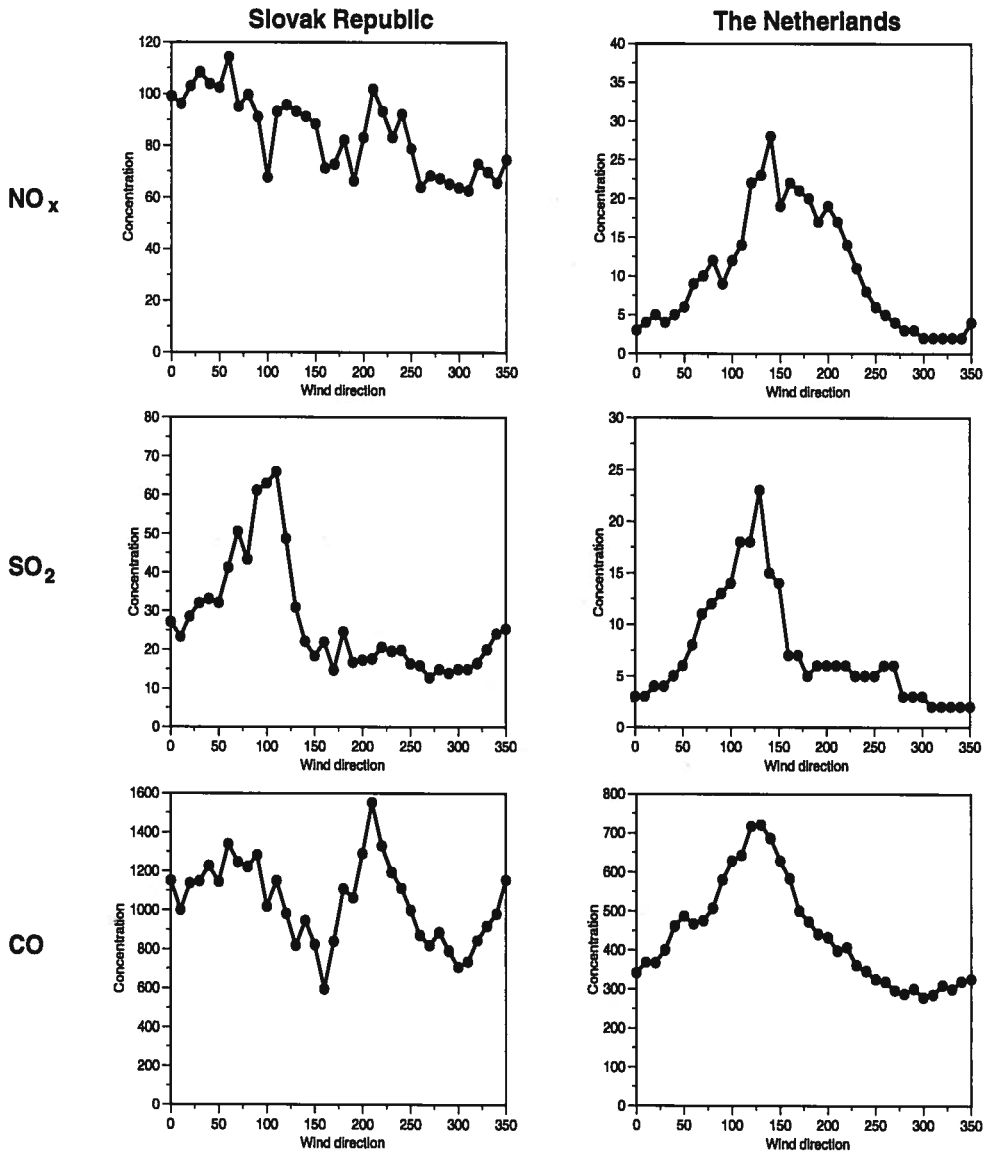


Figure 2. *Wind direction dependent concentrations of NO_x, SO_2 and CO in 1990 as measured in the Slovak Republic (left, latitude 48.0°, longitude 17.0°) and in The Netherlands (right, latitude 53.0°, longitude 5.0°)*

2.3.2 The Netherlands

Within the Netherlands the national air quality monitoring network is operated by the RIVM. Within this network about 80 multi-component measuring sites are installed in a more or less regular pattern over the country. At a number of locations some stations are added to the network to monitor more local characteristics. The network consists among others, of

a) about 80 measuring sites for SO_2
b) about 40 sites measuring NO_x

c) about 20 sites measuring CO.
Based upon meteorological conditions RIVM extracts from these measurements a number of typical wind direction dependencies in different regions within the Netherlands. By doing this very local effects are more or less averaged out of the wind direction dependencies. These results are used in this study.

Figure 2 (page 13, right column) shows the measured wind direction dependencies at a grid cell within the Netherlands. As can be seen for all three components a rather pronounced wind direction dependency is observed. Air pollutant concentrations are higher at south-eastern winds. This is caused by the geographical position of the Netherlands at the borders of the North Sea. As is the case in the Slovak Republic, CO concentrations are higher than a few hundred micrograms per cubic meter at all wind directions.

2.4 Statistical analyses

For the statistical investigation the data were analyzed in 36 10-degrees wind sectors. Averages of pollutant and number of cases per wind sector were further processed. From the measured data in Slovakia the concentrations with low wind velocity (calm) were excluded.

Different approaches were also applied in the evaluation of ground level wind parameters. In Slovakia the monitoring stations are placed in heavy polluted areas and cities. Therefore wind direction is strongly affected by surrounding buildings. Also unlike the Netherlands Slovakia is a hilly country. The representative wind roses for the particular regions were taken from the meteorological observatories. The number of analyzed stations in Slovakia was restricted mainly by the absence of meteorological observations in the vicinity. Also therefore the space distribution of the stations in the Netherlands is better than in Slovakia. For the first calculations were subjected 8 stations in Slovakia and 13 measured averaged wind direction dependencies in the Netherlands. The concentrations are measured 2 m above the surface. The model concentrations represent the air pollution in the lower surface layer.

Comparison between measured and calculated values is obtained by regression analyses on the wind direction dependent averaged concentrations.

3. Results

3.1 LOTOS model runs

3.1.1 Averaged concentrations in Europe

Figure 3a through Figure 3c present the annual averaged concentrations over Europe of NO_x, SO_2 and CO respectively as calculated by the LOTOS model.
High NO_x concentrations are observed in the densely populated areas around the southern North Sea and in Northern Italy. These might be due to the high traffic densities in these regions.

The highest levels of SO_2 are observed in Eastern Germany, southern Poland and Bohemia. The heavy industry and large power plants, operated on high sulphur fuels in this region are the main reason for this result.

CO concentrations are highest in northern Italy, but are rather high all over Europe.

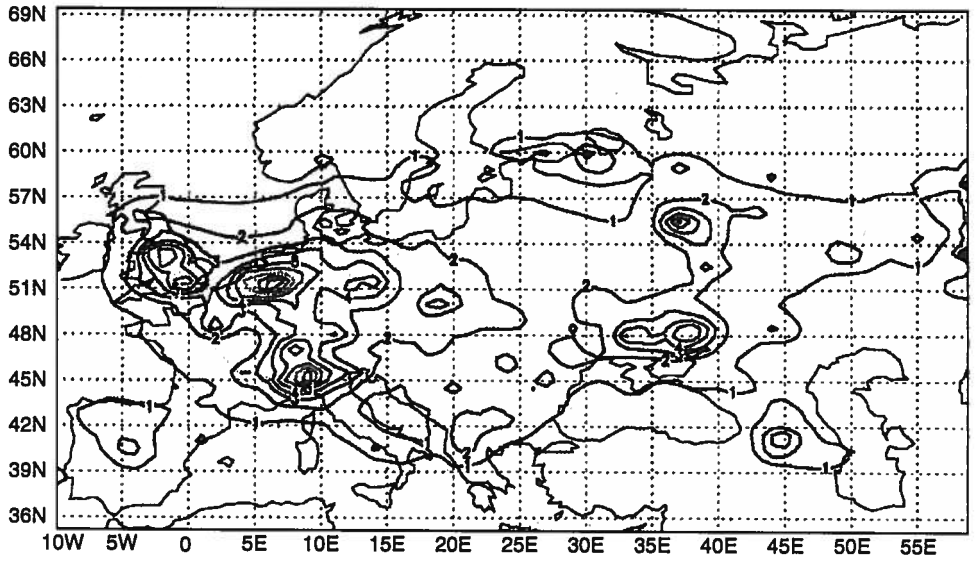


Figure 3.a Calculated annually averaged NO_x concentrations in Europe in 1990

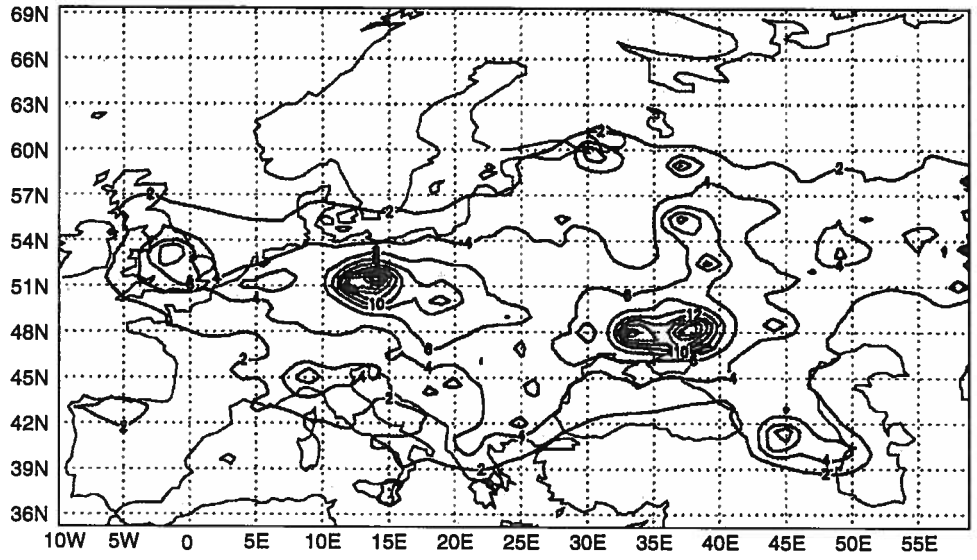


Figure 3.b Calculated annually averaged SO_2 concentrations in Europe in 1990

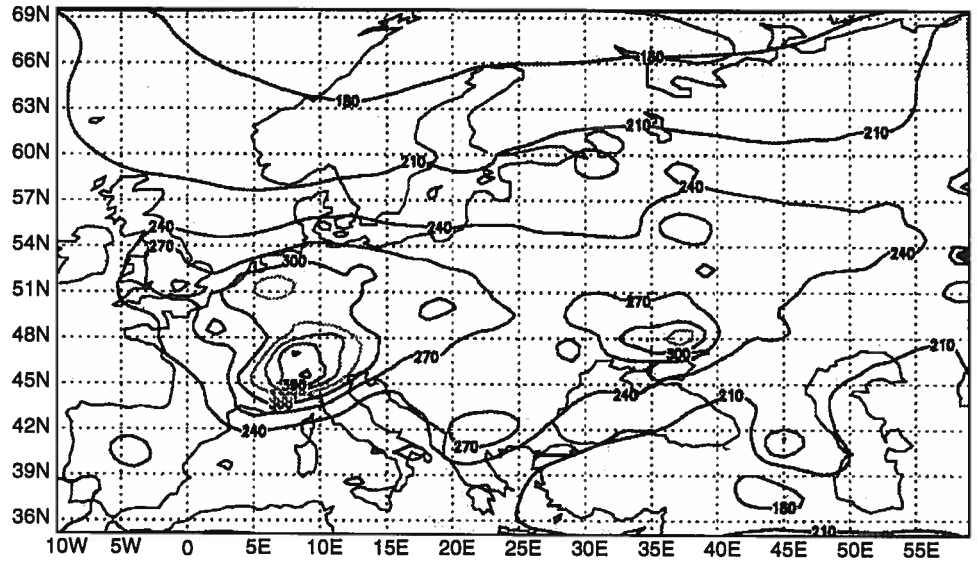


Figure 3.c Calculated annually averaged CO concentrations in Europe in 1990

3.1.2 Wind direction dependency

Figure 4 shows the calculated wind direction dependencies of NO_x, SO_2 and CO concentrations in the Slovak Republic and in the Netherlands at the same grid cells as those used in Figure 2.

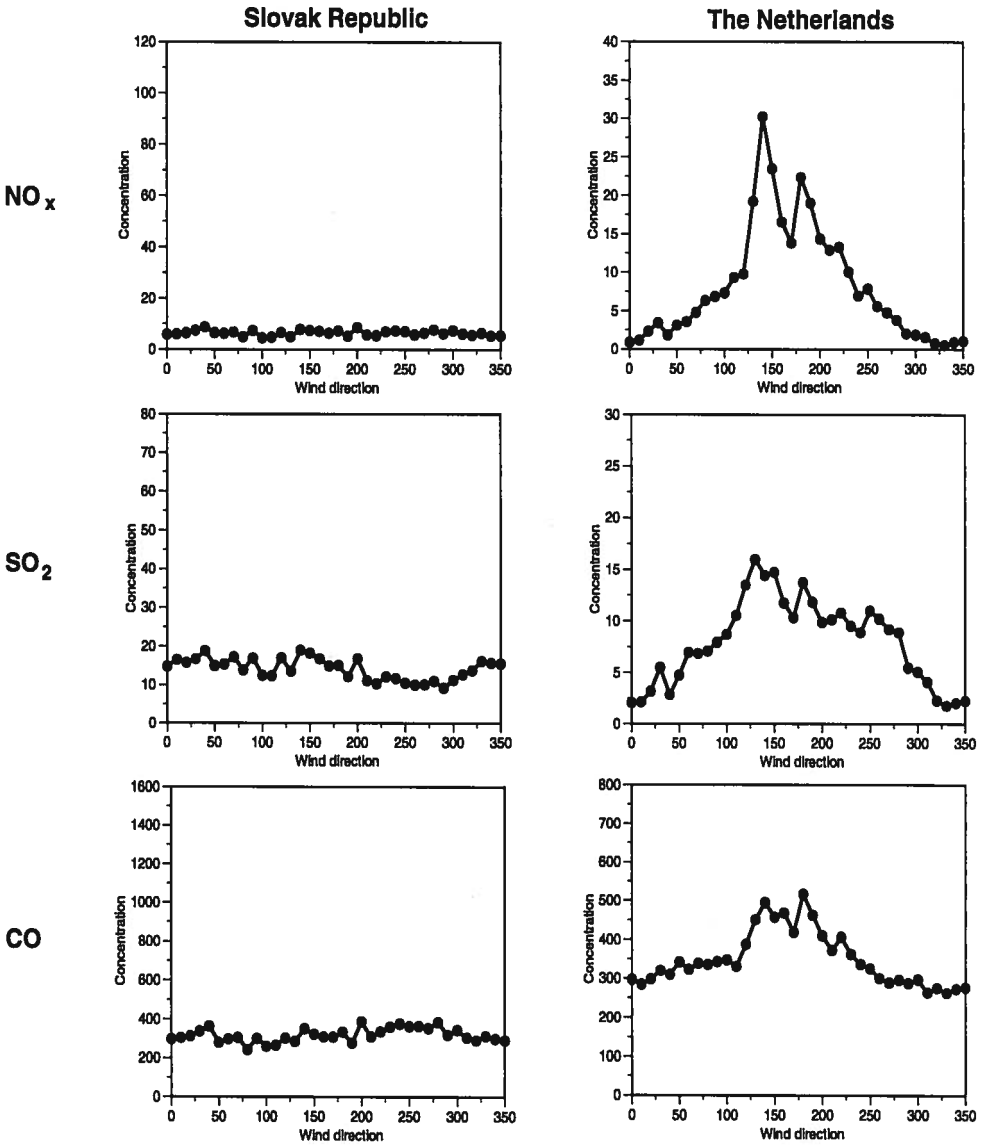


Figure 4. *Wind direction dependent concentrations of NO_x, SO_2 and CO in 1990 as calculated by LOTOS in the Slovak Republic (left, latitude 48.0°, longitude 17.0°) and in The Netherlands (right, latitude 53.0°, longitude 5.0°)*

As with the measurements, only little wind direction dependence is observed in the Slovak Republic. In the Netherlands the wind direction dependence looks similar, although not equal, to the ones measured. The next paragraph compares measurements and calculations in more detail and more quantitatively.

3.2 Comparisons with measurements

This paragraph presents the results of the comparisons between measured and calculated values. The results (regression analyses per component per grid cell) are presented in full detail in the Appendix.

3.2.1 Wind direction dependencies

To study the agreement of the measured and the calculated wind direction dependencies, correlation analyses were performed on the per wind sector averaged measurements and calculations per component and per location. This analysis is done in two steps:

a) comparison of the shape of the wind direction dependencies (this paragraph); and
b) comparison of the absolute values (paragraph 3.2.2)

Table 3. *Correlations between calculated and measured concentrations per grid cell in the Netherlands. For each component and each cell both the correlation coefficient (R) and the covariance (= R^2) are given. R-values above 0.388 are significant at the 99 % confidence level and are printed in bold.*

Latitude		3.0		4.0		5.0		6.0	
Longitude		R	R^2	R	R^2	R	R^2	R	R^2
53.0	NO_x					**.911**	0.830		
	SO_2					**.665**	0.442		
	CO					**.864**	0.747		
52.5	NO_x			**.910**	0.828			**.774**	0.599
	SO_2			**.800**	0.640			**.698**	0.487
	CO			**.885**	0.783			**.806**	0.650
52.0	NO_x			**.779**	0.607	**.703**	0.494	**.792**	0.627
	SO_2			**.784**	0.615	**.669**	0.448	**.626**	0.392
	CO			**.804**	0.646	**.735**	0.540	**.670**	0.449
51.5	NO_x	**.890**	0.792	**.830**	0.689	.315	0.099		
	SO_2	**.708**	0.501	**.796**	0.634	**.594**	0.353		
	CO	**.862**	0.743	**.845**	0.714	**.746**	0.557		
51.0	NO_x			**.889**	0.790	**.740**	0.548		
	SO_2			**.836**	0.699	**.621**	0.386		
	CO			**.831**	0.691	**.764**	0.584		
50.5	NO_x					**.687**	0.472		
	NO_x					**.647**	0.419		
	CO					**.791**	0.626		

Table 3 and Table 4 show results for the Netherlands and the Slovak Republic respectively. For each component and each cell both the correlation coefficient (R) and the covariance (= R2) are given. In this linear regressions with 36 samples (one at

each wind sector) correlations above 0.388 are significant at the 99 % confidence level. This means that with a certainty above 99 % it might be concluded that the observed correlation is not due to chance. The covariance is a measure for the amount of variance in both the measured values and in the calculated ones, that can be explained by a common cause.

Table 4. *Correlations between calculated and measured concentrations per grid cell in the Slovak Republic. For each component and each cell both the correlation coefficient (R) and the covariance (= R^2) are given. R-values above 0.388 are significant at the 99 % confidence level and are printed in bold.*

Latitude		17		18		19	20	21		22	
Longitude		R	R^2	R	R^2			R	R^2	R	R^2
48.5	NO_x							**.631**	0.398		
	SO_2							.155	0.024		
	CO							.172	0.030		
48.0	NO_x	.119	0.014	.007	0.000						
	SO_2	.298	0.089	-.182	0.033						
	CO	-.063	0.004								

The tables show that correlations in The Netherlands are quite good. All correlations except one are significant at the 99 % confidence level. This indicates that the wind direction dependency in this part of Europe of air pollution concentrations is consistent with the ones calculated from the CORINAIR 90 emissions inventory, using the LOTOS model. Highest correlations are observed for NO_x, whereas also the lowest value occurs for this pollutant.

In the Slovak Republic much lower values are observed. Only 1 correlation appears to be significant. Again this might indicate that the measured values in Slovakia are much more influenced by local peculiarities as can be modelled by LOTOS.

3.2.2 Absolute levels

By correlating the calculated concentrations with measured values, depending on wind direction, only the structure in the wind direction dependency is studied. Table 5 presents results of an analysis of the absolute values. If it is assumed that the model, including the emission database predicts the correct wind dependency, the regression analyses should yield regression lines with 0 intercept. The table gives the resulting slopes of such regression lines with 0-intercepts. The numbers in the table differ from the ones in the appendix because in this analysis the regression lines have forced 0 intercepts.

In the western and southern part of the Netherlands measured NO_x and CO concentrations appear to be significantly higher than the calculated ones, whereas in the eastern and northern parts the correspondence is better. This might be due to the in-

fluence of local emissions, which are not very well modelled in the about 60 x 60 km^2 grid cells of LOTOS. This result is more pronounced for NO_x as compared to CO. SO_2 concentrations appear to be underestimated with the exception of the grid cells in the south-eastern grid cells.

Table 5. Slopes of measured vs. calculated concentrations per grid cell in the Netherlands, assuming 0-intercepts.

		3.0	4.0	5.0	6.0
53.0	NO_x			1.103	
	SO_2			.897	
	CO			1.030	
52.5	NO_x		1.302		1.054
	SO_2		.684		.567
	CO		1.000		1.083
52.0	NO_x		1.897	1.782	1.229
	SO_2		.961	.986	.705
	CO		1.285	1.468	1.257
51.5	NO_x	1.874	2.056	1.478	
	SO_2	1.193	.868	.687	
	CO	1.315	1.226	1.399	
51.0	NO_x		1.546	1.646	
	SO_2		1.613	.691	
	CO		1.198	1.212	
50.5	NO_x			2.096	
	NO_x			.947	
	CO			1.218	

Figure 5, Figure 6 and Figure 7 show the wind direction dependent concentrations as calculated by LOTOS and as measured in grid cells over the Netherlands.

For NO_x the central grid cells show an underestimation of concentrations at south-eastern winds. This might indicate an underestimation of NO_x emissions from sources in this direction. Other grid cells show the same difference to a less pronounced extent. It is not very likely that adjacent grid cells would show this result if it was caused by local circumstances. In the northern grid cells the correspondence between measured and calculated values is remarkable.

In many grid cells the calculated values of SO_2 concentrations appear to be higher than the measured ones at several wind directions. The result however does not show a clear pattern. Local peculiarities might be the cause of this.

All grid cells show lower calculated CO concentrations at south-easterly winds as compared to the measurements. This effect is more clear in the southern grid cells than in the northern ones. Like for NO_x, this might indicate an underestimation of emissions in this direction.

The good correspondence in the Northern grid cells suggests that the differences between measured and calculated values are not caused by errors in the model. If the model would underestimate concentrations, this would most probably happen at all grid cells and probably also at all wind directions.

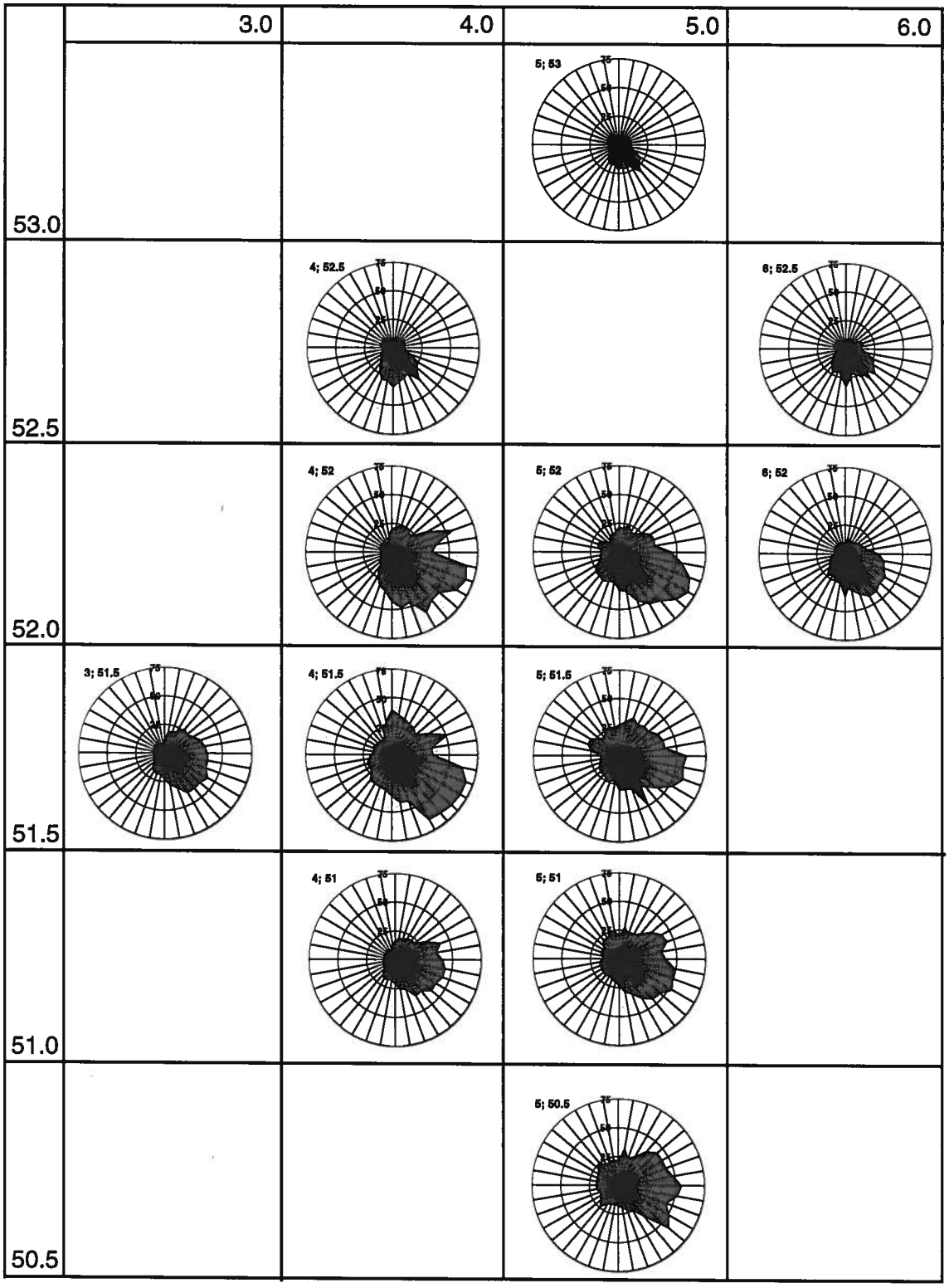


Figure 5. *Calculated (black) and measured (grey) wind roses for NO_x in the Netherlands. All plots are presented at the same scale.*

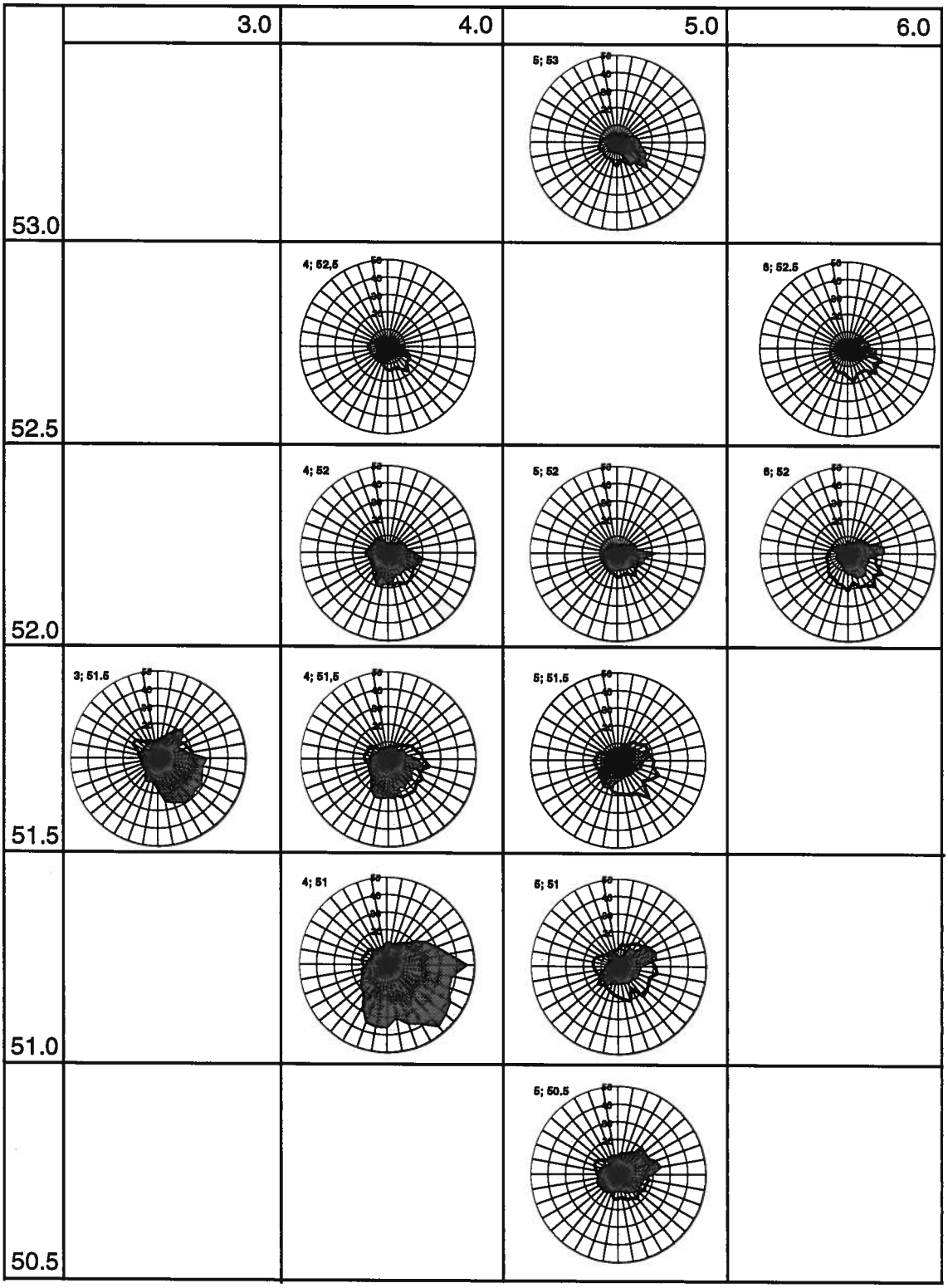


Figure 6. *Calculated (lines) and measured (grey) wind roses for SO_2 in the Netherlands. All plots are presented at the same scale.*

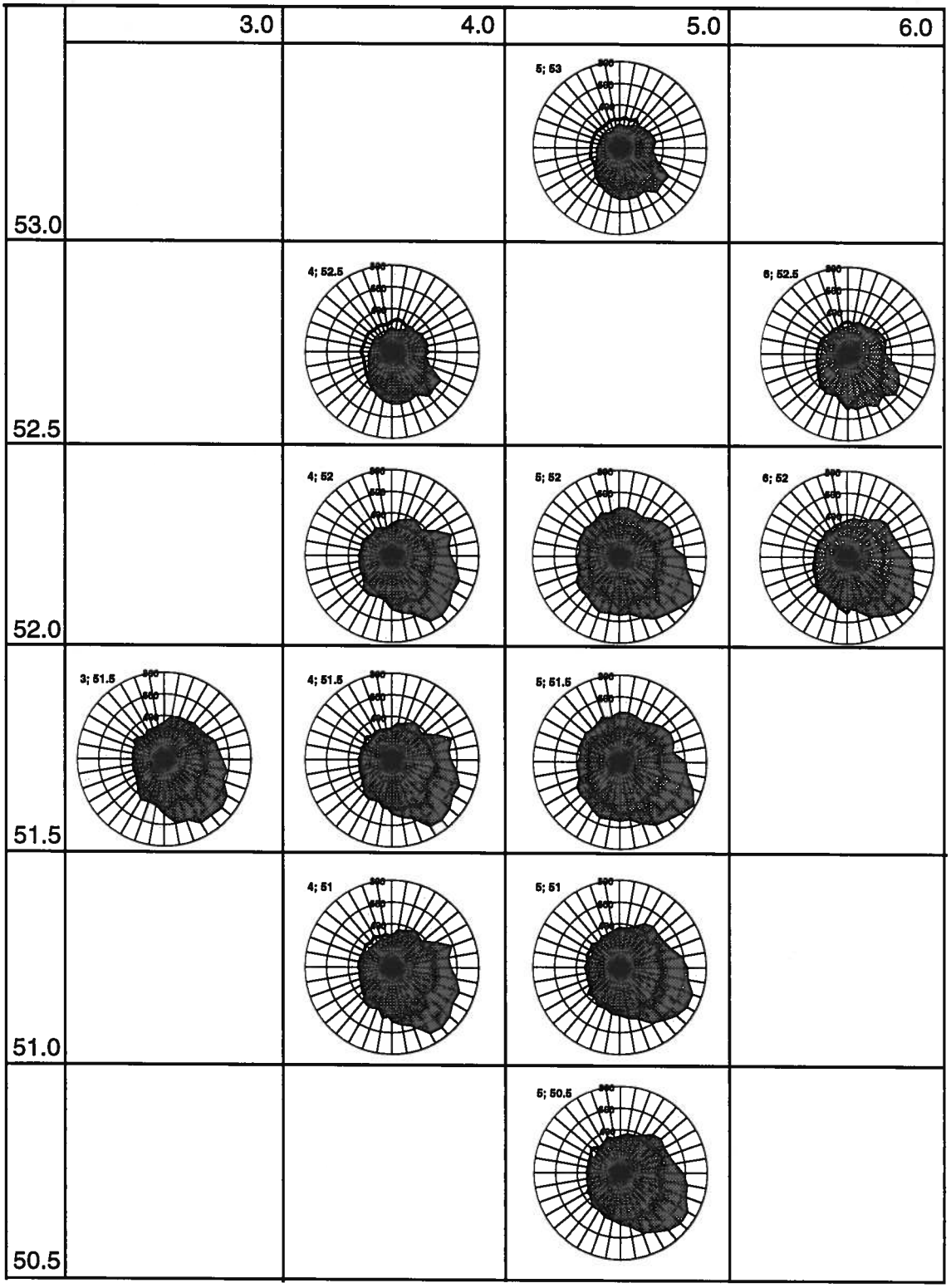
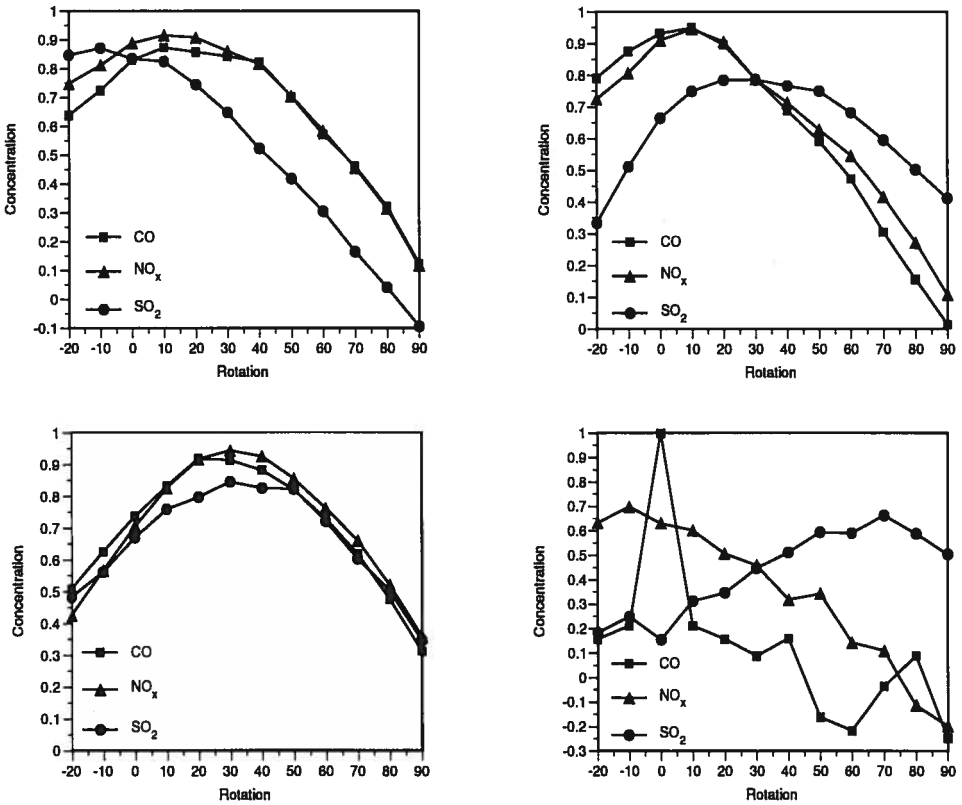


Figure 7. *Calculated (lines) and measured (grey) wind roses for CO in the Netherlands. All plots are presented at the same scale.*

3.2.3 Wind sheer

Transport of air pollutants can take place at higher altitudes, where normally wind directions deviate from the surface ones. This deviation might be as large as several tens of degrees (wind sheer). This phenomenon will result in a counter clockwise shift of the wind direction dependency if measured surface winds are used. This paragraph tries to quantify the effect of wind sheer by analyzing the correlation between measurements and calculations by rotating the first one with respect to the other.

Figure 8 shows an analysis, performed at 3 grid cells in the Netherlands and 1 grid cell in the Slovak Republic, in which the calculated concentration wind roses have been rotated relative to the measured wind roses. The graphs give the correlation coefficients as a function of this shift. Most curves for the Netherlands show an increase in the correlation coefficient by rotating the calculated values over angles of 20 to 30 degrees, indicating a wind sheer of about this values.



Upper Left: longitude: 4.0, latitude: 51.0
Upper Right: longitude: 5.0, latitude: 53.0
Lower Left: longitude: 5.0, latitude: 52.0
Lower Right: longitude: 22.0, latitude: 48.5

Figure 8. Correlations between measured and calculated concentrations at different rotations of measured wind roses.

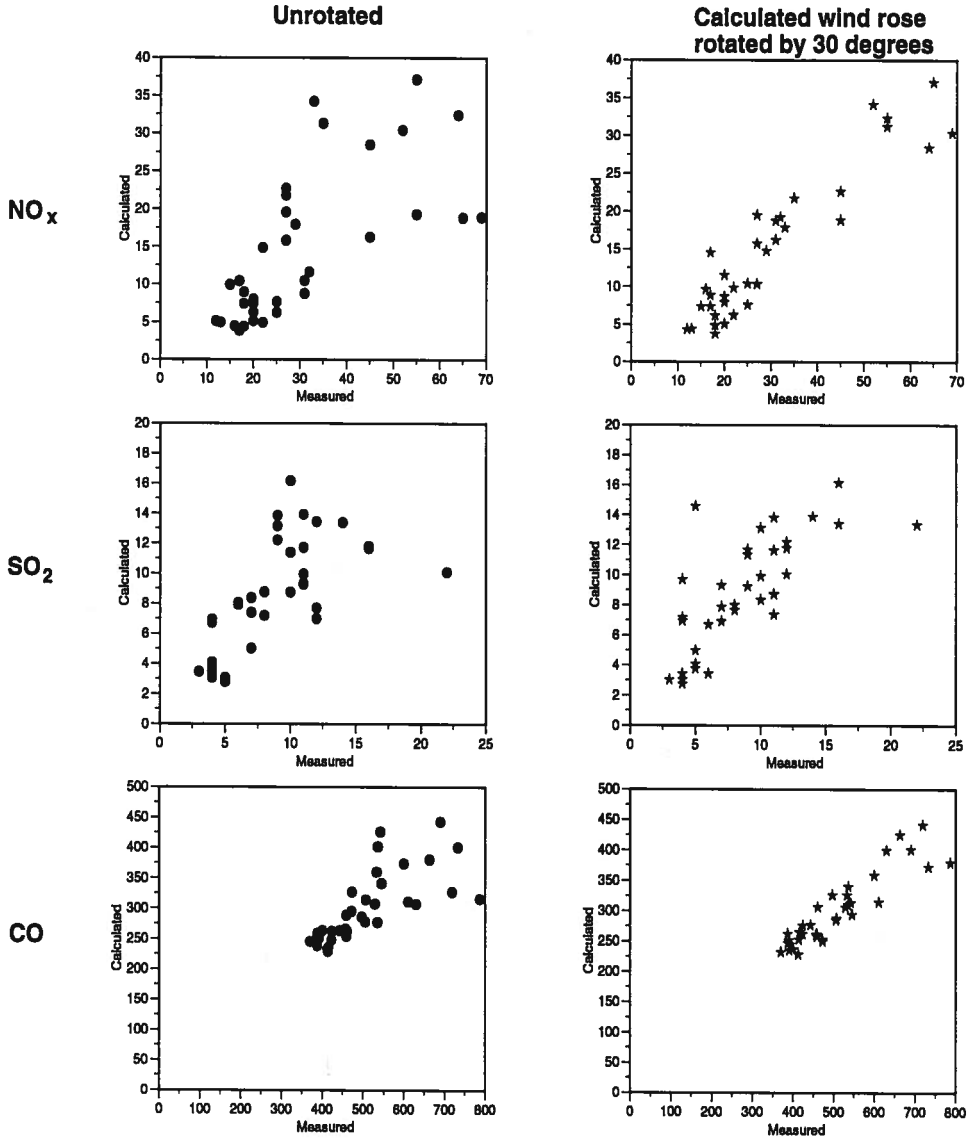


Figure 9. *Scatter plots (longitude 5, latitude 52, the Netherlands) for calculated vs. measured concentrations. Left: unrotated; right: calculated values rotated counter-clockwise over 30 degrees.*

The better correlations in the case of a rotation of 30 degrees (clockwise) of calculated relative to measured wind roses is also shown in Figure 9 at one grid cell in the Netherlands (longitude 5, latitude 52). The right graphs show narrower bands of data points and hence better correspondence for all three components.

3.3 Biogenic NMVOC emissions

Table 11 presents the comparison of the biogenic NMVOC emissions from the CORINAIR 90 inventory with the results from the LOTOS model (Veldt, 1991) and

the global GEIA emissions inventory (Guenther et al. 1995). The left part of table 6 presents the comparison for the biogenic emissions for forests (SNAP codes 11.01 and 11.02), as most of the reported CORINAIR emissions seem to deal with forests. The right part envisages all biogenic sources (SNAP codes 11.01 - 11.09).

The CORINAIR 90 data are compared with results from the LOTOS model (European scale) and the GEIA inventory (global scale). The LOTOS model only calculates forest emissions so far, while the GEIA inventory also includes pastures, shrublands etc. In the left part of only the forest emission data for CORINAIR, LOTOS and GEIA have been presented. For LOTOS two versions are at hand. The original model result version, without isoprene emissions from coniferous forests and a preliminary version, including first estimates of coniferous isoprene emissions. The right part of the table concerns the comparison of the overall results from the GEIA inventory and the biogenic NMVOC data for sector 11 of CORINAIR 90.

Both the GEIA and LOTOS results are based on the relations between temperature and the emission rate of NMVOC and between land use data and leaf mass, and combine these relations with information on the spatial distribution of vegetation/land use types and meteorological data. There are three major differences between the LOTOS and GEIA approaches:

- GEIA probably uses more up to date emission factors than the present version of the LOTOS model. The GEIA factors have been reviewed during the last few years and include a.o. recent results from the EUROTRAC programme. The LOTOS factors are mainly based on information available until the beginning of the nineties. The most striking difference is that the LOTOS model does not calculate isoprene emissions for coniferous species, while the GEIA model does. Also, the emission factors for other substances are higher in the GEIA model.
- GEIA uses land use information from the Olson database (0.5° x 0.5°). LOTOS uses forest statistics (e.g. UNEP) and a handmade forest distribution map based on national forests statistics, forest maps etc. (0.25° x 0.5°). The relation between land use coverage and standing crop (leaf mass) has been described by Veldt (1989) and Guenther et al. (1995). In general, the LOTOS model assumes larger amounts of leaf mass per area than the GEIA model.
- The calculation of emissions in LOTOS is based on hourly values for temperature per 0.5° x 1° grid cell, while the GEIA calculations are based on monthly averages per 0.5° x 0.5°.

From table 6 it can be seen for the CORINAIR 90 data that:

- Not all countries have been in the position to present information on their biogenic emissions. Large parts from the former USSR, Hungary and Norway are lacking.
- A number of countries have not reported biogenic emissions from non-forest vegetation (e.g. Bulgaria, The Netherlands, United Kingdom), while Switzerland only reported NMVOC emissions from non-forest SNAP-codes. The amounts of non-forest NMVOC emissions, whenever reported, generally is very low compared to emissions from forests.

Table 6. Comparison of country totals for biogenic NMVOC

COUNTRY	Forests only (tonnes NMVOC)								Biogenic total (tonnes NMVOC)		
	CORINAIR SNAP 11 01 & 11 02[2]	LOTOS	LOTOS[4]	GEIA	GEIA / COR.[2]	GEIA / LOT.	GEIA / LOT.[4]	LOT.[4] / COR.[2]	CORINAIR SNAP 11 01 to 11 09[3]	GEIA	GEIA / COR.[3]
Albania		6642	10000	46000			4.6			91453	
Austria	214874	64062	190000	120000	0.55	1.9	0.63	0.88	236877	227148	0.96
Belgium	29151	10687	22000	36000	1.2	3.4	1.6	0.75	29497	73410	2.5
Bulgaria	175076	62728	88000	300000	1.7	4.7	3.4	0.5	176079	401960	2.3
Czechoslovakia	127730	92186	230000	300000	2.4	3.3	1.3	1.8	127730	470066	3.7
Denmark	9308	3351	19000	31000	3.3	9.2	1.6	2.0	9308	42749	4.6
Finland	291943	182157	1100000	1000000	3.5	5.5	0.91	3.8	291943	1034094	3.5
France	405071	295515	440000	710000	1.8	2.4	1.6	1.1	461798	1856198	4.0
Germany	386391	220464	820000	740000	1.9	3.3	0.9	2.1	386391	1119128	2.9
Greece	370708	64899	66000	290000			4.4	0.18	392876	443812	1.1
Hungary		47254	51000	150000		3.1	2.9			313585	
Ireland	16600	3893	38000	18000	1.1	4.7	0.47	2.3	16600	130182	7.8
Italy	152182	155404	240000	680000			2.8	1.6	152182	1172145	7.7
Latvia	51706								51706		
Lithuania	4127								4127		
Luxembourg	1669	1997	3400	3900	2.3	1.9	1.1	2.0	1712	3885	2.3
Netherlands	3400	6102	15000	12000	3.4	1.9	0.8	4.4	3400	65582	19
Norway		56104	340000	410000		7.3	1.2			467840	
Poland	290100	171581	400000	540000	1.9	3.1	1.4	1.4	290100	941977	3.2
Portugal	341918	124263	124000	510000				0.36	438097	512400	1.2
Romania	150333	124618	170000	360000	2.4	2.9	2.1	1.1	150333	648390	4.3
Spain	736655	707075	720000	860000			1.2	0.98	775489	1962216	2.5
Sweden	268935	248988	1200000	1100000	4.0	4.4	0.92	4.5	270447	1124161	4.2
Switzerland		23975	73000	43000		1.8	0.59		606	102456	170
Turkey		316052									
United Kingdom	80014	37428	110000	75000	0.94	2.0	0.68	1.4	80014	380235	4.8
Yugoslavia		189387	230000	470000		2.5	2.0			863808	
Column total	**4107891**	**3216812**	**6699400**	**8804900**			**1.3**	**1.6**	**4347312**	**14448860**	
Total [1]	**4052058**	**2601373**	**6100000**	**7700000**	**1.91**	**3**	**1.3**	**1.5**	**2684411**	**9691290**	**3.6**

1 Total for countries listed in both CORINAIR and LOTOS

2 Sum of "non managed deciduous forests" and "non managed coniferous forests"

3 Sum of 3), "forest fires" and "natural grasslands" (=total natural NMVOC in CORINAIR)

4 LOTOS forest & preliminary isoprene estimate

Comparison for forests

On average LOTOS emission estimates are 1.5 times higher than the CORINAIR 90 data. Only Belgium, Bulgaria, Greece and Portugal reported larger forest emissions than calculated by LOTOS. The results vary considerably between the countries. For Austria, France, and Spain there is quite a good correspondence. The differences for the other countries can be quite large.

The estimated emissions from the GEIA inventory are 1.9 times higher than the CORINAIR 90 forest emission data. Only Austria and United Kingdom reported higher values to CORINAIR 90 for forest emissions than GEIA has calculated. For Ireland and United Kingdom there is a good correspondence between the two methods.

Comparison for all biogenic sources

When also the contribution from pastures, crops etc. to biogenic emissions are considered in the comparison, the difference between the CORINAIR 90 data and the GEIA inventory is even much more distinct. The GEIA results are 3.6 times higher than the data reported to CORINAIR 90. Only the data for Austria are in good resemblance. The data for Greece and Portugal seem to correspond well, but show dissimilarities for the forest emissions. The variation in difference between the countries is much larger than it is for only the forest emissions. The large number for Switzerland has to do with the lacking of CORINAIR 90 data for forest emissions.

4. Discussion and conclusions

This study aims at a validation of the CORINAIR 90 emission inventory by a comparison with measured air quality in the Netherlands and in the Slovak Republic. The CORINAIR 90 emission inventory has been used to calculate air quality on a ± 60 x 60 km^2 grid over Europe, using the LOTOS model. The calculations have been performed by using the 1990 meteorological situation. Application of the model results in calculated 1990 wind direction dependent concentrations of NO_x, SO_2 and CO, which are statistically compared with measurements of national air quality monitoring networks for the same year. The total emissions stored in CORINAIR appears to be in fair agreement with the LOTOS-emission database for most components.

The results for the grid cells within the Slovak Republic are not conclusive, probably because of local influences on the measurements that could not be reproduced by the relatively large scale LOTOS model. The location of measuring sites within Slovakia moreover, is primarily aimed at monitoring heavily polluted areas in stead of estimating large scale air pollution concentration fields.

Within the Netherlands a fair agreement between measurements and calculated air pollution concentrations is observed:

- Measurements and calculations agree mostly within a factor of 2. measured NO_x and CO concentrations appear to be higher than the calculated ones, whereas the SO_2 concentrations in most cases are lower.
- Correspondence is better in the more rural (Northern) parts of the country, where measurements are representative for large scale air pollution movements. Measured NO_x and CO concentrations differ less than 10 % from the calculated ones. The differences appear to be somewhat larger for SO_2.
- In the more densely populated and industrialized parts of the country differences between measurements and calculations are larger (upto a factor of 2). This might be due to the fact that for calculating concentrations fields in these heavily industrialized and densily populated aras the spatial resolution of the model (60 x 60 km^2) is too low.

For SO_2 no clear pattern in the wind direction dependence is recognized in the comparison. Differences between measurements and calculations are mainly attributed to local influences.

NO_x and CO concentrations appear to be underestimated at winds from south-easterly directions. At many grid cells calculated concentrations are lower than the measured ones, whereas at other wind directions correspondence is better. The difference seems to be larger for NO_x than for CO. This might indicate underestimation of emissions in areas to the south-east of the Netherlands. Further anayses will be needed to explore the causes of this differences. If indeed emissions in the direction as indicated are underestimated, comparisons in other grid cells (Belgium, France, Germany) might confirm this difference at other wind directions. On the other hand, increasing

the model emissions input at certain locations should decrease the difference between calculated and measured concentrations.

The results of the comparison in the Netherlands show that an inventory like CORINAIR can be used to calculate and predict large scale air quality to a satisfying extent. Apparently the Netherlands have a measuring setup that allows for measurement of large scale (60 km resolution) air pollution patterns. The CORINAIR data in combination with the LOTOS model can be used to analyse air quality on this scale. The CORINAIR 90 database therefore is a very useful tool for large scale air pollution policy: contributions to ground level pollutant concentrations by different source categories or geographical entities can be estimated by using this approach.

In Slovakia the situation is different. We were not able to use measured data at the same spatial representativity as obtained from the combined usage of CORINAIR emission data and the LOTOS model. This is the main reason for the mismatch between measured and calculated concentrations here. The Slovak network is primarily aimed at measuring local scale air quality which is not modelled correctly by either the scale of the model and of the inventory. This conclusion can be tested as soon as the Slovak Republic has regional scale measuring stations.

We conclude that the CORINAIR system now seems to work at the regional scale (NUTS-3, same order of magnitude as the 60 km grid of LOTOS) and produces reliable emission estimates. For air quality studies at the local level a spatially more detailed inventory and modelling system is needed. The CORINAIR system will (at the EU level) not be extended to the scale that is needed for such analyses of local air quality. However the CORINAIR methodology can be used at higher spatial resolutions (down to 1 x 1 km). The topic centre and EEA should help countries by providing emission factors and inventory compilation procedures, to produce these kind of inventories.

Biogenic NMVOC emissions stored in the CORINAIR 90 emission inventory have been compared with the LOTOS and GEIA estimates. The following conclusions are drawn:

- The LOTOS and GEIA calculation methods apply for all countries the same estimation procedure. Therefore, for both methods, the results for the different countries can be regarded as mutually comparable. The CORINAIR data show clear inconsistencies, which have to be subject for further examination.
- The NMVOC emissions for forests might well be underestimated by a factor of 1.5-1.9 on an average. The overall biogenic NMVOC emissions might well be underestimated by a factor of 3-4 on an average.
- The NMVOC data for CORINAIR 90 lack detailed information about speciation. Therefore, the applicability for studies on e.g. ozone formation will be hampered, as these will require information on the reactivity of the species.

5. References

[1] **CORINAIR 90** Summary Report nr. 1 (Final Draft), June 1995

[2] **P.J.H. Builtjes (1992)**, The LOTOS - Long Term Ozone Simulation project; Summary report, TNO report TNO-MW R 92/240.

[3] **Guenther, A., C.N. Hewitt, D. Erickson, R. Fall, C. Geron, T. Graedel, P. Harley, L. Klinger, M. Lerdau, W.A. McKay, T. Pierce, B. Scholes, R. Steinbrecher, R. Tallamraju, J. Tayler, and P. Zimmerman (1995)**. A global model of natural volatile organic compound emissions. Journal of Geophysical Research, Vol. 100, PP 8873-8892.

[4] **Olson, J. (1992)** World ecosystems (WE1.4). Digital raster data on a 10 minute geographic 1080 x 2160 grid. In: Global ecosystems database, Version 1.0; Disc A, NOAA, Boulder, CO.

[5] **Veldt, C. (1989)**. Leaf biomass data for the estimation of biogenic VOC emissions. TNO report 89-306.

[6] **Veldt, C. (1991)**. The use of biogenic VOC measurements in emission inventories. TNO report 91-323.

6. Authentication

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Period in which the research took place

November - December 1995

Signatures

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Date:
26 02 '96

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Dr. R. Guicherit
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Assessment

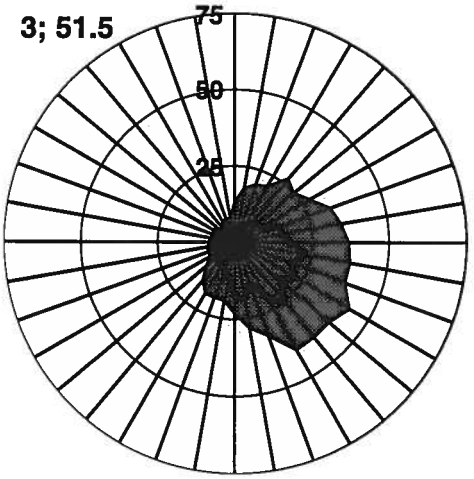
Date: 26-02-96

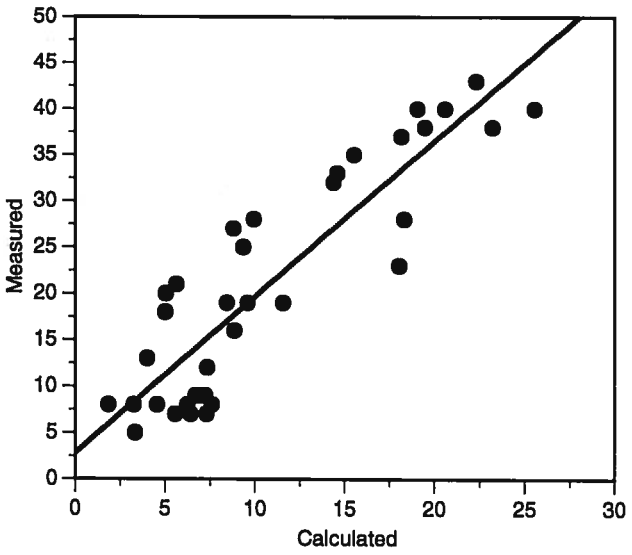
Appendix

Regressions between measured and calculated concentrations per component and per grid cell

Appendix A 1 *Measured and calculated concentrations of NO_x at 3 latitude and 51.5 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	2.13	4.00	13
10	2.67	5.01	18
20	2.68	5.04	20
30	3.00	5.63	21
40	4.69	8.80	27
50	4.98	9.35	25
60	5.29	9.94	28
70	7.77	14.60	33
80	8.27	15.54	35
90	9.68	18.17	37
100	12.36	23.21	38
110	10.37	19.47	38
120	11.89	22.31	43
130	10.15	19.06	40
140	13.61	25.55	40
150	10.97	20.60	40
160	7.66	14.39	32
170	9.76	18.32	28
180	9.61	18.05	23
190	6.17	11.58	19
200	5.11	9.59	19
210	4.49	8.43	19
220	4.71	8.85	16
230	3.91	7.33	12
240	3.84	7.22	9
250	3.64	6.84	9
260	3.56	6.69	9
270	4.04	7.59	8
280	3.88	7.29	7
290	3.42	6.41	7
300	3.32	6.23	8
310	2.96	5.56	7
320	1.77	3.33	5
330	1.73	3.25	8
340	2.42	4.55	8
350	0.98	1.84	8
		10.8	21.0
Linear regression		**6.5**	12.3
	Intercept	**2.837**	
	Slope	**1.681**	
	Correlation	**0.890**	
	Force 0	**1.8743**	

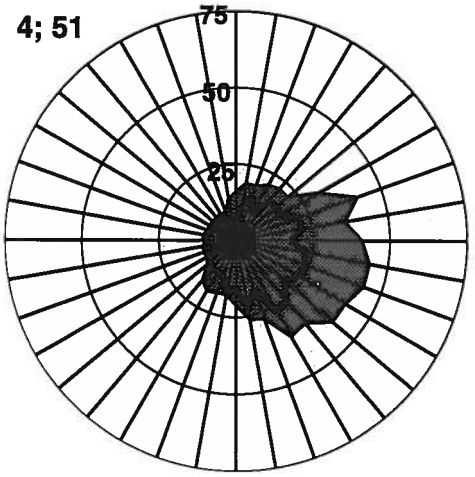


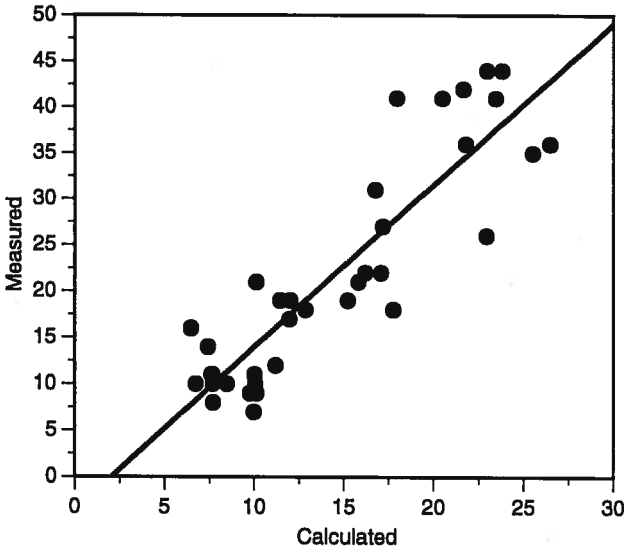


f(x) = 1.681E+0*x + 2.836E+0
R^2 = 7.923E-1

Appendix A 2 Measured and calculated concentrations of NO_x at 4 latitude and 51 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	3.45	6.48	16
10	6.09	11.44	19
20	8.11	15.22	19
30	5.38	10.11	21
40	8.62	16.19	22
50	9.09	17.06	22
60	8.92	16.75	31
70	11.53	21.64	42
80	11.60	21.78	36
90	9.56	17.95	41
100	12.22	22.95	44
110	12.67	23.79	44
120	12.49	23.44	41
130	10.91	20.47	41
140	13.58	25.50	35
150	14.10	26.46	36
160	9.14	17.17	27
170	12.21	22.92	26
180	8.42	15.81	21
190	9.45	17.75	18
200	6.84	12.85	18
210	6.39	12.00	19
220	6.36	11.95	17
230	5.95	11.18	12
240	5.33	10.01	11
250	4.08	7.66	11
260	4.06	7.61	11
270	5.35	10.05	10
280	5.38	10.11	9
290	4.51	8.47	10
300	4.09	7.68	10
310	5.20	9.76	9
320	4.10	7.70	8
330	5.31	9.97	7
340	3.59	6.74	10
350	3.95	7.42	14
		14.5	21.9
Linear regression		**6.1**	**12.1**
	Intercept	**-3.564**	
	Slope	**1.755**	
	Correlation	**0.889**	
	Force 0	**1.54583**	

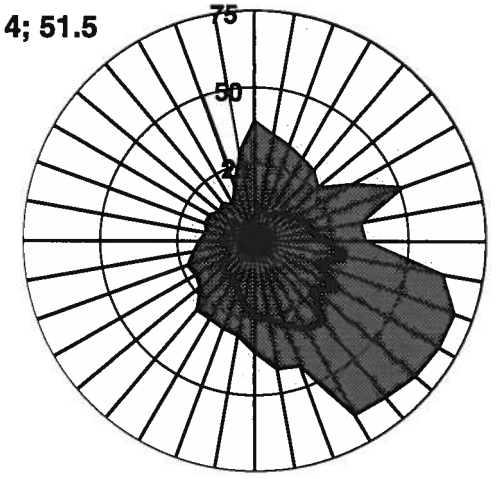


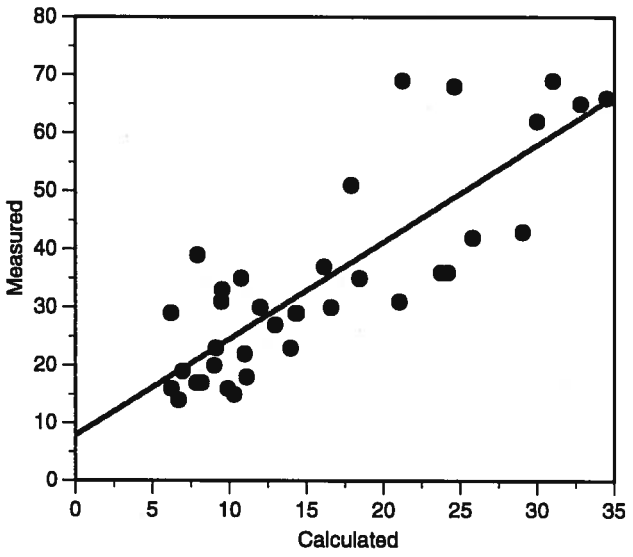


f(x) = 1.755E+0*x + -3.567E+0
R^2 = 7.912E-1

Appendix A 3 Measured and calculated concentrations of NO_x at 4 latitude and 51.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	4.21	7.91	39
10	5.73	10.76	35
20	5.07	9.52	33
30	5.04	9.47	31
40	6.39	12.00	30
50	6.90	12.96	27
60	8.60	16.14	37
70	9.53	17.90	51
80	9.82	18.44	35
90	12.64	23.74	36
100	15.97	29.99	62
110	11.31	21.24	69
120	16.52	31.02	69
130	13.11	24.62	68
140	18.39	34.52	66
150	17.47	32.81	65
160	15.47	29.05	43
170	13.74	25.79	42
180	12.89	24.20	36
190	11.20	21.03	31
200	8.83	16.59	30
210	7.60	14.28	29
220	7.65	14.37	29
230	7.44	13.96	23
240	5.85	10.99	22
250	4.86	9.12	23
260	4.80	9.01	20
270	5.92	11.12	18
280	5.27	9.89	16
290	4.18	7.85	17
300	4.36	8.18	17
310	5.49	10.30	15
320	3.31	6.22	16
330	3.56	6.69	14
340	3.70	6.95	19
350	3.30	6.20	29
		16.0	34.5
Linear regression		8.4	17.0
	Intercept	**7.856**	
	Slope	**1.669**	
	Correlation	**0.830**	
	Force 0	**2.05557**	

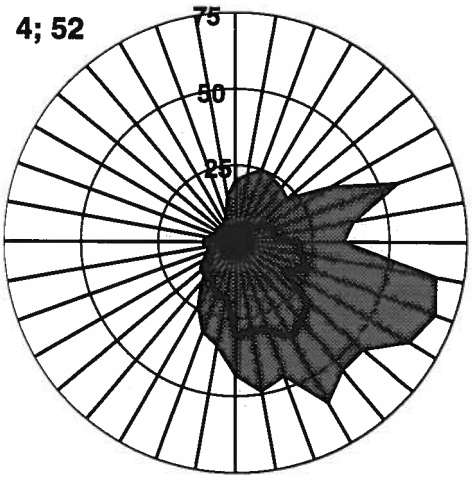


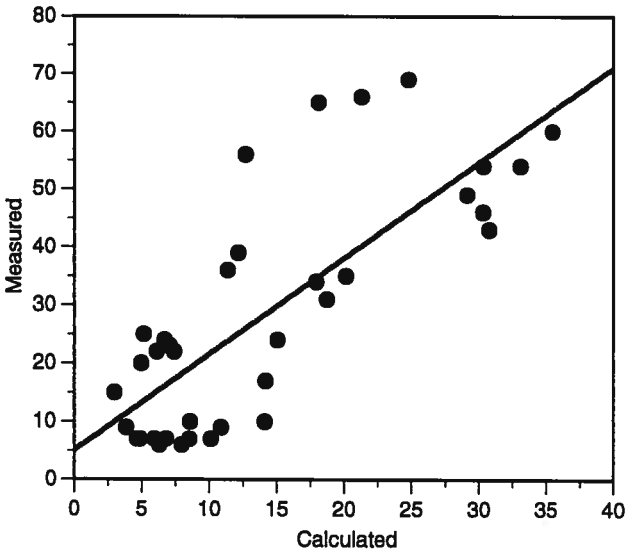


f(x) = 1.669E+0*x + 7.857E+0
R^2 = 6.892E-1

Appendix A 4 *Measured and calculated concentrations of NO_x at 4 latitude and 52 longitude.*

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	2.65	4.97	20
10	3.27	6.13	22
20	2.75	5.15	25
30	3.57	6.70	24
40	3.78	7.09	23
50	3.95	7.41	22
60	6.07	11.39	36
70	6.77	12.71	56
80	6.48	12.17	39
90	10.74	20.16	35
100	11.35	21.31	66
110	13.21	24.80	69
120	9.65	18.12	65
130	16.16	30.35	54
140	17.63	33.10	54
150	18.90	35.49	60
160	16.15	30.32	46
170	15.52	29.14	49
180	16.39	30.78	43
190	9.56	17.95	34
200	9.98	18.74	31
210	8.02	15.06	24
220	7.55	14.17	17
230	7.51	14.11	10
240	5.80	10.88	9
250	4.57	8.59	10
260	4.55	8.54	7
270	5.40	10.13	7
280	4.24	7.97	6
290	3.36	6.31	6
300	3.63	6.82	7
310	3.17	5.95	7
320	2.60	4.88	7
330	2.48	4.66	7
340	2.05	3.85	9
350	1.59	2.98	15
		14.1	28.4
Linear regression		9.6	20.3
	Intercept	**5.027**	
	Slope	**1.651**	
	Correlation	**0.779**	
	Force 0	**1.89677**	

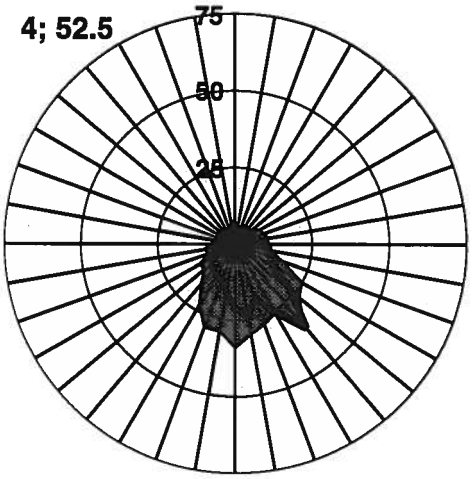


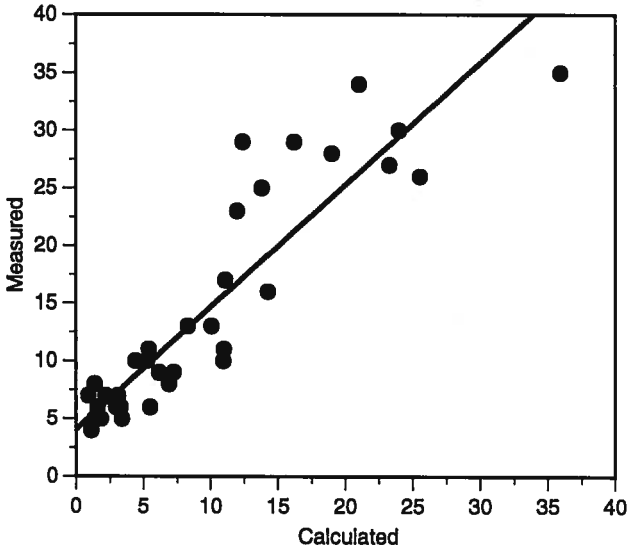


f(x) = 1.651E+0*x + 5.029E+0
R^2 = 6.063E-1

Appendix A 5 Measured and calculated concentrations of NO_x at 4 latitude and 52.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	0.74	1.39	8
10	0.99	1.85	5
20	1.72	3.23	6
30	1.17	2.19	7
40	1.58	2.96	6
50	1.66	3.12	7
60	3.29	6.17	9
70	2.36	4.43	10
80	2.88	5.40	11
90	5.84	10.96	10
100	5.86	11.01	11
110	7.61	14.28	16
120	7.37	13.83	25
130	12.41	23.30	27
140	19.14	35.94	35
150	13.61	25.55	26
160	10.13	19.02	28
170	12.78	24.00	30
180	11.21	21.05	34
190	8.64	16.23	29
200	6.61	12.41	29
210	6.38	11.99	23
220	5.92	11.11	17
230	5.37	10.08	13
240	4.43	8.32	13
250	3.86	7.25	9
260	2.82	5.29	10
270	3.68	6.91	8
280	2.94	5.51	6
290	1.81	3.39	5
300	1.82	3.42	5
310	1.77	3.32	6
320	0.86	1.61	6
330	0.61	1.14	4
340	0.75	1.40	5
350	0.49	0.92	7
		9.4	14.1
Linear regression		8.4	9.8
	Intercept	**4.050**	
	Slope	**1.059**	
	Correlation	**0.910**	
	Force 0	**1.30162**	

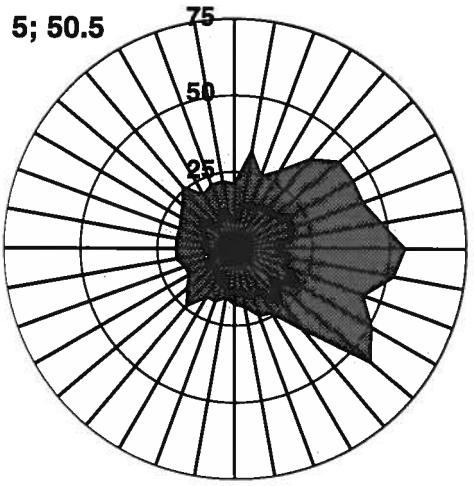


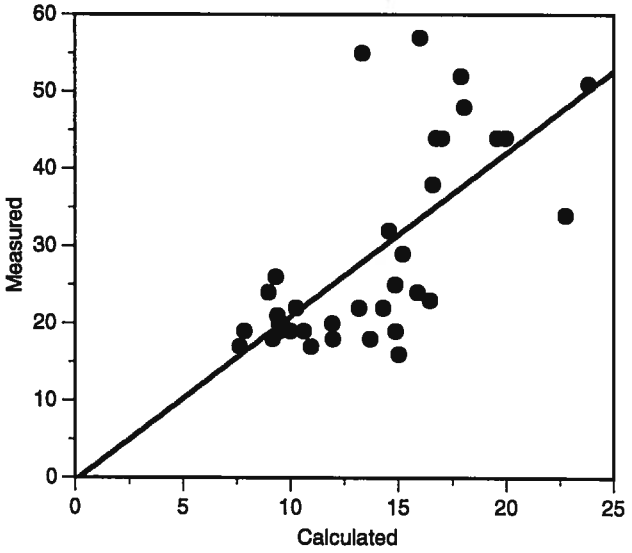


f(x) = 1.060E+0*x + 4.049E+0
R^2 = 8.289E-1

Appendix A 6 Measured and calculated concentrations of NO_x at 5 latitude and 50.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	5.01	9.41	21
10	7.77	14.58	32
20	7.92	14.87	25
30	8.11	15.22	29
40	8.84	16.60	38
50	8.93	16.76	44
60	10.63	19.97	44
70	10.43	19.58	44
80	9.62	18.05	48
90	7.10	13.34	55
100	9.53	17.90	52
110	9.07	17.02	44
120	12.69	23.83	51
130	8.53	16.01	57
140	12.12	22.76	34
150	8.47	15.90	24
160	8.77	16.47	23
170	7.93	14.89	19
180	7.30	13.71	18
190	8.00	15.03	16
200	4.89	9.18	18
210	6.38	11.98	18
220	4.79	8.99	24
230	5.05	9.47	20
240	5.84	10.96	17
250	4.07	7.65	17
260	4.19	7.87	19
270	5.33	10.01	19
280	5.06	9.51	19
290	5.66	10.63	19
300	6.37	11.95	20
310	7.62	14.31	22
320	4.97	9.33	26
330	5.13	9.62	20
340	7.03	13.19	22
350	5.47	10.27	22
		13.8	28.9
Linear regression		4.1	12.8
	Intercept	**-0.393**	
	Slope	**2.122**	
	Correlation	**0.687**	
	Force 0	**2.09559**	

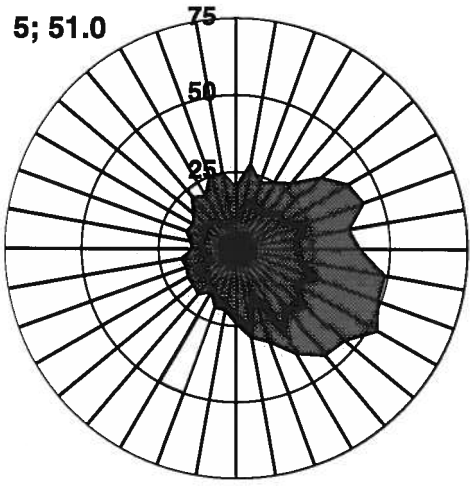


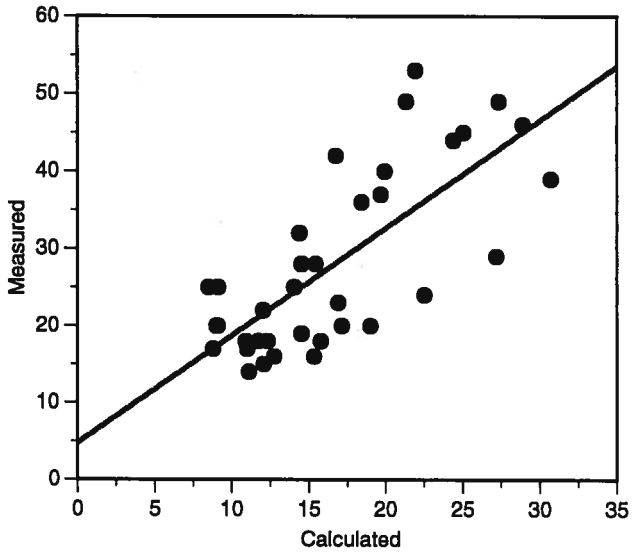


f(x) = 2.122E+0*x + -3.894E-1
R^2 = 4.718E-1

Appendix A 7 Measured and calculated concentrations of NO_x at 5 latitude and 51.0 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	4.84	9.09	20
10	7.73	14.52	28
20	9.00	16.90	23
30	7.49	14.05	25
40	8.21	15.42	28
50	9.82	18.43	36
60	8.91	16.74	42
70	12.98	24.38	44
80	10.48	19.68	37
90	10.62	19.94	40
100	11.35	21.32	49
110	14.56	27.33	49
120	11.67	21.92	53
130	15.39	28.89	46
140	13.34	25.04	45
150	16.35	30.70	39
160	7.66	14.38	32
170	14.47	27.17	29
180	11.99	22.51	24
190	10.12	19.00	20
200	9.12	17.13	20
210	8.16	15.32	16
220	7.73	14.51	19
230	6.56	12.32	18
240	8.40	15.76	18
250	4.68	8.78	17
260	5.80	10.90	18
270	5.92	11.11	14
280	6.43	12.07	15
290	5.86	11.00	17
300	6.79	12.74	16
310	6.23	11.71	18
320	6.41	12.04	22
330	4.79	9.00	20
340	4.52	8.49	25
350	4.85	9.11	25
		16.6	28.0
Linear regression		6.2	11.6
	Intercept	**4.786**	
	Slope	**1.393**	
	Correlation	**0.740**	
	Force 0	**1.64608**	

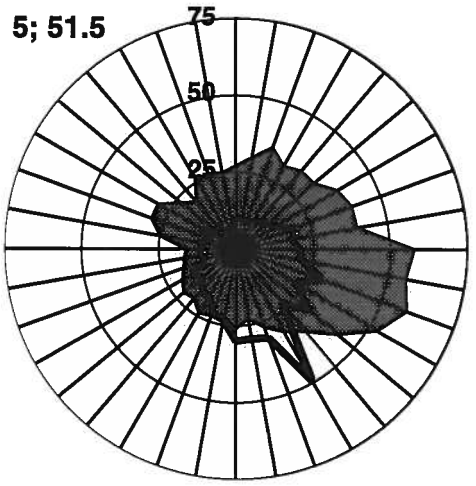


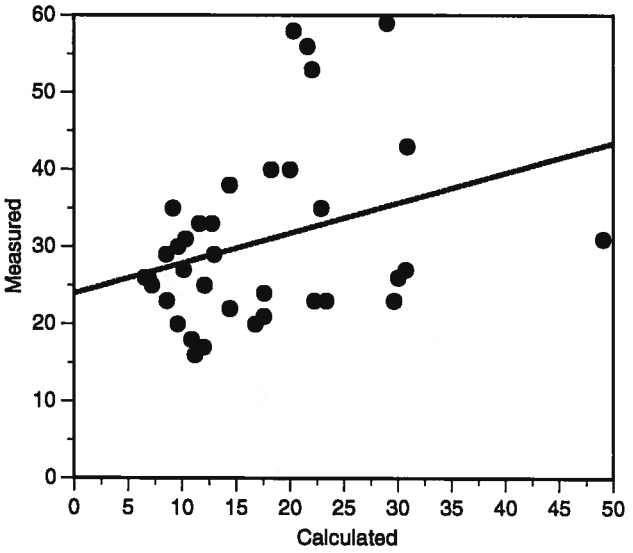


f(x) = 1.393E+0*x + 4.781E+0
R^2 = 5.480E-1

Appendix A 8 Measured and calculated concentrations of NO_x at 5 latitude and 51.5 longitude.

	ppb Calculated	ug/m^3 Calculated	Measured
0	5.41	10.16	27
10	5.14	9.65	30
20	4.89	9.18	35
30	5.51	10.34	31
40	6.19	11.62	33
50	6.80	12.77	33
60	7.68	14.41	38
70	10.66	20.01	40
80	9.73	18.26	40
90	10.84	20.35	58
100	11.52	21.64	56
110	15.45	29.01	59
120	11.76	22.07	53
130	16.45	30.89	43
140	12.20	22.91	35
150	26.14	49.08	31
160	16.37	30.74	27
170	15.79	29.65	23
180	16.01	30.06	26
190	12.44	23.36	23
200	11.86	22.27	23
210	9.38	17.61	24
220	9.36	17.58	21
230	7.68	14.42	22
240	8.93	16.78	20
250	5.79	10.88	18
260	6.39	11.99	17
270	5.97	11.20	16
280	6.44	12.09	25
290	4.56	8.56	29
300	6.92	12.98	29
310	4.58	8.60	23
320	5.11	9.60	20
330	3.50	6.56	26
340	3.68	6.90	26
350	3.84	7.21	25
		17.3	30.7
Linear regression		9.2	11.4
	Intercept	**23.969**	
	Slope	**0.390**	
	Correlation	**0.315**	
	Force 0	**1.47771**	

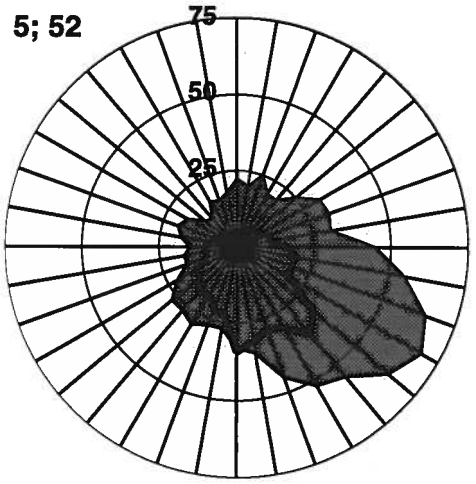


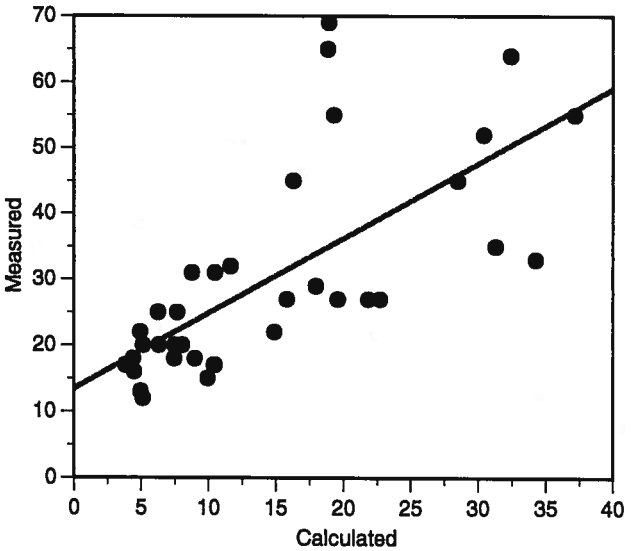


f(x) = 3.896E-1*x + 2.397E+1
R^2 = 9.948E-2

Appendix A 9 *Measured and calculated concentrations of NO_x at 5 latitude and 52 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	2.62	4.92	22
10	2.73	5.13	20
20	3.33	6.25	25
30	3.36	6.30	20
40	4.27	8.02	20
50	4.08	7.66	25
60	4.67	8.76	31
70	6.20	11.63	32
80	5.58	10.48	31
90	8.68	16.29	45
100	10.27	19.29	55
110	10.04	18.85	65
120	10.08	18.92	69
130	17.26	32.41	64
140	19.79	37.15	55
150	16.20	30.41	52
160	15.18	28.50	45
170	16.67	31.29	35
180	18.23	34.23	33
190	12.09	22.70	27
200	11.62	21.82	27
210	9.56	17.95	29
220	10.42	19.56	27
230	8.42	15.81	27
240	7.92	14.87	22
250	5.56	10.45	17
260	5.54	10.39	17
270	5.29	9.92	15
280	4.77	8.96	18
290	3.97	7.45	20
300	3.96	7.44	18
310	2.64	4.95	13
320	2.73	5.12	12
330	2.04	3.82	17
340	2.38	4.47	16
350	2.35	4.40	18
		14.6	30.1
Linear regression		9.7	15.8
	Intercept	**13.445**	
	Slope	**1.139**	
	Correlation	**0.703**	
	Force 0	**1.78177**	



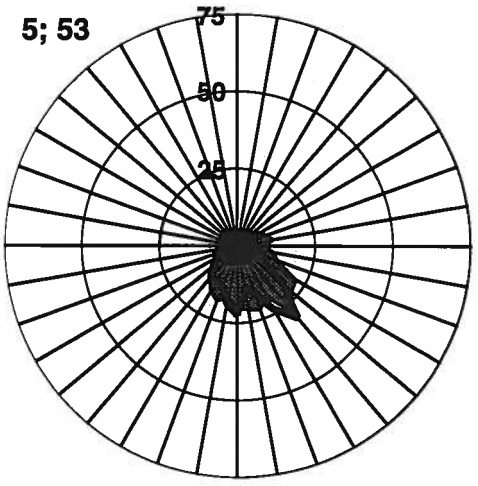


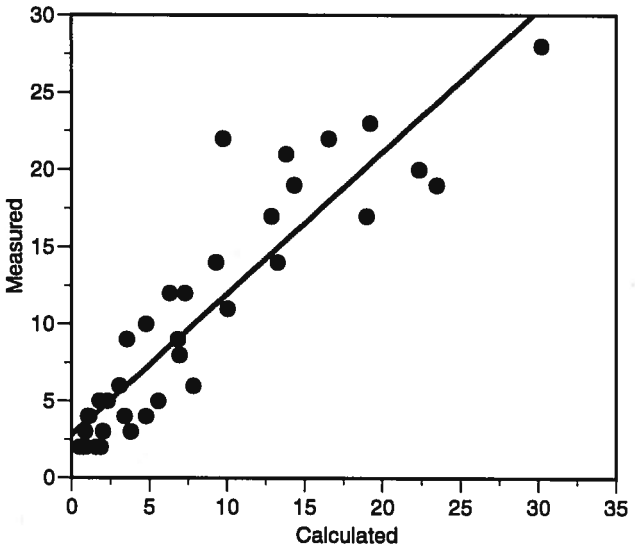
f(x) = 1.139E+0*x + 1.345E+1

R^2 = 4.945E-1

Appendix A 10 Measured and calculated concentrations of NO_x at 5 latitude and 53 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	0.46	0.87	3
10	0.61	1.14	4
20	1.23	2.31	5
30	1.81	3.40	4
40	0.96	1.80	5
50	1.64	3.08	6
60	1.89	3.55	9
70	2.54	4.78	10
80	3.37	6.32	12
90	3.64	6.84	9
100	3.89	7.30	12
110	4.96	9.31	14
120	5.19	9.75	22
130	10.23	19.22	23
140	16.09	30.20	28
150	12.50	23.47	19
160	8.81	16.55	22
170	7.36	13.82	21
180	11.90	22.35	20
190	10.11	18.99	17
200	7.64	14.34	19
210	6.86	12.88	17
220	7.07	13.27	14
230	5.35	10.04	11
240	3.70	6.95	8
250	4.18	7.84	6
260	2.97	5.57	5
270	2.54	4.77	4
280	2.02	3.79	3
290	1.07	2.01	3
300	1.00	1.87	2
310	0.83	1.57	2
320	0.41	0.76	2
330	0.28	0.52	2
340	0.48	0.91	2
350	0.56	1.06	4
		8.1	10.3
Linear regression		7.6	7.6
	Intercept	**2.780**	
	Slope	**0.917**	
	Correlation	**0.911**	
	Force 0	**1.10311**	

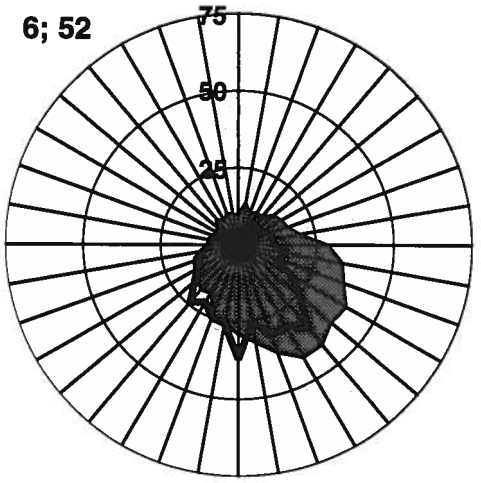


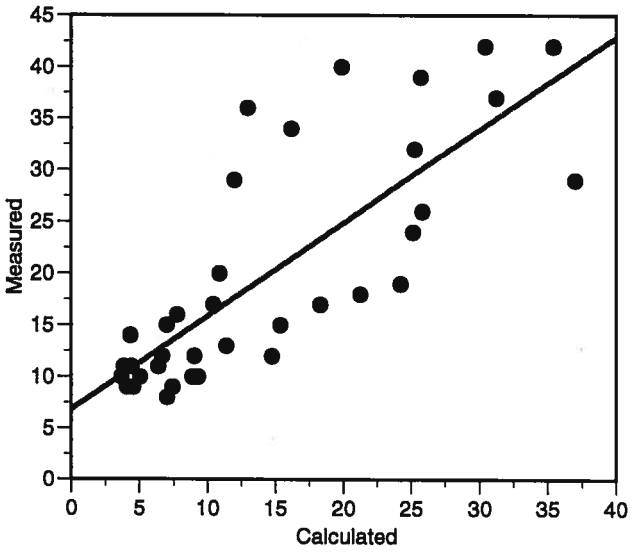


f(x) = 9.172E-1*x + 2.780E+0
R^2 = 8.298E-1

Appendix A 11 *Measured and calculated concentrations of NO_x at 6 latitude and 52 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	2.68	5.04	10
10	2.31	4.33	14
20	3.39	6.37	11
30	3.54	6.65	12
40	4.80	9.02	12
50	3.73	7.00	15
60	4.13	7.75	16
70	5.55	10.43	17
80	5.79	10.88	20
90	6.39	11.99	29
100	8.61	16.16	34
110	6.90	12.96	36
120	10.58	19.87	40
130	13.69	25.70	39
140	18.85	35.39	42
150	16.19	30.40	42
160	16.63	31.21	37
170	13.45	25.25	32
180	19.70	36.98	29
190	13.75	25.82	26
200	13.39	25.13	24
210	11.32	21.25	18
220	12.90	24.22	19
230	9.75	18.30	17
240	8.18	15.36	15
250	7.85	14.74	12
260	6.07	11.39	13
270	4.95	9.30	10
280	4.73	8.87	10
290	3.95	7.41	9
300	3.74	7.02	8
310	2.42	4.54	9
320	2.35	4.41	11
330	2.17	4.08	9
340	2.05	3.85	11
350	1.95	3.67	10
		14.5	19.9
Linear regression		9.8	11.1
	Intercept	**6.850**	
	Slope	**0.902**	
	Correlation	**0.792**	
	Force 0	**1.2291**	

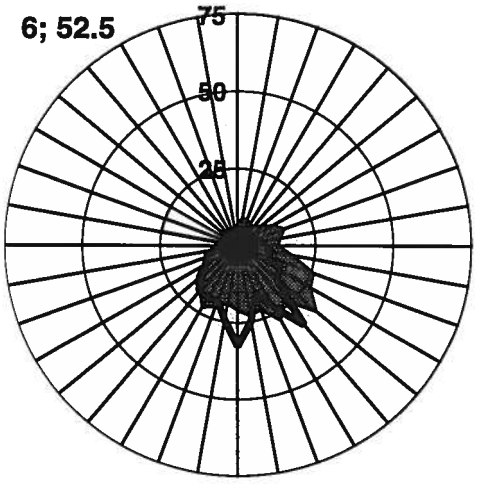


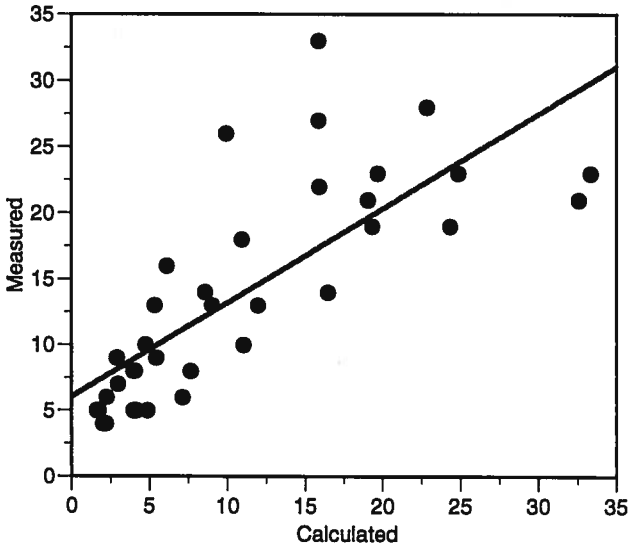


f(x) = 9.018E-1*x + 6.850E+0
R^2 = 6.269E-1

Appendix A 12 Measured and calculated concentrations of NO_x at 6 latitude and 52.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	1.58	2.96	7
10	1.54	2.89	9
20	2.89	5.42	9
30	2.10	3.94	8
40	2.16	4.05	8
50	2.52	4.73	10
60	2.84	5.33	13
70	3.25	6.11	16
80	4.55	8.55	14
90	4.79	8.99	13
100	5.82	10.93	18
110	5.28	9.92	26
120	8.45	15.87	27
130	8.45	15.86	33
140	17.76	33.35	23
150	12.17	22.85	28
160	8.47	15.91	22
170	13.23	24.84	23
180	17.36	32.59	21
190	12.96	24.34	19
200	10.15	19.06	21
210	10.49	19.69	23
220	10.29	19.31	19
230	8.77	16.47	14
240	6.37	11.97	13
250	5.88	11.04	10
260	4.07	7.65	8
270	3.80	7.13	6
280	2.58	4.85	5
290	2.21	4.16	5
300	2.11	3.96	5
310	1.07	2.00	4
320	0.85	1.60	5
330	1.17	2.19	4
340	0.92	1.73	5
350	1.19	2.24	6
		11.0	13.9
Linear regression		8.8	8.1
	Intercept	**6.062**	
	Slope	**0.714**	
	Correlation	**0.774**	
	Force 0	**1.05369**	

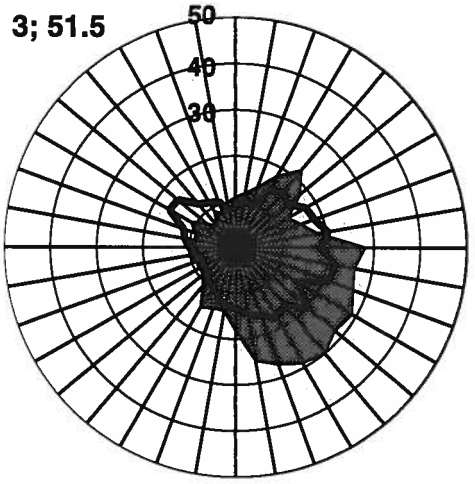


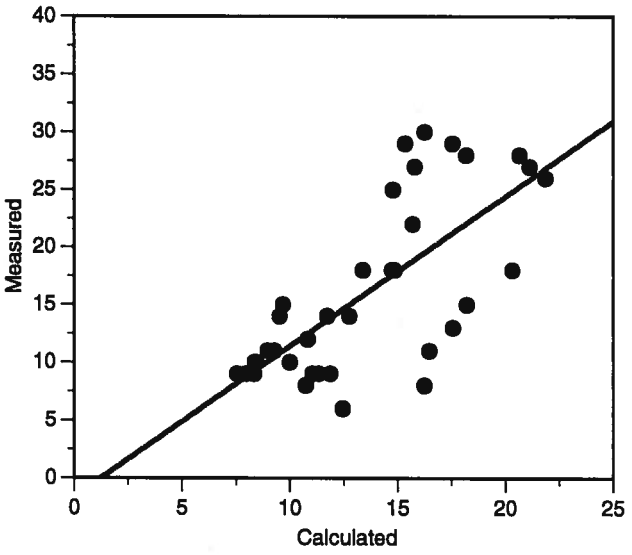


f(x) = 7.142E-1*x + 6.062E+0
R^2 = 5.990E-1

Appendix A 13 Measured and calculated concentrations of SO_2 at 3 latitude and 51.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	3.43	8.97	11
10	4.14	10.82	12
20	3.64	9.52	14
30	5.68	14.84	18
40	6.00	15.68	22
50	5.64	14.74	18
60	6.72	17.56	13
70	6.97	18.20	15
80	7.77	20.31	18
90	6.96	18.17	28
100	8.36	21.85	26
110	8.08	21.12	27
120	5.87	15.33	29
130	7.90	20.65	28
140	6.22	16.24	30
150	6.71	17.54	29
160	6.04	15.78	27
170	5.66	14.77	25
180	5.12	13.38	18
190	4.88	12.75	14
200	4.49	11.72	14
210	3.70	9.67	15
220	3.55	9.27	11
230	3.04	7.94	9
240	3.19	8.34	9
250	2.89	7.54	9
260	3.20	8.36	10
270	3.82	9.99	10
280	4.23	11.04	9
290	4.54	11.86	9
300	6.30	16.46	11
310	6.21	16.23	8
320	4.76	12.44	6
330	4.33	11.32	9
340	4.11	10.73	8
350	3.23	8.45	10
		13.6	16.1
Linear regression		4.2	7.6
	Intercept	**-1.624**	
	Slope	**1.302**	
	Correlation	**0.708**	
	Force 0	**1.19261**	

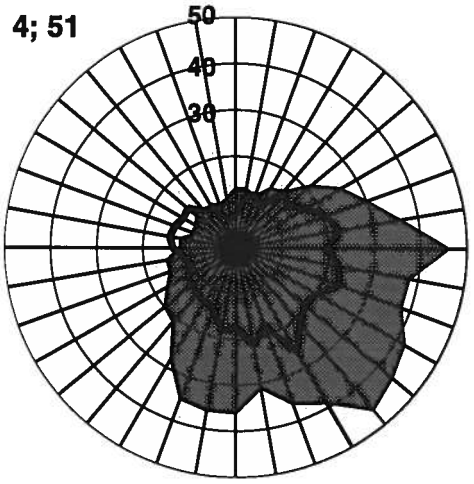


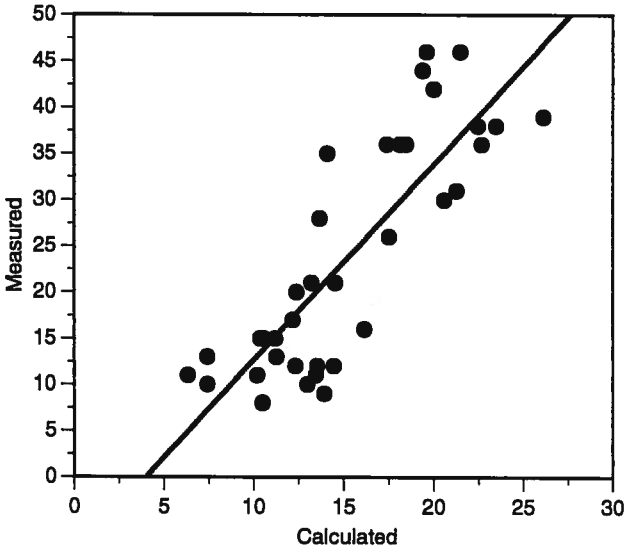


f(x) = 1.302E+0*x + -1.624E+0
R^2 = 5.010E-1

Appendix A 14 Measured and calculated concentrations of SO_2 at 4 latitude and 51 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	2.83	7.40	13
10	4.31	11.25	13
20	4.71	12.31	12
30	4.03	10.54	15
40	6.18	16.13	16
50	5.55	14.51	21
60	6.70	17.51	26
70	7.87	20.57	30
80	8.67	22.65	36
90	7.50	19.60	46
100	8.59	22.45	38
110	8.98	23.46	38
120	7.65	19.99	42
130	7.42	19.38	44
140	8.22	21.47	46
150	9.99	26.11	39
160	6.93	18.11	36
170	8.13	21.25	31
180	6.66	17.39	36
190	7.06	18.45	36
200	5.39	14.08	35
210	5.22	13.65	28
220	5.05	13.20	21
230	4.73	12.37	20
240	4.65	12.16	17
250	3.96	10.35	15
260	4.28	11.18	15
270	5.18	13.52	12
280	5.53	14.44	12
290	5.33	13.91	9
300	4.97	12.98	10
310	5.14	13.44	11
320	4.01	10.48	8
330	3.90	10.18	11
340	2.84	7.41	10
350	3.36	6.31	11
		15.3	23.9
Linear regression		5.0	12.6
	Intercept	**-8.484**	
	Slope	**2.116**	
	Correlation	**0.833**	
	Force 0	**1.61281**	

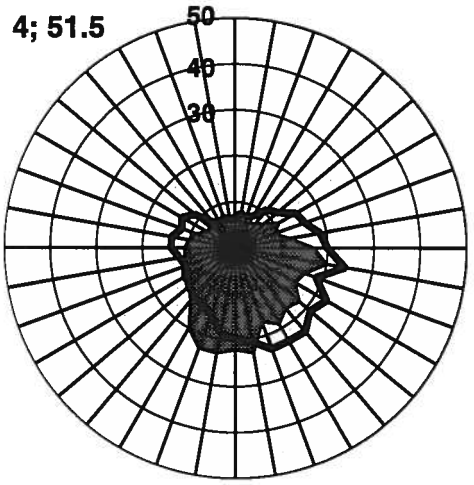


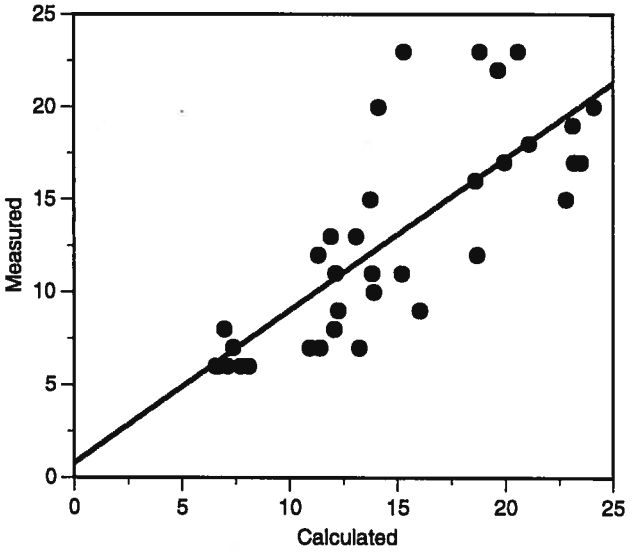


f(x) = 2.116E+0*x + -8.479E+0
R^2 = 6.945E-1

Appendix A 15 Measured and calculated concentrations of SO_2 at 4 latitude and 51.5 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	2.52	6.59	6
10	2.96	7.72	6
20	2.73	7.12	6
30	3.10	8.10	6
40	4.18	10.92	7
50	4.62	12.06	8
60	5.81	15.19	11
70	6.13	16.02	9
80	7.15	18.67	12
90	7.63	19.94	17
100	9.22	24.10	20
110	7.12	18.60	16
120	8.73	22.79	15
130	8.07	21.09	18
140	8.99	23.48	17
150	8.85	23.11	19
160	8.87	23.17	17
170	7.87	20.56	23
180	7.52	19.64	22
190	7.19	18.79	23
200	5.85	15.28	23
210	5.40	14.11	20
220	5.26	13.74	15
230	5.01	13.08	13
240	4.56	11.90	13
250	4.33	11.32	12
260	4.64	12.13	11
270	5.29	13.82	11
280	5.32	13.90	10
290	4.68	12.24	9
300	5.06	13.22	7
310	4.36	11.40	7
320	2.82	7.38	7
330	2.57	6.71	6
340	2.66	6.96	8
350	2.51	6.57	6
		14.5	12.7
Linear regression		5.6	5.7
	Intercept	**0.798**	
	Slope	**0.819**	
	Correlation	**0.796**	
	Force 0	**0.867636**	

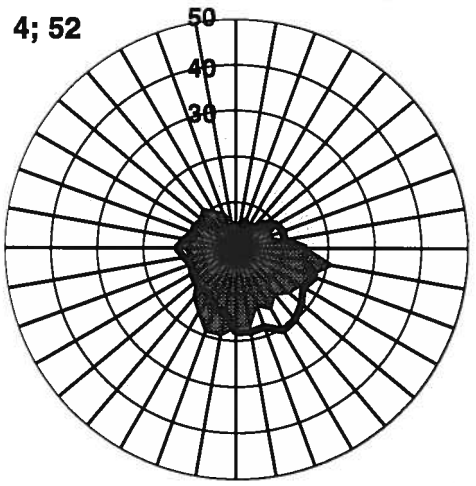


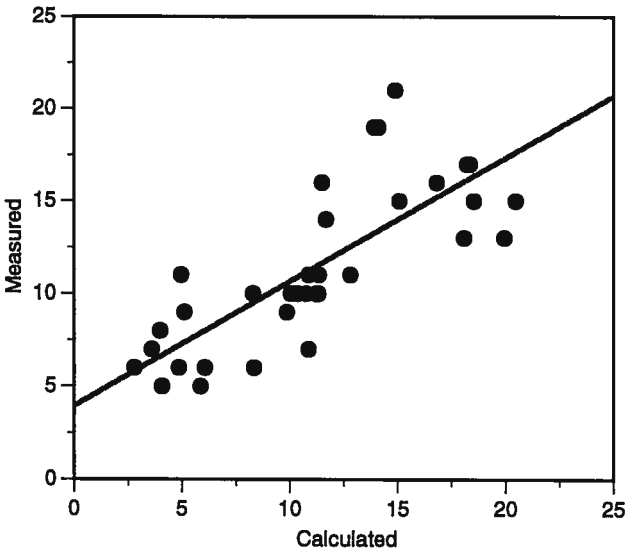


f(x) = 8.194E-1*x + 7.981E-1
R^2 = 6.335E-1

Appendix A 16 Measured and calculated concentrations of SO_2 at 4 latitude and 52 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	1.56	4.07	5
10	1.86	4.85	6
20	1.38	3.60	7
30	2.24	5.86	5
40	2.32	6.06	6
50	3.19	8.34	6
60	3.92	10.25	10
70	4.17	10.88	7
80	4.12	10.77	10
90	5.77	15.08	15
100	5.70	14.89	21
110	6.98	18.22	17
120	6.44	16.82	16
130	7.09	18.53	15
140	7.63	19.94	13
150	7.83	20.46	15
160	6.92	18.08	13
170	7.02	18.35	17
180	6.99	18.25	17
190	5.40	14.11	19
200	5.33	13.91	19
210	4.40	11.50	16
220	4.47	11.68	14
230	4.34	11.35	11
240	4.17	10.89	11
250	3.97	10.37	10
260	4.34	11.33	10
270	4.90	12.81	11
280	4.30	11.22	10
290	3.78	9.87	9
300	3.85	10.05	10
310	3.17	8.29	10
320	1.89	4.95	11
330	1.96	5.11	9
340	1.52	3.98	8
350	1.07	2.79	6
		11.3	11.5
Linear regression		5.1	4.4
	Intercept	**3.945**	
	Slope	**0.670**	
	Correlation	**0.784**	
	Force 0	**0.960613**	

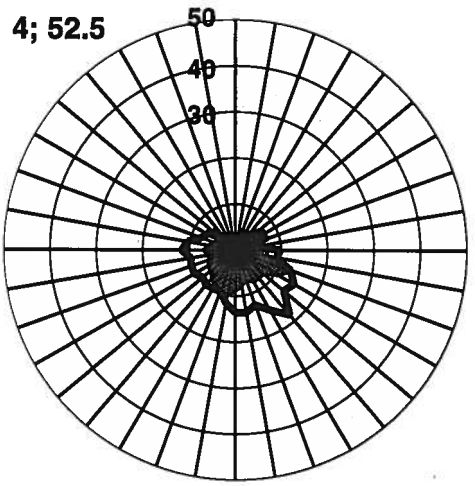


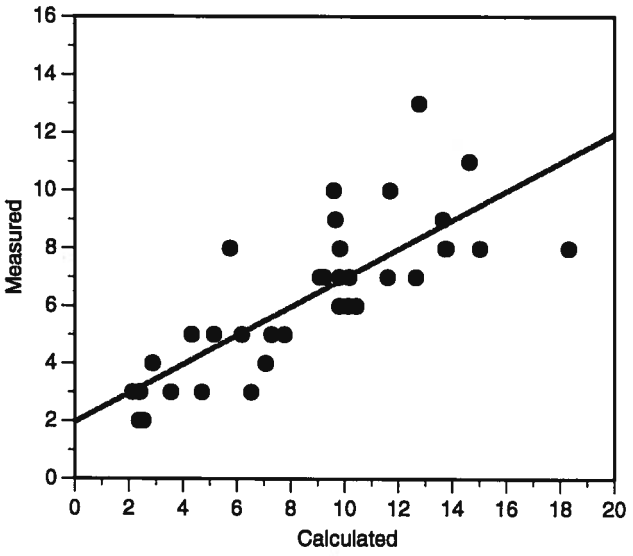


f(x) = 6.700E-1*x + 3.943E+0
R^2 = 6.149E-1

Appendix A 17 Measured and calculated concentrations of SO_2 at 4 latitude and 52.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	0.93	2.42	3
10	0.90	2.35	3
20	1.80	4.71	3
30	1.10	2.88	4
40	1.66	4.33	5
50	2.37	6.19	5
60	2.79	7.29	5
70	2.20	5.75	8
80	1.98	5.16	5
90	3.75	9.80	7
100	3.48	9.09	7
110	4.89	12.78	13
120	5.61	14.64	11
130	5.28	13.79	8
140	7.02	18.32	8
150	5.75	15.03	8
160	4.85	12.66	7
170	5.27	13.76	8
180	5.23	13.66	9
190	4.48	11.70	10
200	3.67	9.60	10
210	3.70	9.66	9
220	3.76	9.83	8
230	3.54	9.24	7
240	3.89	10.17	7
250	3.88	10.14	6
260	3.75	9.81	6
270	4.45	11.61	7
280	4.00	10.44	6
290	2.97	7.77	5
300	2.71	7.08	4
310	2.50	6.54	3
320	1.36	3.56	3
330	0.91	2.38	2
340	0.97	2.54	2
350	0.82	2.14	3
		8.6	6.3
Linear regression		4.3	2.7
	Intercept	**1.954**	
	Slope	**0.501**	
	Correlation	**0.800**	
	Force 0	**0.684229**	

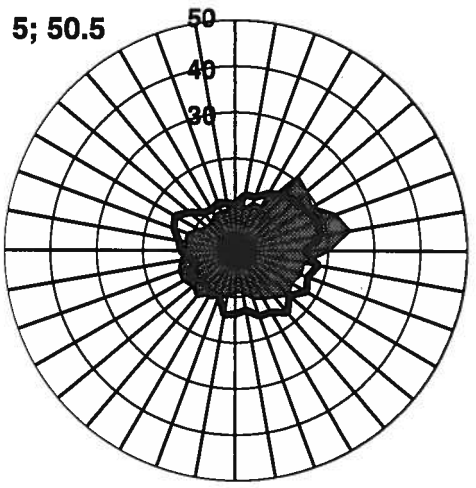


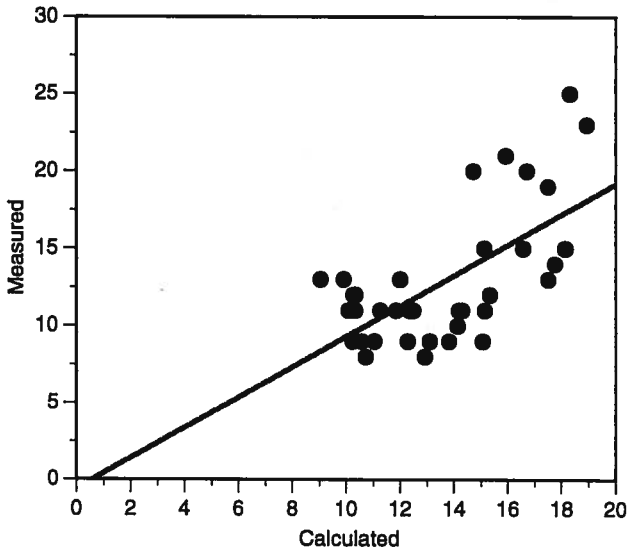


f(x) = 5.009E-1*x + 1.953E+0
R^2 = 6.406E-1

Appendix A 18 Measured and calculated concentrations of SO_2 at 5 latitude and 50.5 longitude.

	ppb Calculated	ug/m^3 Calculated	Measured
0	3.86	10.09	11
10	4.70	12.28	9
20	4.73	12.35	11
30	5.47	14.29	11
40	6.10	15.92	21
50	6.39	16.70	20
60	6.70	17.49	19
70	7.24	18.92	23
80	7.01	18.30	25
90	5.63	14.71	20
100	6.94	18.13	15
110	6.34	16.56	15
120	6.79	17.75	14
130	5.79	15.12	15
140	6.70	17.50	13
150	5.87	15.33	12
160	5.79	15.14	11
170	5.01	13.09	9
180	5.29	13.81	9
190	5.41	14.13	10
200	3.95	10.33	11
210	4.31	11.26	11
220	3.80	9.91	13
230	3.93	10.27	12
240	4.59	12.00	13
250	3.47	9.05	13
260	3.96	10.34	12
270	4.54	11.85	11
280	4.78	12.48	11
290	5.43	14.18	11
300	5.77	15.06	9
310	4.94	12.91	8
320	4.10	10.72	8
330	3.92	10.24	9
340	4.23	11.04	9
350	4.04	10.57	9
		13.6	12.9
Linear regression		2.9	4.4
	Intercept	**-0.574**	
	Slope	**0.988**	
	Correlation	**0.647**	
	Force 0	**0.947011**	

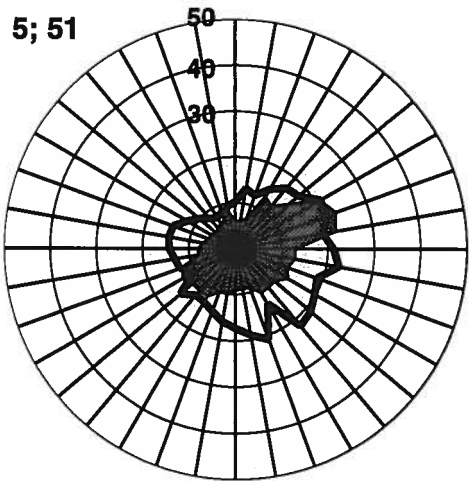


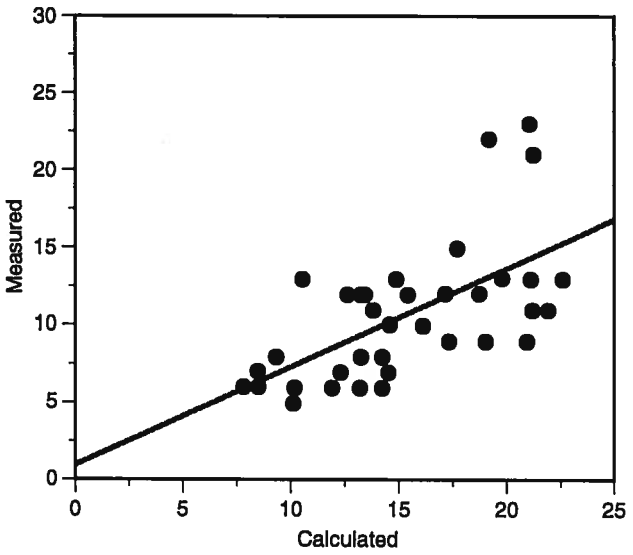


f(x) = 9.875E-1*x + -5.744E-1
R^2 = 4.186E-1

Appendix A 19 *Measured and calculated concentrations of SO_2 at 5 latitude and 51 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	3.90	10.19	6
10	5.06	13.21	6
20	4.72	12.32	7
30	5.70	14.90	13
40	6.57	17.16	12
50	6.79	17.73	15
60	8.13	21.24	21
70	8.06	21.06	23
80	7.34	19.19	22
90	8.09	21.13	13
100	8.66	22.61	13
110	7.17	18.73	12
120	7.58	19.79	13
130	8.12	21.21	11
140	8.39	21.93	11
150	5.30	13.83	11
160	8.02	20.95	9
170	7.29	19.04	9
180	6.64	17.34	9
190	6.18	16.14	10
200	5.58	14.58	10
210	5.15	13.45	12
220	5.07	13.25	12
230	5.91	15.44	12
240	4.05	10.57	13
250	4.84	12.65	12
260	5.08	13.26	8
270	5.45	14.25	8
280	5.56	14.53	7
290	5.45	14.24	6
300	4.57	11.93	6
310	3.88	10.13	5
320	3.26	8.50	6
330	2.99	7.82	6
340	3.25	8.48	7
350	3.57	9.34	8
		15.3	10.7
Linear regression		4.3	4.4
	Intercept	**0.945**	
	Slope	**0.634**	
	Correlation	**0.621**	
	Force 0	**0.691115**	

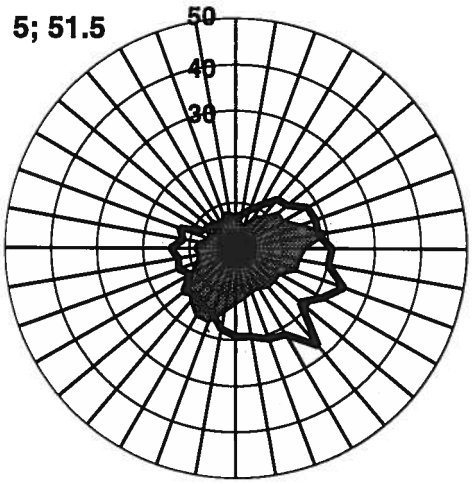


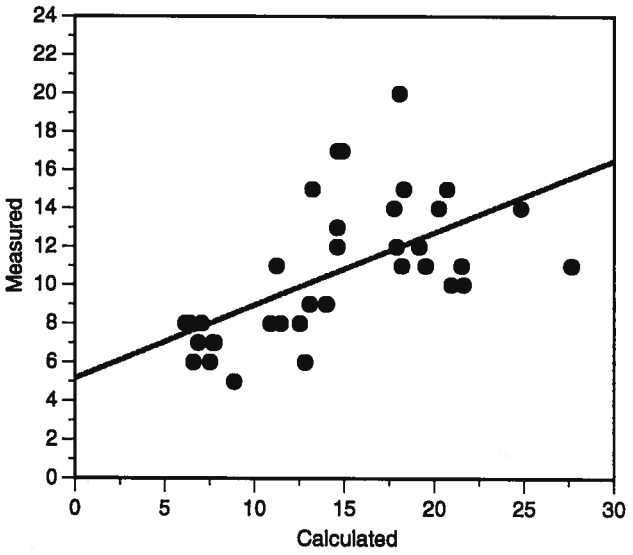


f(x) = 6.340E-1*x + 9.436E-1
R^2 = 3.856E-1

Appendix A 20 Measured and calculated concentrations of SO_2 at 5 latitude and 51.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	2.63	6.87	7
10	2.88	7.52	6
20	3.40	8.87	5
30	4.17	10.90	8
40	5.37	14.03	9
50	5.60	14.62	12
60	6.86	17.92	12
70	7.01	18.32	15
80	6.92	18.08	20
90	7.93	20.71	15
100	7.75	20.25	14
110	9.50	24.82	14
120	8.24	21.52	11
130	6.97	18.21	11
140	10.59	27.66	11
150	8.28	21.62	10
160	8.02	20.95	10
170	7.47	19.52	11
180	7.34	19.17	12
190	6.81	17.79	14
200	5.70	14.90	17
210	5.61	14.65	17
220	5.07	13.24	15
230	5.60	14.62	13
240	4.31	11.25	11
250	5.01	13.09	9
260	4.80	12.54	8
270	5.35	13.99	9
280	4.39	11.47	8
290	4.91	12.83	6
300	2.98	7.78	7
310	2.94	7.67	7
320	2.35	6.15	8
330	2.52	6.60	6
340	2.46	6.42	8
350	2.70	7.05	8
		14.5	10.7
Linear regression		5.7	3.6
	Intercept	**5.166**	
	Slope	**0.378**	
	Correlation	**0.594**	
	Force 0	**0.687468**	

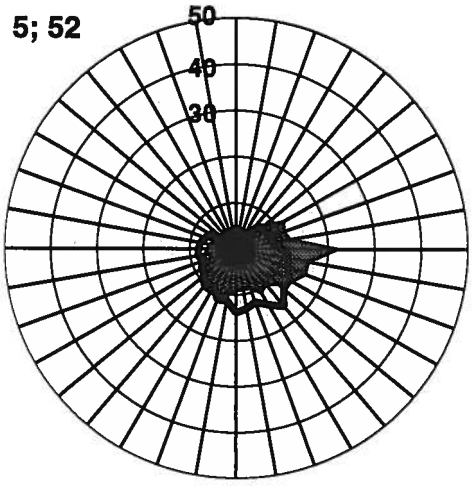


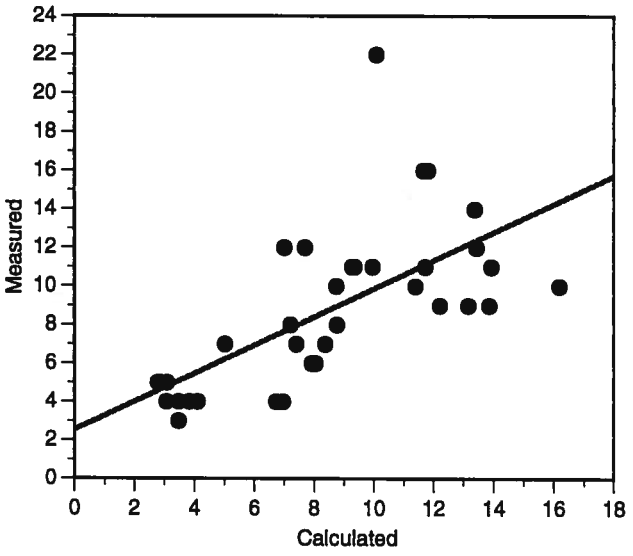


f(x) = 3.782E-1*x + 5.166E+0
R^2 = 3.530E-1

Appendix A 21 *Measured and calculated concentrations of SO_2 at 4 latitude and 52 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	1.53	2.87	5
10	1.53	2.87	5
20	2.03	3.82	4
30	2.19	4.10	4
40	2.67	5.02	7
50	3.83	7.20	8
60	3.73	7.00	12
70	4.98	9.34	11
80	4.10	7.69	12
90	5.37	10.08	22
100	6.21	11.66	16
110	6.29	11.81	16
120	7.12	13.37	14
130	7.15	13.43	12
140	8.62	16.18	10
150	7.41	13.92	11
160	6.50	12.21	9
170	7.01	13.16	9
180	7.37	13.85	9
190	6.06	11.39	10
200	6.24	11.72	11
210	4.94	9.27	11
220	5.30	9.95	11
230	4.66	8.74	10
240	4.67	8.76	8
250	3.94	7.40	7
260	4.46	8.37	7
270	4.28	8.04	6
280	4.22	7.92	6
290	3.70	6.95	4
300	3.59	6.73	4
310	1.85	3.47	3
320	1.85	3.47	4
330	1.48	2.78	5
340	1.64	3.07	4
350	1.64	3.08	5
		8.4	8.7
Linear regression		3.8	4.2
	Intercept	**2.547**	
	Slope	**0.733**	
	Correlation	**0.669**	
	Force 0	**0.985612**	

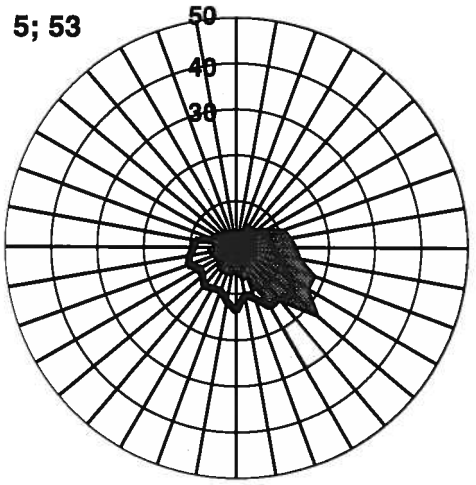


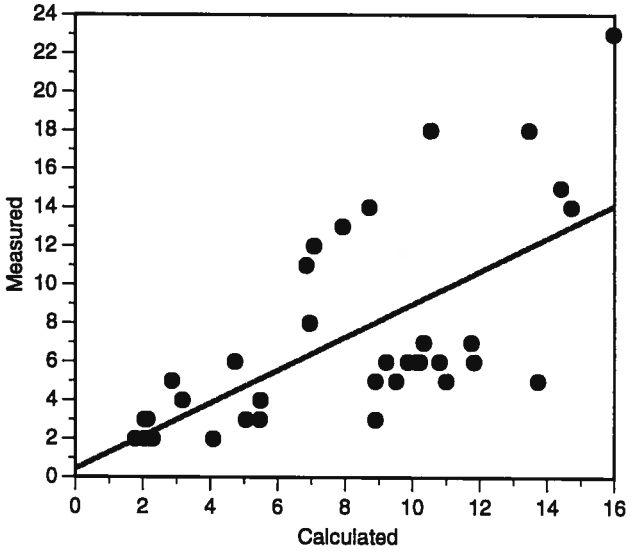


f(x) = 7.325E-1*x + 2.549E+0
R^2 = 4.467E-1

Appendix A 22 Measured and calculated concentrations of SO_2 at 5 latitude and 53 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	0.79	2.06	3
10	0.82	2.13	3
20	1.21	3.17	4
30	2.10	5.49	4
40	1.10	2.86	5
50	1.81	4.73	6
60	2.66	6.95	8
70	2.62	6.85	11
80	2.71	7.08	12
90	3.04	7.93	13
100	3.34	8.72	14
110	4.04	10.55	18
120	5.16	13.47	18
130	6.11	15.97	23
140	5.52	14.42	15
150	5.64	14.73	14
160	4.50	11.75	7
170	3.96	10.34	7
180	5.26	13.73	5
190	4.53	11.83	6
200	3.78	9.87	6
210	3.88	10.14	6
220	4.13	10.80	6
230	3.65	9.52	5
240	3.41	8.90	5
250	4.21	11.00	5
260	3.91	10.22	6
270	3.53	9.22	6
280	3.41	8.90	3
290	2.09	5.47	3
300	1.94	5.06	3
310	1.57	4.09	2
320	0.88	2.29	2
330	0.68	1.78	2
340	0.78	2.05	2
350	0.87	2.26	2
		8.0	7.2
Linear regression		4.1	5.3
	Intercept	**0.431**	
	Slope	**0.854**	
	Correlation	**0.665**	
	Force 0	**0.896839**	

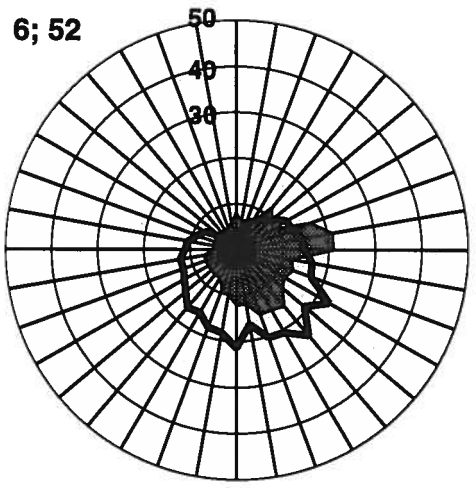


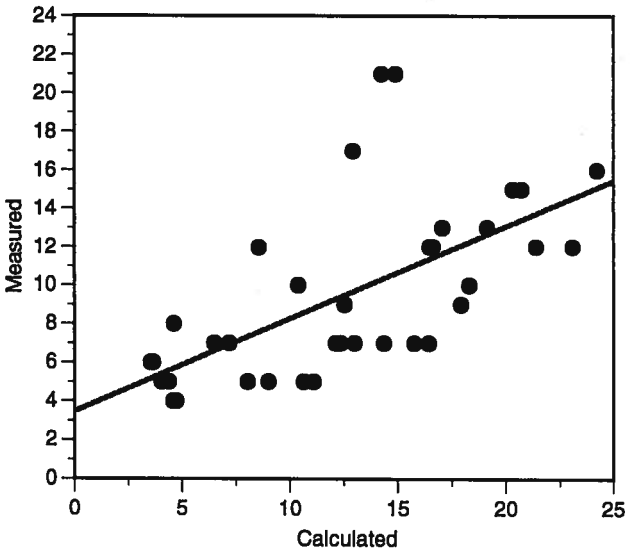


f(x) = 8.537E-1*x + 4.322E-1
R^2 = 4.426E-1

Appendix A 23 *Measured and calculated concentrations of SO_2 at 6 latitude and 52 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	1.76	4.61	8
10	1.35	3.54	6
20	2.48	6.49	7
30	2.75	7.18	7
40	3.27	8.55	12
50	3.97	10.38	10
60	4.79	12.52	9
70	4.94	12.91	17
80	5.45	14.24	21
90	5.70	14.88	21
100	6.36	16.61	12
110	6.53	17.05	13
120	8.84	23.08	12
130	7.32	19.12	13
140	9.27	24.22	16
150	7.92	20.70	15
160	7.78	20.31	15
170	6.31	16.47	12
180	8.19	21.38	12
190	7.01	18.30	10
200	6.86	17.92	9
210	6.03	15.76	7
220	6.29	16.42	7
230	5.49	14.35	7
240	4.97	12.98	7
250	4.64	12.12	7
260	4.72	12.34	7
270	4.25	11.09	5
280	4.07	10.64	5
290	3.45	9.00	5
300	3.07	8.03	5
310	1.81	4.72	4
320	1.75	4.58	4
330	1.68	4.38	5
340	1.55	4.04	5
350	1.39	3.64	6
		12.6	9.5
Linear regression		6.0	4.6
	Intercept	**3.475**	
	Slope	**0.479**	
	Correlation	**0.626**	
	Force 0	**0.705027**	

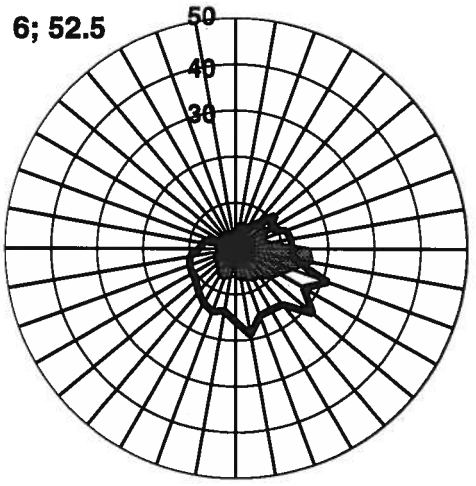


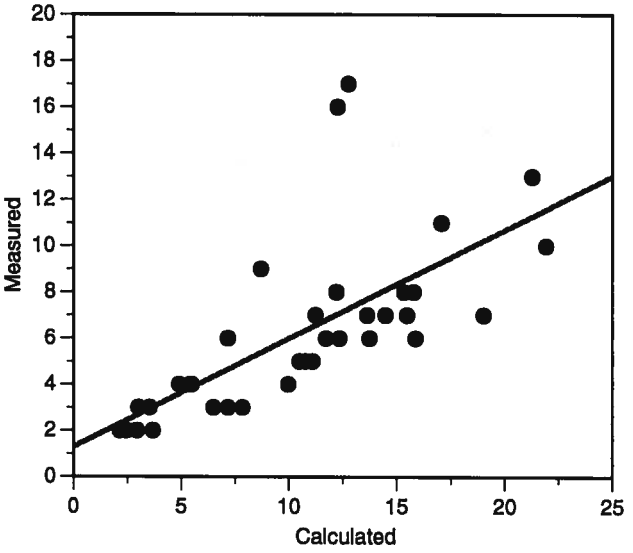


f(x) = 4.794E-1*x + 3.475E+0
R^2 = 3.914E-1

Appendix A 24 Measured and calculated concentrations of SO_2 at 6 latitude and 52.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	1.35	3.52	3
10	2.10	5.48	4
20	1.88	4.90	4
30	2.07	5.40	4
40	2.75	7.17	6
50	4.30	11.24	7
60	3.34	8.71	9
70	4.49	11.73	6
80	4.67	12.21	8
90	4.70	12.27	16
100	4.89	12.77	17
110	8.14	21.27	13
120	6.54	17.07	11
130	8.39	21.92	10
140	6.04	15.79	8
150	5.88	15.35	8
160	5.93	15.49	7
170	7.28	19.02	7
180	6.08	15.88	6
190	5.27	13.75	6
200	5.55	14.49	7
210	5.22	13.64	7
220	4.72	12.34	6
230	4.25	11.10	5
240	4.13	10.78	5
250	4.02	10.50	5
260	3.82	9.98	4
270	3.01	7.85	3
280	2.74	7.17	3
290	2.49	6.50	3
300	1.13	2.94	2
310	0.94	2.44	2
320	1.41	3.67	2
330	0.82	2.15	2
340	0.95	2.47	2
350	1.15	3.01	3
		10.3	6.1
Linear regression		5.5	3.7
	Intercept	**1.285**	
	Slope	**0.470**	
	Correlation	**0.698**	
	Force 0	**0.567277**	

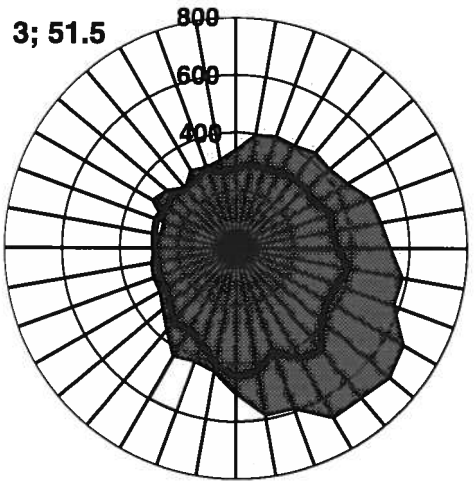


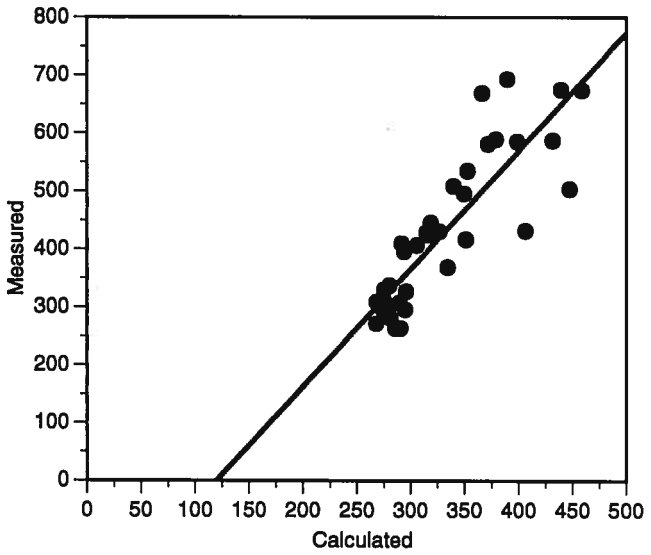


f(x) = 4.697E-1*x + 1.286E+0
R^2 = 4.864E-1

Appendix A 25 Measured and calculated concentrations of CO at 3 latitude and 51.5 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	245.14	280.16	337
10	257.23	293.97	396
20	255.19	291.64	410
30	267.51	305.73	407
40	275.42	314.76	429
50	276.21	315.66	425
60	279.00	318.85	446
70	305.97	349.68	496
80	297.43	339.92	509
90	308.62	352.71	535
100	348.89	398.73	585
110	325.17	371.62	581
120	320.40	366.17	669
130	340.79	389.47	693
140	401.31	458.63	674
150	384.38	439.29	675
160	331.50	378.86	589
170	377.62	431.56	587
180	391.41	447.32	504
190	355.57	406.36	432
200	307.36	351.27	417
210	286.05	326.91	431
220	292.70	334.52	369
230	258.72	295.68	327
240	244.27	279.17	302
250	245.15	280.17	301
260	239.98	274.26	296
270	246.30	281.48	281
280	249.94	285.64	264
290	254.31	290.64	264
300	241.05	275.49	330
310	253.60	289.82	308
320	234.46	267.95	272
330	241.52	276.02	309
340	257.99	294.84	296
350	234.73	268.26	309
		331.20	429.3
Linear regression		56.45	133.2
	Intercept	**-244.420**	
	Slope	**2.034**	
	Correlation	**0.862414**	
	Force 0	**1.316479**	

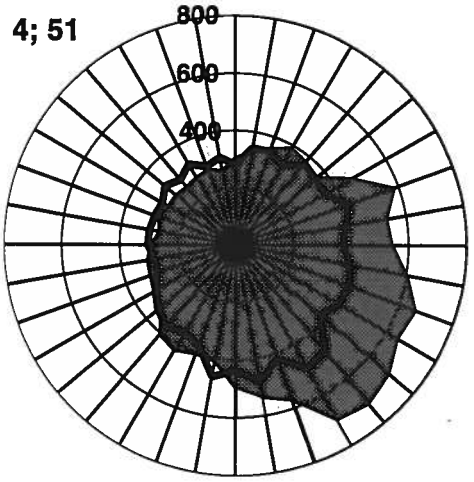


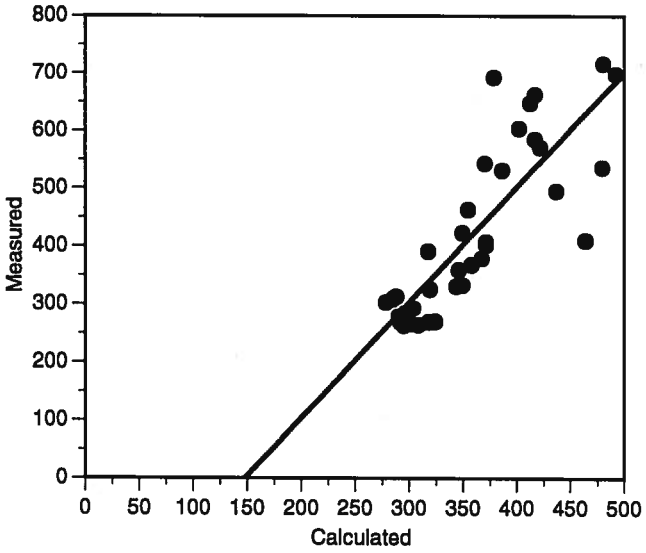


f(x) = 2.034E+0*x + -2.444E+2
R^2 = 7.438E-1

Appendix A 26 Measured and calculated concentrations of CO at 4 latitude and 51 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	243.63	278.43	303
10	300.62	343.57	331
20	302.61	345.84	359
30	277.88	317.57	391
40	321.31	367.22	379
50	313.12	357.85	368
60	310.21	354.53	463
70	364.51	416.58	585
80	337.99	386.27	531
90	323.87	370.14	543
100	351.70	401.95	604
110	364.53	416.60	663
120	360.77	412.31	648
130	331.20	378.52	693
140	420.30	480.34	717
150	430.54	492.04	699
160	368.87	421.57	571
170	419.49	479.42	536
180	382.06	436.64	495
190	405.94	463.93	410
200	324.87	371.28	402
210	305.59	349.24	423
220	324.76	371.15	407
230	305.63	349.29	333
240	279.23	319.12	326
250	251.98	287.98	313
260	249.33	284.95	309
270	265.61	303.56	293
280	263.91	301.61	266
290	259.41	296.47	285
300	255.38	291.87	269
310	278.02	317.74	269
320	258.29	295.19	263
330	283.46	323.96	270
340	254.01	290.30	279
350	269.78	308.32	264
		360.65	423.9
Linear regression		61.12	146.4
	Intercept	**-294.138**	
	Slope	**1.991**	
	Correlation	**0.8314035**	
	Force 0	**1.1975066**	

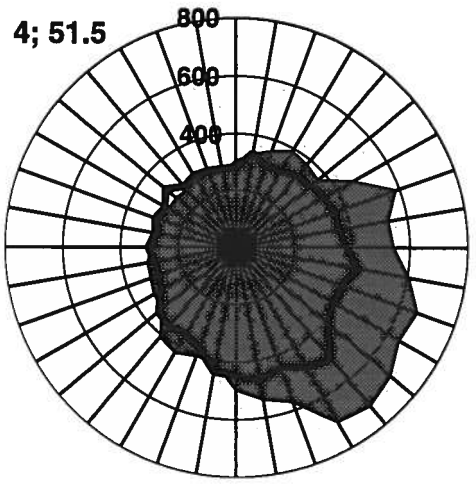


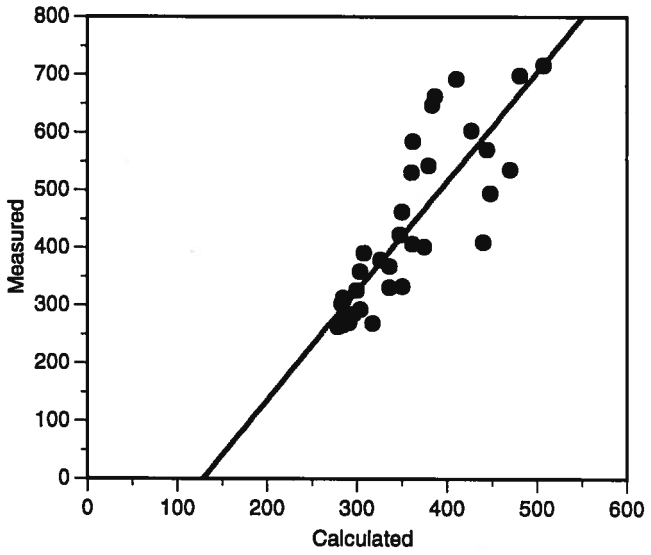


f(x) = 1.991E+0*x + -2.942E+2
R^2 = 6.912E-1

Appendix A 27 *Measured and calculated concentrations of CO at 4 latitude and 51.5 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	247.79	283.19	303
10	294.34	336.39	331
20	265.68	303.64	359
30	269.45	307.94	391
40	285.60	326.40	379
50	294.31	336.35	368
60	306.57	350.37	463
70	317.06	362.36	585
80	315.80	360.91	531
90	332.44	379.93	543
100	373.79	427.18	604
110	338.48	386.83	663
120	335.87	383.86	648
130	359.25	410.57	693
140	444.08	507.52	717
150	421.19	481.36	699
160	388.83	444.37	571
170	411.66	470.47	536
180	392.19	448.21	495
190	385.20	440.23	410
200	327.95	374.80	402
210	304.38	347.87	423
220	316.51	361.72	407
230	306.62	350.43	333
240	262.26	299.73	326
250	249.23	284.84	313
260	249.90	285.60	309
270	265.72	303.68	293
280	249.02	284.59	266
290	259.49	296.56	285
300	247.75	283.14	269
310	277.68	317.35	269
320	243.86	278.70	263
330	255.13	291.57	270
340	251.35	287.26	279
350	245.59	280.67	264
		352.13	423.9
Linear regression		65.35	146.4
	Intercept	**-242.781**	
	Slope	**1.893**	
	Correlation	**0.8453056**	
	Force 0	**1.2261281**	

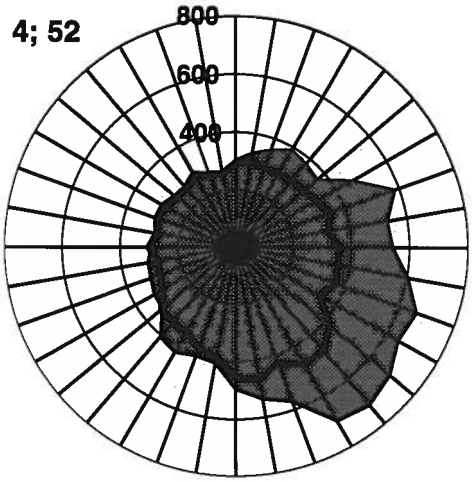


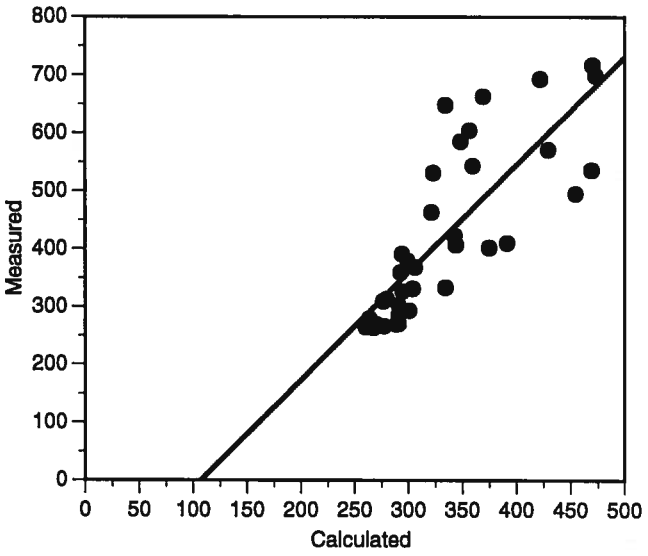


f(x) = 1.893E+0*x + -2.428E+2
R^2 = 7.145E-1

Appendix A 28 Measured and calculated concentrations of CO at 4 latitude and 52 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	254.37	290.71	303
10	265.99	303.99	331
20	256.05	292.63	359
30	257.22	293.96	391
40	261.71	299.10	379
50	267.67	305.91	368
60	281.15	321.32	463
70	304.63	348.15	585
80	282.55	322.92	531
90	314.48	359.40	543
100	311.86	356.41	604
110	323.00	369.14	663
120	292.44	334.22	648
130	369.26	422.02	693
140	411.43	470.21	717
150	413.89	473.01	699
160	375.49	429.13	571
170	410.69	469.36	536
180	397.91	454.75	495
190	342.30	391.20	410
200	327.93	374.78	402
210	299.61	342.41	423
220	300.92	343.91	407
230	292.57	334.37	333
240	257.62	294.42	326
250	244.98	279.97	313
260	241.91	276.47	309
270	263.08	300.67	293
280	242.74	277.41	266
290	254.46	290.81	285
300	252.43	288.49	269
310	236.76	270.58	269
320	233.97	267.40	263
330	254.67	291.05	270
340	230.82	263.80	279
350	227.84	260.38	264
		335.12	423.9
Linear regression		63.31	146.4
	Intercept	**-199.315**	
	Slope	**1.860**	
	Correlation	**0.804453**	
	Force 0	**1.284818**	

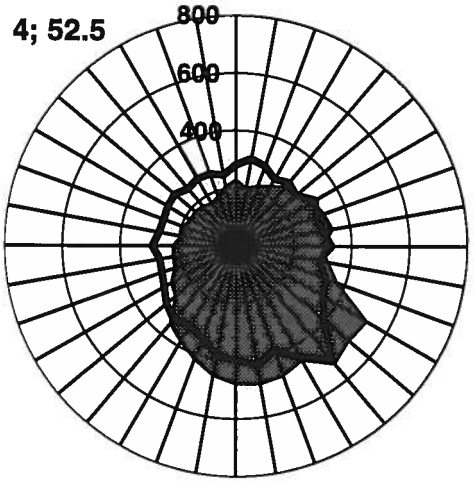


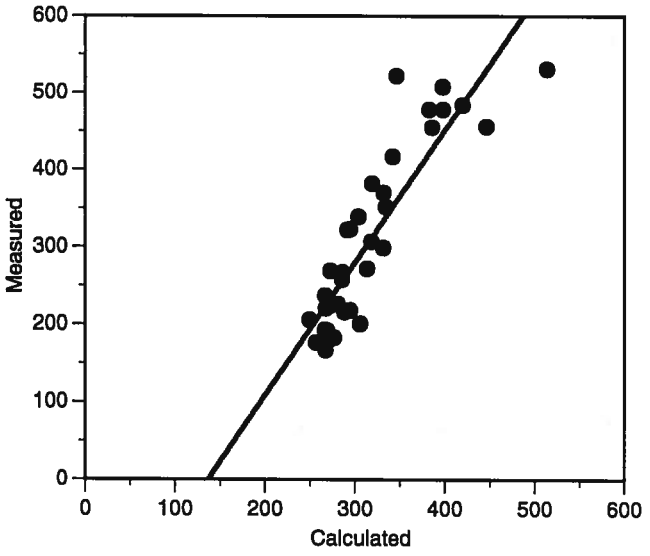


f(x) = 1.860E+0*x + -1.993E+2
R^2 = 6.472E-1

Appendix A 29 *Measured and calculated concentrations of CO at 4 latitude and 52.5 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	245.35	280.40	226
10	267.11	305.27	201
20	257.34	294.10	218
30	233.30	266.62	237
40	249.88	285.58	267
50	238.41	272.47	269
60	278.25	318.00	307
70	257.37	294.13	323
80	265.53	303.46	339
90	289.68	331.06	299
100	254.88	291.29	322
110	289.98	331.40	370
120	302.75	346.00	522
130	347.59	397.25	508
140	449.16	513.33	531
150	390.12	445.86	456
160	334.71	382.53	478
170	366.98	419.41	484
180	347.93	397.64	478
190	337.44	385.65	455
200	299.05	341.78	417
210	278.79	318.62	382
220	292.26	334.01	352
230	273.98	313.12	272
240	249.70	285.37	258
250	237.49	271.41	224
260	233.93	267.34	221
270	251.95	287.94	216
280	235.08	268.66	192
290	232.76	266.01	193
300	237.98	271.98	181
310	241.76	276.30	183
320	224.28	256.32	177
330	233.55	266.92	167
340	231.80	264.92	180
350	218.08	249.23	206
		316.70	308.6
Linear regression		60.27	116.6
	Intercept	**-233.488**	
	Slope	**1.712**	
	Correlation	**0.884575**	
	Force 0	**0.999605**	

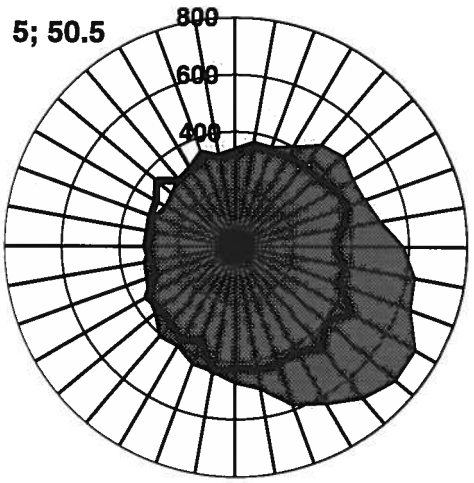


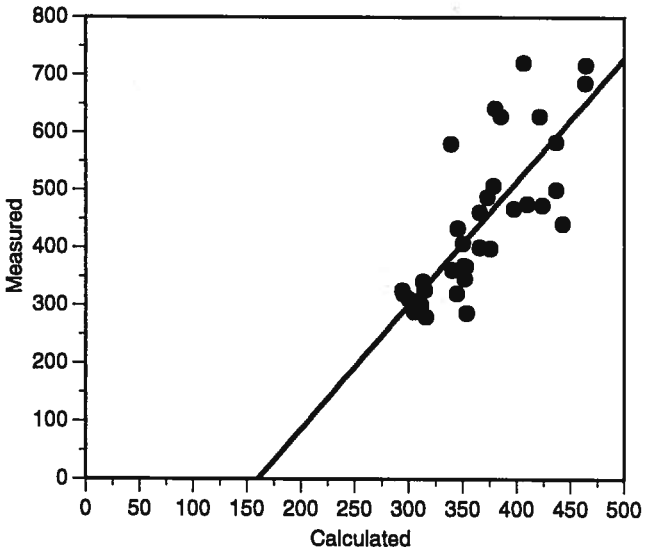


f(x) = 1.712E+0*x + -2.335E+2
R^2 = 7.825E-1

Appendix A 30 Measured and calculated concentrations of CO at 5 latitude and 50.5 longitude.

	ppb Calculated	ug/m^3 Calculated	Measured
0	274.19	313.36	341
10	307.21	351.09	368
20	308.54	352.61	367
30	319.80	365.49	400
40	319.95	365.66	461
50	326.12	372.71	487
60	347.57	397.22	467
70	358.39	409.59	475
80	331.10	378.40	507
90	296.77	339.16	580
100	337.04	385.18	628
110	332.35	379.83	642
120	406.08	464.09	717
130	355.59	406.39	721
140	405.53	463.47	686
150	368.53	421.18	628
160	381.86	436.41	583
170	381.89	436.44	500
180	370.73	423.69	473
190	387.27	442.59	441
200	302.10	345.25	433
210	328.37	375.28	398
220	306.35	350.11	407
230	297.87	340.42	361
240	307.75	351.71	346
250	257.46	294.24	325
260	258.47	295.39	319
270	270.82	309.51	297
280	266.84	304.96	288
290	272.08	310.95	301
300	276.47	315.96	279
310	309.30	353.49	286
320	262.73	300.27	310
330	272.46	311.38	300
340	301.47	344.53	320
350	275.56	314.92	326
		364.53	438.0
Linear regression		49.31	133.7
	Intercept	**-343.693**	
	Slope	**2.144**	
	Correlation	**0.790945**	
	Force 0	**1.218041**	

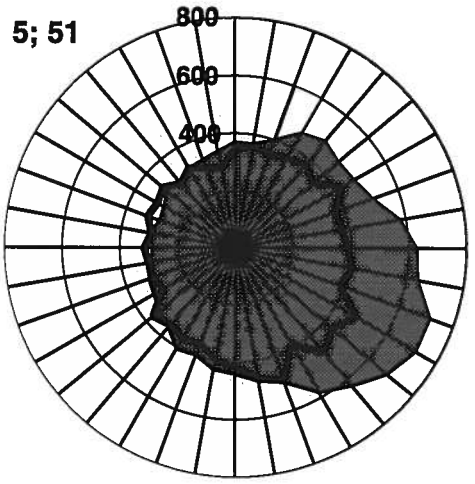


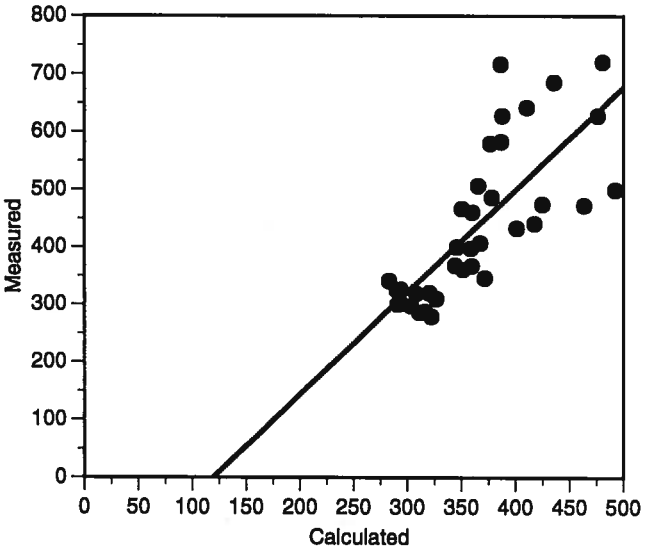


f(x) = 2.144E+0*x + -3.437E+2
R^2 = 6.256E-1

Appendix A 31 *Measured and calculated concentrations of CO at 5 latitude and 51 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	301.26	344.30	368
10	314.59	359.54	367
20	302.81	346.06	400
30	315.11	360.13	461
40	330.90	378.17	487
50	306.78	350.61	467
60	371.82	424.94	475
70	319.93	365.63	507
80	329.65	376.75	580
90	339.35	387.83	628
100	359.21	410.52	642
110	338.07	386.36	717
120	420.75	480.86	721
130	381.23	435.69	686
140	416.81	476.36	628
150	338.49	386.84	583
160	431.15	492.75	500
170	405.68	463.63	473
180	365.47	417.68	441
190	351.09	401.24	433
200	313.89	358.73	398
210	321.39	367.30	407
220	307.71	351.67	361
230	325.14	371.59	346
240	253.80	290.06	325
250	269.71	308.24	319
260	265.58	303.52	297
270	276.81	316.35	288
280	257.87	294.71	301
290	281.82	322.08	279
300	272.14	311.02	286
310	285.80	326.63	310
320	254.37	290.71	300
330	280.28	320.32	320
340	257.15	293.89	326
350	247.84	283.24	341
		365.44	438.0
Linear regression		57.35	133.7
	Intercept	**-212.717**	
	Slope	**1.781**	
	Correlation	**0.7638109**	
	Force 0	**1.2121572**	

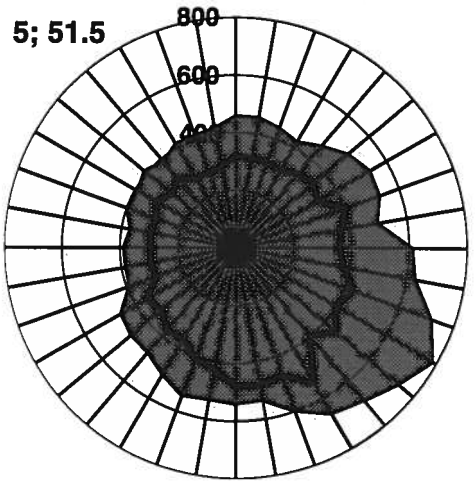


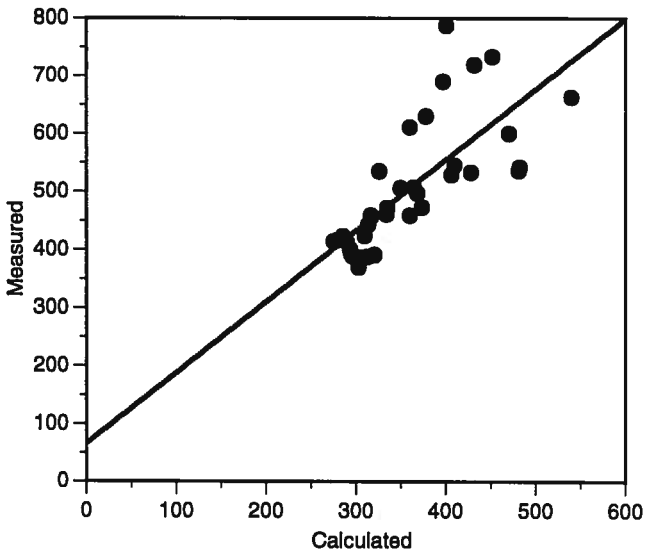


f(x) = 1.781E+0*x + -2.127E+2
R^2 = 5.834E-1

Appendix A 32 Measured and calculated concentrations of CO at 5 latitude and 51.5 longitude.

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	277.19	316.79	457
10	277.04	316.62	459
20	274.32	313.51	443
30	270.73	309.41	424
40	292.10	333.82	461
50	305.93	349.64	506
60	285.15	325.88	535
70	355.50	406.28	529
80	322.41	368.47	497
90	314.96	359.96	611
100	330.59	377.82	630
110	377.76	431.72	719
120	350.43	400.49	787
130	395.20	451.65	733
140	347.25	396.86	690
150	472.34	539.82	663
160	411.39	470.16	600
170	420.83	480.95	536
180	421.78	482.04	542
190	374.49	427.98	533
200	358.40	409.60	545
210	318.92	364.48	507
220	326.72	373.39	473
230	292.94	334.78	472
240	315.07	360.08	459
250	257.73	294.54	393
260	264.60	302.40	387
270	259.28	296.32	389
280	264.96	302.81	370
290	256.50	293.14	401
300	280.24	320.28	391
310	253.49	289.70	413
320	273.65	312.75	388
330	243.37	278.14	416
340	240.55	274.91	414
350	249.63	285.29	423
		359.79	505.4
Linear regression		67.42	110.4
	Intercept	**66.168**	
	Slope	**1.221**	
	Correlation	**0.7455142**	
	Force 0	**1.3987564**	

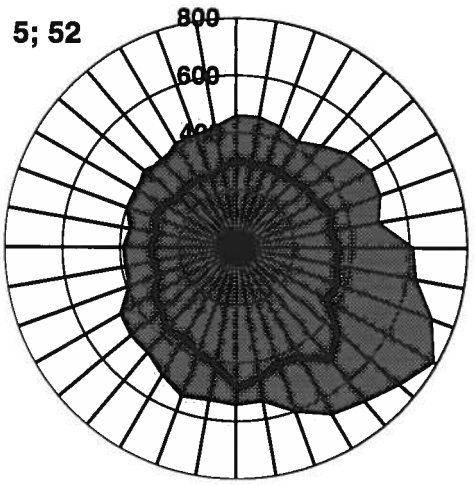


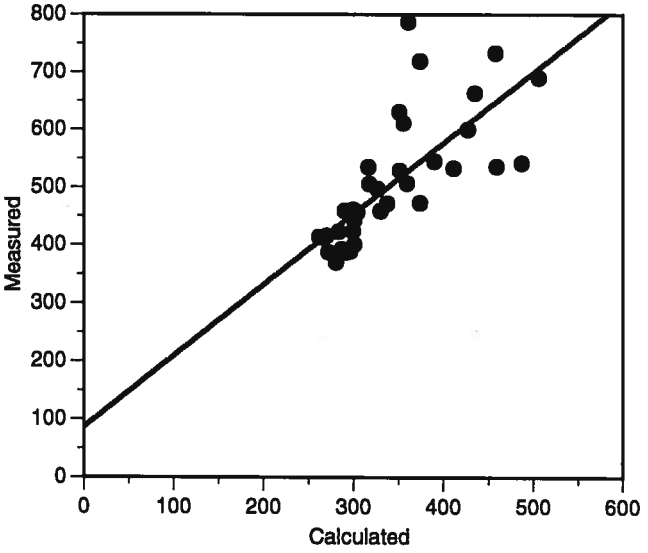


f(x) = 1.221E+0*x + 6.617E+1
R^2 = 5.558E-1

Appendix A 33 *Measured and calculated concentrations of CO at 5 latitude and 52 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	266.12	304.14	457
10	254.03	290.32	459
20	263.11	300.70	443
30	261.99	299.42	424
40	262.26	299.73	461
50	277.79	317.48	506
60	276.95	316.52	535
70	307.45	351.37	529
80	286.05	326.91	497
90	311.06	355.50	611
100	307.06	350.93	630
110	327.28	374.03	719
120	315.81	360.93	787
130	400.54	457.76	733
140	442.74	505.99	690
150	380.39	434.73	663
160	373.72	427.11	600
170	401.61	458.98	536
180	426.01	486.86	542
190	359.91	411.32	533
200	341.33	390.10	545
210	314.43	359.35	507
220	327.34	374.10	473
230	295.22	337.39	472
240	288.80	330.06	459
250	251.02	286.87	393
260	254.30	290.63	387
270	259.01	296.01	389
280	245.38	280.43	370
290	263.14	300.73	401
300	252.52	288.59	391
310	232.97	266.25	413
320	238.08	272.09	388
330	236.26	270.01	416
340	229.22	261.96	414
350	248.16	283.61	423
		342.19	505.4
Linear regression		66.29	110.4
	Intercept	**86.553**	
	Slope	**1.224**	
	Correlation	**0.734965**	
	Force 0	**1.468174**	

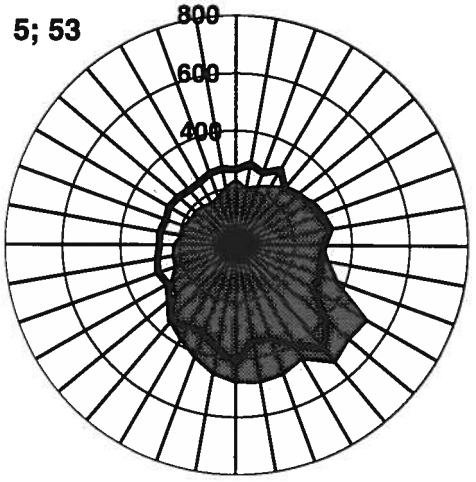


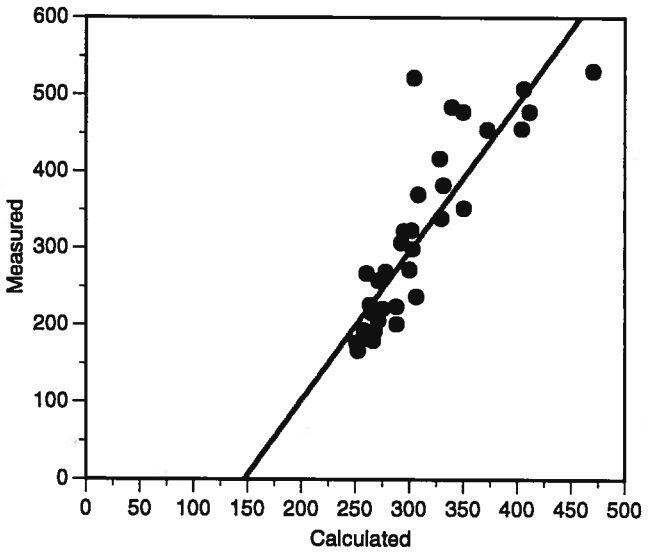


f(x) = 1.224E+0*x + 8.655E+1
R^2 = 5.402E-1

Appendix A 34 Measured and calculated concentrations of CO at 5 latitude and 53 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	230.7	263.65	226
10	252.26	288.30	201
20	235.30	268.91	218
30	268.25	306.57	237
40	227.99	260.56	267
50	243.43	278.21	269
60	256.19	292.79	307
70	264.34	302.10	323
80	288.36	329.55	339
90	265.26	303.16	299
100	258.36	295.27	322
110	269.89	308.44	370
120	266.57	304.65	522
130	355.45	406.23	508
140	411.72	470.54	531
150	353.84	404.39	456
160	306.19	349.93	478
170	297.08	339.52	484
180	360.07	411.51	478
190	325.80	372.35	455
200	287.41	328.47	417
210	290.16	331.61	382
220	306.64	350.44	352
230	262.65	300.17	272
240	237.60	271.55	258
250	252.02	288.02	224
260	240.82	275.22	221
270	231.99	265.13	216
280	234.50	268.00	192
290	225.90	258.18	193
300	233.17	266.48	181
310	227.25	259.71	183
320	219.70	251.08	177
330	220.98	252.55	167
340	233.25	266.57	180
350	237.48	271.41	206
		307.26	308.6
Linear regression		52.34	116.6
	Intercept	**-282.787**	
	Slope	**1.925**	
	Correlation	**0.863849**	
	Force 0	**1.029751**	

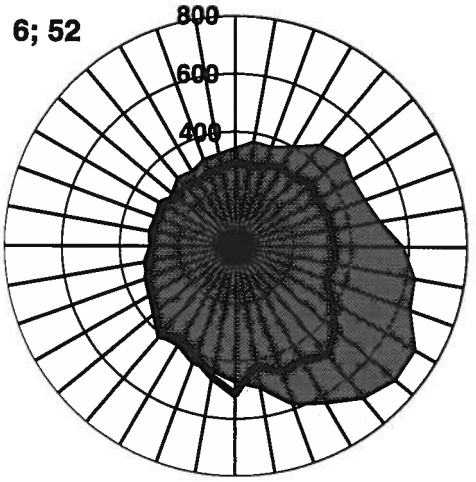


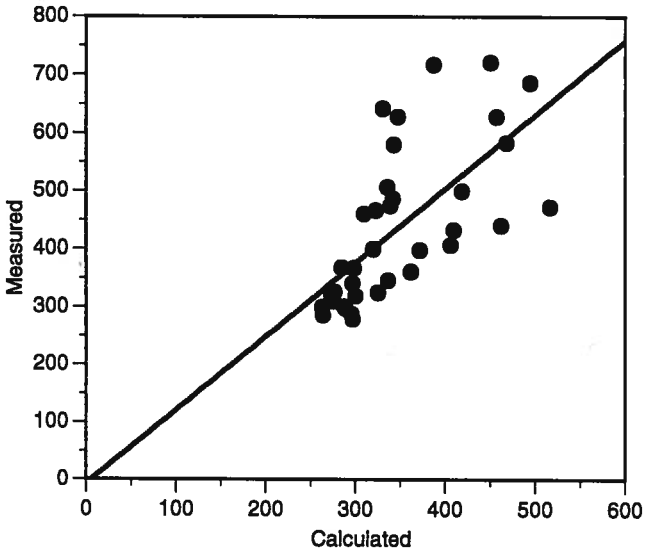


f(x) = 1.925E+0*x + -2.828E+2
R^2 = 7.462E-1

Appendix A 35 *Measured and calculated concentrations of CO at 6 latitude and 52 longitude.*

	ppb	ug/m^3	
	Calculated	**Calculated**	**Measured**
0	259.47	296.53	341
10	248.92	284.48	368
20	260.75	298.00	367
30	279.84	319.81	400
40	270.78	309.47	461
50	298.97	341.68	487
60	282.44	322.79	467
70	296.37	338.71	475
80	293.61	335.55	507
90	299.87	342.71	580
100	303.84	347.25	628
110	289.26	330.58	642
120	338.89	387.31	717
130	394.07	450.36	721
140	432.48	494.26	686
150	399.82	456.94	628
160	409.40	467.89	583
170	366.24	418.56	500
180	451.82	516.36	473
190	404.24	461.99	441
200	358.27	409.46	433
210	325.32	371.79	398
220	355.38	406.15	407
230	316.62	361.86	361
240	294.25	336.28	346
250	284.48	325.12	325
260	262.41	299.90	319
270	252.90	289.03	297
280	258.50	295.43	288
290	251.28	287.18	301
300	259.65	296.74	279
310	230.66	263.62	286
320	240.75	275.14	310
330	229.74	262.56	300
340	238.78	272.90	320
350	241.98	276.55	326
		348.64	438.0
Linear regression		70.22	133.7
	Intercept	**-6.636**	
	Slope	**1.275**	
	Correlation	**0.66992**	
	Force 0	**1.257043**	

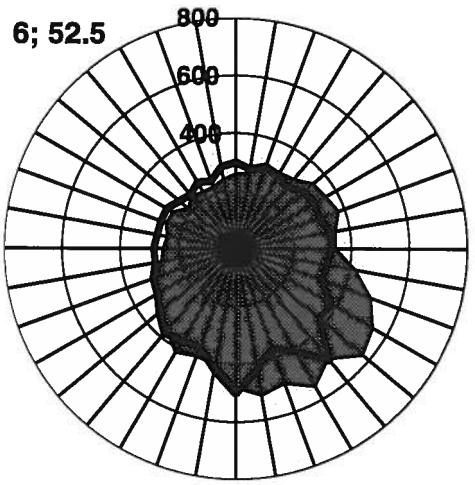


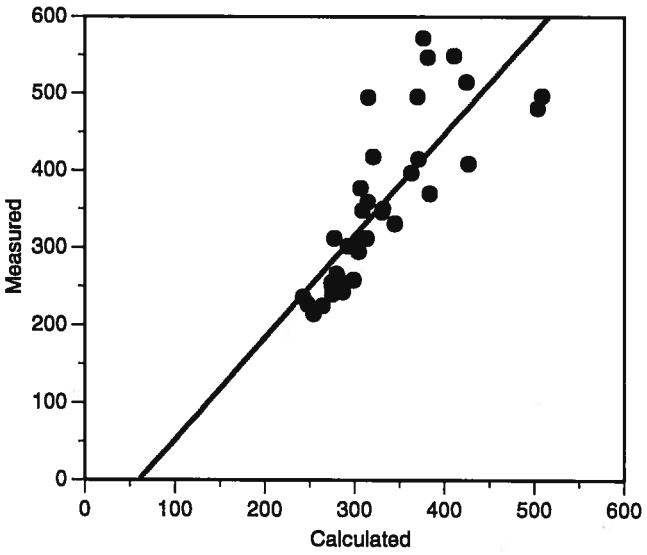


f(x) = 1.275E+0*x + -6.636E+0
R^2 = 4.488E-1

Appendix A 36 Measured and calculated concentrations of CO at 6 latitude and 52.5 longitude.

	ppb	ug/m^3	
	Calculated	Calculated	Measured
0	261.34	298.67	258
10	244.89	279.88	266
20	265.86	303.84	309
30	256.11	292.70	302
40	242.78	277.46	312
50	270.01	308.59	348
60	275.12	314.42	359
70	268.23	306.55	377
80	290.10	331.54	350
90	289.12	330.43	346
100	280.58	320.66	418
110	275.79	315.19	495
120	333.98	381.69	547
130	329.48	376.55	572
140	445.00	508.57	497
150	359.60	410.98	549
160	323.79	370.05	496
170	371.50	424.57	515
180	440.78	503.75	481
190	373.38	426.72	409
200	317.79	363.19	397
210	324.79	371.18	415
220	335.76	383.73	370
230	301.62	344.71	331
240	273.98	313.12	312
250	266.36	304.41	295
260	246.37	281.56	261
270	248.14	283.59	250
280	240.55	274.91	240
290	240.09	274.39	255
300	240.42	274.76	250
310	222.57	254.37	215
320	212.40	242.75	236
330	231.22	264.25	225
340	216.75	247.71	227
350	251.01	286.86	243
		329.12	353.6
Linear regression		65.48	106.9
	Intercept	**-79.354**	
	Slope	**1.315**	
	Correlation	**0.806019**	
	Force 0	**1.083181**	





f(x) = 1.315E+0*x + -7.936E+1
R^2 = 6.497E-1