DEDICATION

This thesis is dedicated to my brother Kalamazad, who died in a tragic accident on 11th February, 1976. Kalamazad, 4 years my senior, was my mathematics teacher for 2 years and his inspiration has certainly made me a better mathematics student which is reflected largely in this thesis.

Kalamazad was not only a great teacher, but he also earned the distinction of being one of the top engineers in Guyana. Had he not such an untimely death, he would have made, I am sure, an outstanding contribution to engineering.

It seems fitting therefore to dedicate this thesis as a small tribute to my brother whose tragic loss will be felt for years to come.

SPECTRAL ANALYSIS
OF
ATMOSPHERIC POLLUTANTS
IN THE
UPPER GREAT LAKES REGION

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ATMOSPHERIC POLLUTANTS
IN THE
UPPER GREAT LAKES REGION

By

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ABSTRACT

Chemical data from atmospheric precipitation in the Upper Great Lakes region were collected on a monthly basis by various organizations from late 1972 to late 1975. The data for pairs of chemical parameters are regarded as simultaneously recorded time series and are analyzed by spectral analysis to bring out periodic components present in both time series and also to reflect the correlation and phase difference between the pairs of time series.

The precipitation data have four major bands of periodicities: (1) a yearly cycle representing high and low atmospheric loadings in the summer and winter respectively with regards to Fe, PART, SO_4 and P; (2) a 6 monthly period reflecting the four seasons of the year and affecting Fe, PART, P and pH; (3) a 9 monthly period for the heavy metals (Cu, Ni, Cd, Pb and Zn); and (4) 2 to 5 cycles/month periodicities caused by atmospheric turbulence and affecting all the parameters.

The mining-smelting industry at Sudbury has been identified as a major point source of atmospheric emissions of acid, sulfate and the heavy metals, with the exception of iron. This is evident in view of the facts that the spectral intensities at the major periodicities decrease away from the mining-smelting centre; the strikingly high correlation and practically inphase relationship between Cu and Ni, and the 9 monthly period for the heavy metals which is associated with the industry.

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1. INTRODUCTION

1.1 Statement of Problem

An international network of stations is set up in the Upper Great Lakes region to collect chemical data from atmospheric precipitation. Samples of precipitation are collected on a monthly interval and the concentration of various chemical parameters are determined. The large amount of data ranging from late 1973 to late 1975 is particularly suited for computer manipulations and is therefore analyzed by spectral analysis.

The aim of the present study is to analyze the data (time series) for any periodic components. Furthermore, to see if any loading parameters are correlated and what are their possible sources of origin. If a model is suggested as a result of this analysis, then it can be used to make predictions. That is, the estimation of future values $x(t + T)$ of the time series in some future range $0 \leqslant T \leqslant T'$ from some values of the series up to and including time t.

1.2 Outline of Analysis

The obvious method of analysis of periodic data is the classical Fourier analysis. The precise use of the technique in this study is to approximate non-periodic functions by sine and cosine functions. The approximation becomes better as the number of points increases and becomes exact if the number of points is infinite.

To analyze for cross correlation between two series involves statistical analysis. Thus statistical questions, to be discussed later, do arise from the fact that it is necessary to estimate the accuracy of the various functions obtained from finite amounts of data.

Consequently two very important approaches are considered, the statistical analysis of time series and the method of Fourier analysis. These two approaches constitute spectral analysis.

The main advantage of spectral analysis is that large amounts of data are digested into a few graphical spectral presentations, the main features of which can be readily interpreted and which disclose persistent periodicities in the data, if present.

The power spectrum shows the frequency distribution of the energy (variance) of a time series over a defined frequency range. Peaks in particular frequency bands of the spectrum indicate higher contents of energy in those bands than in neighbouring ones. The positions of the peaks give the periodicities.

To disclose relationships between loading parameters, they must be analyzed in pairs. The pairs are regarded as simultaneously recorded time series. Cross spectral analysis of such series contains two different types of information. The first is the squared coherency spectrum which effectively measures the correlation between the two series at each frequency, and the second is the phase spectrum which measures the phase difference between the two series at each frequency.

Energy distribution with frequency is not a continuous distribution, but is a "histogram" made up of estimates of successive small sub-divisions of the frequency range. The number of such sub-divisions is called the number of "lags". It determines the statistical reliability of the results obtained and the frequency resolution. The resolution increases as the number of lags L increases. However, the number of degrees of freedom ($\sim \frac{2N}{L}$ where N is the number of points in the time series) and hence the statistical reliability, decreases as L increases (i.e. the confidence limits widen as L increases).

When large amounts of data are analyzed, for example 10,000 points, it is not possible to look at the data and the spectra consistently and the number of lags becomes crucial. However, with an average of 40 points displayed graphically together with the spectra, interpretation can be consistent and less emphasis can be placed on the number of lags. Thus in the present work the number of lags is set equal to N/2 to give an adequate amount of resolution to disclose gross features.

The data used in this study were collected on a monthly interval and thus the maximum frequency f_n is 0.5 cycles/month. If there is appreciable energy in the time series corresponding to frequencies greater than f_n, then this energy can generate spurious additions and peaks in the range 0 to f_n, which in severe cases can completely mask the true spectrum. For the present data, a period of less than two months is unlikely. In any event, it would be very costly to sample, say, every $\frac{1}{4}$ month to see a $\frac{1}{2}$ monthly period, if present.

Fourier analysis requires a continuous series. In most cases the data are complete but in the few cases where data are missing for less than 3 months then such gaps are filled by the value of the month immediately preceeding the gap.

Spectral analysis is applicable to stationary time series. Hence any trends present in the data must be removed. Atmospheric loading is presumably due to industries and therefore broad trends are expected which reflect the gradual growth of the economy. Such trends are rather difficult to remove if the data are not well understood. At the present time, the best one can do is to assume a linear trend and remove it by a least squares fit to all the data and subtraction to obtain the residuals which are used in the analysis. Negative residuals do not have physical meaning for the data and are set equal to zero.

2. METHODS

2.1 Mathematical Development

Spectral analysis brings together two very important theoretical approaches, the statistical analysis of time series and the methods of Fourier analysis. The role of Fourier analysis in applied mathematics and engineering is well documented. The analytic techniques are particularly important in three applications: (a) for studying periodic solutions to physical problems described by differential equations; (b) as an operational device for solving differential equations; (c) for approximating non-periodic functions.

The present study is concerned primarily with the latter case. Before going into the analysis in some details, it is necessary to define a few basic terms.

A function is said to be periodic of period T, if for all t,

$$f(t) = f(t + T). \tag{2.1.1}$$

The function between time t and t + T can be of any shape whatsoever. A particularly simple shape is the cosine function,

$$f(t) = A \cos 2\pi f_1 t, \quad -\infty < t < \infty, \tag{2.1.2}$$

which has a period $T = \frac{1}{f_1}$, since

$$A \cos 2\pi f_1 t = A \cos 2\pi f_1 \left[t+\left(\frac{1}{f_1}\right)\right]. \tag{2.1.3}$$

Furthermore, a function f(t) is said to be a (real) harmonic with frequency w and amplitude A, where A and w are positive constants, if it is of the form

$$f(t) = A \cos wt$$

or

$$f(t) = A \sin wt. \tag{2.1.4}$$

A harmonic with frequency, w, has period

$$T = \frac{2\pi}{w}, \tag{2.1.5}$$

and since it is a harmonic, it must satisfy

$$T = k\frac{2\pi}{w} \text{ for some integer } k=1,2,\ldots, \tag{2.1.6}$$

since

$$\cos\,(w(t+\theta)) = \cos wt \Rightarrow \theta=k\frac{2\pi}{w}. \tag{2.1.7}$$

The frequency, w, represents the number of complete cycles in 2π units of time and is therefore called the angular frequency, to distinguish it from the true frequency, f, measured in cycles per unit time. The true frequency f of a harmonic with angular frequency w is given by

$$f = \frac{w}{2\pi} = \frac{1}{T}. \tag{2.1.8}$$

For completeness, we may define zero frequency by a constant function

$$f(t) = A, \tag{2.1.9}$$

which may be regarded as the value of the harmonic function A cos wt with $w = 0$.

2.2 Fourier Analysis

It is possible to represent any non-periodic functions using any class of periodic functions. In Fourier analysis, the periodic functions used are sine and cosine functions. They have the important properties that an approximation consisting of a given number of terms achieves the minimum mean square error between the signal and the approximation, and also they are orthogonal, so the coefficients may be determined independently of one another.

Consider a signal which is specified only at discrete times, and suppose it is required to be expanded in terms of periodic functions. The discrete signal can be regarded as having been derived from a continuous signal $S(t)$ of duration T by sampling the values of the signal at spacing Δ, as shown in Figure 2.2.1. This produces $N = T/\Delta$ sample values S_r, where

$$S_r = S(t=r\Delta). \qquad (2.2.1)$$

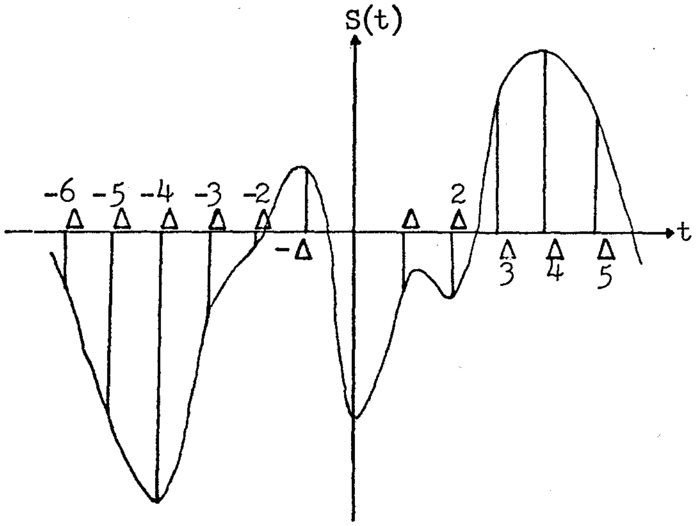


Figure 2.2.1: A discrete signal obtained by sampling a continuous signal.

For convenience it is assumed that N is even and equal to $2n$ so that r may run through the integers $-n,\ldots,0,1,\ldots,n-1$.

Note that periodic functions which pass through the sample values may be chosen in an infinite number of ways. For example, the finite Fourier series

$$\bar{S}(t) = A_o + 2 \sum_{m=1}^{n-1} \left\{ A_m \cos 2\pi m f_1 t + B_m \sin 2\pi m f_1 t \right\} + A_n \cos 2\pi n f_1 t \tag{2.2.2}$$

contains N constants, the A_m and B_m, which can be determined so that the discrete and continuous values coincide at the points $t = r\Delta$, that is, $\bar{S}(t) = S_r$. Thus the function $\bar{S}(t)$ provides an <u>approximation</u> to the original continuous function $S(t)$ in the interval $-T/2 \leqslant t < T/2$.

On substituting $t = r\Delta$ in (2.2.2) and setting $\bar{S}(r\Delta) = S_r$, a set of N equations for the N unknown constants is obtained. The equations are

$$S_r = A_o + 2 \sum_{m=1}^{n-1} \left\{ A_m \cos 2\pi m f_1 r\Delta + B_m \sin 2\pi m f_1 r\Delta \right\} + A_n \cos 2\pi n f_1 r\Delta,$$

$$(r = -n, \ldots, 0, 1, \ldots, n-1). \tag{2.2.3}$$

Choosing $f_1 = 1/N\Delta$ simplifies the solution of equation (2.2.3), because then the sines and cosines are orthogonal. The frequency $f_1 = 1/N\Delta$ is called the <u>fundamental</u> <u>frequency</u> of the signal $\bar{S}(t)$, and it corresponds to a period T_1 equal to the length of the record,

$$f_1 = \frac{1}{N\Delta}; \; T_1 = N\Delta. \tag{2.2.4}$$

The function $\bar{S}(t)$ is composed of a sum of sine and cosine functions whose frequencies are multiples or harmonics of the fundamental f_1. Note that $(N = 2n)$ the

$$\text{2nd harmonic:} \quad f_2 = \frac{2}{N\Delta}; \; T_2 = \frac{N\Delta}{2}; \; f_2 = 2f_1$$

and (2.2.5)

$$\text{nth harmonic:} \quad f_n = \frac{n}{N\Delta} = \frac{N/2}{N\Delta} = \frac{1}{2\Delta}, \; T_n = 2\Delta, \; f_n = nf_1$$

The highest frequency f_n present is $1/2\Delta$, which corresponds to a Period of 2 sampling intervals. The parameter f_n is called the "Nyquist frequency". If the number of points N is odd, say $2n - 1$, (2.2.2) can be solved by similar reasoning, the only difference being that the A_n term vanishes. Furthermore, it can be shown that A_o is the mean or average value of S_r if N is either even or odd.

To solve (2.1.3) for the constants, the A_m and B_m, the discrete Fourier transform of the series of N terms would require approximately N^2 operations. This is very expensive to operate when large number of data points are involved. However, a recent innovation in spectral analysis is the fast Fourier transform (FFT).

2.3 Fast Fourier Transform

The FFT is an algorithm for computing discrete Fourier transforms much more quickly than the direct method given above, but at the same time retaining accuracy. For a series of N terms, the FFT requires only $2N\log_2 N$ operations compared to N^2 operations required for the direct method. Thus savings in computer time is very large if one is interested in the Fourier analysis of long series.

The relevance of the FFT to spectral analysis is that it is now faster to compute the power spectrum directly using an FFT, then smooth the spectrum rather than compute the autocorrelation function, smooth with a lag window and finally transform.

Description of the Fast Fourier Transform

Suppose it is required to find the Fourier transform

$X_m, m = 0,1,\ldots,N-1$, of the series $x_t, t = 1,2,\ldots,N$, where N is even. The series can be partitioned into two series y_t and z_t, where

$$y_t = x_{2t-1};$$
$$z_t = x_{2t}, \quad t = 1,2,\ldots,N/2. \tag{2.3.1}$$

The series y_t, z_t each consist of N/2 values and hence have Fourier transforms

$$Y_m^{(N/2)} = \frac{2}{N}\sum_{t=1}^{N/2} y_t\, e^{-j(4\pi tm/N)},$$

and

$$Z_m^{(N/2)} = \frac{2}{N}\sum_{t=1}^{N/2} z_t\, e^{-j(4\pi tm/N)}, \tag{2.3.2}$$

where the superscript on the transform denotes the number of terms in the series and the transform. But $X_m^{(N)}$ and $Y_m^{(N/2)}$, $Z_m^{(N/2)}$ are related. In fact,

$$X_m^{(N)} = \frac{e^{j(2\pi m/N)}}{2}\, Y_m^{(N/2)} + \frac{1}{2} Z_m^{(N/2)}$$

and

$$X_{m+(N/2)}^{(N)} = -\frac{e^{j(2\pi m/N)}}{2}\, Y_m^{(N/2)} + \frac{1}{2} Z_m^{(N/2)}, \quad 0 \leqslant m \leqslant \frac{N}{2} - 1. \tag{2.3.3}$$

Therefore, the Fourier transform for the series x_t is easily obtained from the Fourier series of the half-series y_t and z_t. Likewise, if N/2 is even, the series y_t and z_t are partitioned into two series y_t', z_t', and y_t'', z_t'' respectively, and an appropriate version of (2.3.3) is used to construct the transforms $Y_m^{(N/2)}$ and $Z_m^{(N/2)}$ from the transforms of the series of length N/4.

For series of length $N = 2^k$, the procedure is followed until partitions of only one term are obtained, for which the Fourier transform equals the term itself. In practice, N is not an exact power of 2, thus sufficient zeros must be appended to the data to make N an exact power of 2. This procedure does not alter the accuracy of the answers in anyway.

2.4 Power Spectrum

The power spectrum is the Fourier transform of the autocovariance function (acvf). The spectrum shows how the variance (energy) is distributed with frequency for a time series over a defined frequency range. Subject to statistical considerations, peaks in particular frequency bands of the power spectrum indicate higher contents of energy in those bands than in neighbouring ones.

The variance or average power of a discrete signal x(t) observed at times $t = -n\Delta, -(n-1)\Delta, \ldots, (n-1)\Delta$, can be decomposed into contributions at a finite number of harmonics of the fundamental frequency $f_1 = 1/N\Delta$ $(N = 2n)$ according to

$$s_T^2 = \frac{1}{N} \sum_{t=-n}^{n-1} x_t^2 = \sum_{m=-n}^{n-1} |X_m|^2, \qquad (2.4.1)$$

X_m is called the complex amplitude at the harmonic frequency f_m and measures the amplitudes of the sine and cosine terms at frequency f_m in x(t). X_m is given by

$$X_m = \frac{1}{N} \sum_{t=-n}^{n-1} x_t \, e^{-j2\pi m t \Delta / N\Delta} = \frac{1}{N} \sum_{t=-n}^{n-1} x_t \, e^{-j2\pi m t/N}. \qquad (2.4.2)$$

The contribution of $|X_m|^2$ to the average power of frequency f_m is called the <u>intensity</u> at this frequency.

The power spectrum $C_{xx}(f)$ is the Fourier transform of the acvf $\gamma_{xx}(k)$,

$$C_{xx}(f) = \Delta \sum_{k=-\infty}^{\infty} \gamma_{xx}(k) \, e^{-j2\pi k f \Delta}, \quad -\frac{1}{2\Delta} < f < \frac{1}{2\Delta}, \quad k = 0, \pm 1, \pm 2, \ldots \qquad (2.4.3)$$

$C_{xx}(f)$ shows how the variance of the $X(t)$ process is distributed over frequency and is non-negative for all f. The spectrum is usually plotted on a logarithmic scale which shows more detail in the spectrum over a wider amplitude range. Another reason why the log. scale is a sensible choice is that only proportional changes in power is important. Furthermore, if confidence intervals is to be represented on the plot, then on a log. scale it is simply a constant interval about the spectral estimate.

<u>2.5 Cross Spectrum</u>

Cross spectral analysis of two simultaneously recorded time series provides information on the relationships between them, in the form of estimates of coherences and phase differences, as functions of frequency.

The acvf has a Fourier transform called the <u>cross spectrum</u>. This spectrum is a complex quantity which may be written as the product of a real function called the <u>cross amplitude spectrum</u> and a complex function called the <u>phase spectrum</u>. However, a more useful quantity than the cross amplitude spectrum is the coherency square spectrum. The latter is dimensionless.

2.6 Phase and Cross Amplitude Spectra

Suppose that $x_1(t)$ and $x_2(t)$ are arbitrary real signals with Fourier transforms $X_1(f)$ and $X_2(f)$ respectively. These Fourier transforms give the amplitude and phase distribution of the signals, i.e.

$$X_i(f) = A_i(f)\, e^{jFi(f)}, \quad i = 1,2. \tag{2.6.1}$$

The cross spectrum can be written as

$$C_{12}(f) = A_{12}(f)\, e^{jF12(f)}. \tag{2.6.2}$$

The covariance between the two series $x_1(t)$ and $x_2(t)$ may be described by the phase spectrum $F_{12}(f)$,

$$F_{12}(f) = F_2(f) - F_1(f) \tag{2.6.3}$$

and the cross amplitude spectrum

$$A_{12}(f) = A_1(f)A_2(f)/T. \tag{2.6.4}$$

The phase spectrum $F_{12}(f)$ shows whether the frequency components in one series lead or lag the components at the same frequency in the other series. Similarly, the cross amplitude spectrum $A_{12}(f)$ shows whether the amplitude of the components at a particular frequency in one series is associated with a large or small amplitude at the same frequency in the other series.

2.7 Co- and Quadrature Spectra

Since (2.6.2) is a complex quantity, it can be written as a sum of a real and imaginary part, that is,

$$C_{12}(f) = L_{12}(f) - jQ_{12}(f),$$

where

$$L_{12}(f) = A_{12}(f)\cos F_{12}(f), \qquad Q_{12}(f) = -A_{12}(f)\sin F_{12}(f), \quad (2.7.1)$$

and

$$A^2_{12}(f) = L^2_{12}(f) + Q^2_{12}(f), \qquad F_{12}(f) = \arctan - \frac{Q_{12}(f)}{L_{12}(f)}.$$

$L_{12}(f)$ is called the co-spectrum and it measures the variance between the in-phase components of the two series. $Q_{12}(f)$ is called the quadrature spectrum and it measures the out of phase components of the two series.

2.8 Squared Coherency Spectrum

The squared coherency is defined as

$$K^2_{12}(f) = \frac{A^2_{12}(f)}{C_{11}(f)C_{22}(f)} \qquad (2.8.1)$$

where $C_{11}(f)$ and $C_{22}(f)$ are the power spectra of series 1 and series 2 respectively. A plot of $K^2_{12}(f)$ versus f is called the squared coherency spectrum. It resembles the ordinary correlation coefficient. In fact, the coherency plays the role of the correlation coefficient defined at each frequency f. A value near to unity indicates that the two series correlate closely, in terms of variance contribution, at a particular frequency interval, while a value near to zero indicates little correlation between the two series at that frequency interval.

The coherency spectrum is useful in practice because it provides non-dimensional measure of the correlation between the two time series as a function of frequency. Thus it should be preferred to the cross amplitude spectrum, which depends on the scale of measurement of $X_1(t)$ and $X_2(t)$.

Consequently the cross correlation properties of two time series can be described by the squared coherency spectrum and the phase spectrum. The main uses of the co- and quadrature spectrum are in their estimation of the above two spectra.

2.9 Smoothed Cross Spectral Estimators

The estimators of the sample cross spectra have the undesirable properties that their variances are dominated by a constant term which does not tend to zero as the record length increases. Hence cross spectral estimators must be smooth using a spectral (lag) window. The lag window Hanning is used to smooth the data analyzed in this present study. Hanning is defined as

$$\left.\begin{aligned} D(u) &= \tfrac{1}{2}\left(1 + \cos \frac{\pi u}{T_m}\right) && \text{for } |u| < T_m, \\ &= 0 && \text{for } |u| > T_m, \end{aligned}\right\} \quad (2.9.1)$$

where T_m is the maximum lag desired to be used. Note that $D(u)$ is an even function of u and is regarded as a window of variable transmission which modifies the values of the autocovariance function at different lags.

2.10 Description of Computer Program (Fortran)

The program is presented in Appendix A. Except for the main program OFF and subroutine PLOTT, the program was obtained from Fee (1969) and little or no alteration was made to it.

The program is simple to understand. The major steps of operation of the program are as follows:

(1) PROGRAM OFF: This is the main program which reads the data of two time series and their identification from tape. The two series represent data collected within the same period of time. The number of data points N is also noted.

(2) SUBROUTINE FOUR: DELTAT (the time interval between observations), NUMBER (the number of time series to be analysed) and LAGS (the number of spectral estimates to be made) are set equal to 1,2, and N/2 respectively. This subroutine also labels the output.

(3) SUBROUTINE INPUT: The trend in the data is removed by a least squares fit to the data and subtraction to obtain the residuals. Negative residuals are set equal to zero. Finally, zeros are appended to the end of the data to make N an exact power of 2 ($N = 2^k$) as required for the fast Fourier transform.

(4) SUBROUTINE FOURT: The time series is Fourier transformed to obtain the Fourier coefficients ($a_{1k} + jb_{1k}$, $k = o,\ldots,N$).

Steps (3) and (4) are repeated for the second time series to obtain $a_{2k} + jb_{2k}$. Since two series are analysed, SUBROUTINE SPEC1 is inactive. However, it has been retained for generalization.

(5) SUBROUTINE SPEC2: The real and imaginary Fourier coefficients are squared and summed over the number of lags L and normalized

by the number of terms squared,

$$\sum_{k=i}^{L} (a_k^2 + b_k^2) \; / \; (L - i + 1),$$

to obtain the raw spectral estimates of both series.

The raw cross spectrum is obtained by taking the sum of the cross products of the real and imaginary parts of the transformed series,

$$\sum_{k=i}^{L} (a_{1k} \text{ X } a_{2k}) + (b_{1k} \text{ X } b_{2k}) \;\; / \; (L - i + 1).$$

(6) The raw estimates are smoothed by Hanning (SUBROUTINE HANN) before estimation of the phase and coherency squared spectra.

(7) $\text{PHASE} = \arctan \dfrac{(\text{Quadrature spectrum }_k)_{\text{smoothed}}}{(\text{Co-spectrum }_k)_{\text{smoothed}}}$, where k refers to the kth lag.

(8) $\text{COHERENCE}_k = \dfrac{(\text{Co-spectrum}_k)^2 + (\text{Quadrature spectrum}_k)^2}{(\text{Spectrum}(1)_k \text{ X spectrum}(2)_k)}$

(9) SUBROUTINE PLOTT: This subroutine produces a series of plots of the data and spectra.

a) Raw data of series 1 (and trend) vs. time;

b) Series 1 (trend removed) vs. time;

c) Same as a) and b) for series 2;

d) Power spectrum for both series 1 and 2;

e) Phase spectrum;

f) Squared coherency spectrum.

2.11 Testing of Computer Program

The program is tested on simple periodic functions, namely the sine and cosine functions. The values for the functions are estimated at ninety-six equally spaced sampling intervals on the domain 0 to 8π. A linear least squares fit of all the points is also estimated.

The power spectra for the trigonometric functions are estimated for various lags, which include the lag to be used for the precipitation data of the Upper Great Lakes region. The expected results were obtained. They are discussed in Section 3.3.1.

3. RESULTS

3.1 Station Locations

Figure 3.1.1 shows the distribution of the monthly stations in the Upper Great Lakes (U.G.L.) region. Table 3.1.1 gives the latitude and longitude of the stations.

TABLE 3.1.1: Station locations

No.	Station	Latitude	Longitude
2	Skead	46.658N	80.752W
3	Killarney	45.990N	81.447W
4	Gore Bay	45.881N	82.570W
5	Jamot	46.105N	80.625W
6	Windy Lake	46.615N	81.458W
8	Mount Lake	46.680N	82.725W
9	Gogama	47.675N	81.727W
10	Timagami	47.067N	79.783W
11	Espanola	46.255N	81.768W
12	Sudbury South	46.420N	80.955W
13	Sault Ste. Marie	46.505N	84.250W
14	Chapleau	47.833N	83.400W
15	Wawa	48.050N	84.725W
16	Timmins	48.477N	81.364W
17	Kapuskasing	49.417N	82.433W
18	Sparrow Lake	44.798N	79.383W
23	Lake St. Peter	45.300N	78.033W
24	Lake Traverse	45.947N	78.064W
25	Shawanaga	45.533N	80.200W
26	Mattawa	46.287N	78.873W
27	Hearst	49.700N	83.667W
28	Hornepayne	49.218N	84.787W
29	Powassan	46.123N	79.246W

TABLE 3.1.1 (Continued)

No.	Station	Latitude	Longitude
31	Marten River	46.725N	79.833W
32	Verner	46.412	80.120W
33	River Valley	46.577N	80.188W
34	Sudbury North	46.488N	81.012W
35	South Bay Mouth	45.585N	82.012W
36	Tobermory	45.207N	81.523W
37	Owen Sound	44.491N	80.871W
38	Duck Island	45.821N	82.948W
39	Goderich	43.726N	81.724W
40	Goderich Tower	43.726N	81.724W
41	Goderich Buoy	43.766N	81.859W
42	Manitoulin Tower	45.821N	82.948W
43	Manitoulin Buoy	45.118N	82.939W
44	Wawa Tower	47.723N	84.809W
45	Red Rock	47.723N	84.809W
46	Goderich Tower 50	43.726N	81.724W
47	Goderich Tower 30	43.726N	81.724W
110	Gore Bay Airport r	45.881N	82.570W
140	Gore Bay Airport s	45.881N	82.570W
112	Thunder Bay Airport r	48.373N	89.320W
141	Thunder Bay Airport s	48.373N	89.320W
113	Wiarton Airport r	44.650N	81.233W
142	Wiarton Airport s	44.650N	81.233W
120	Sarnia Airport r	42.983N	82.283W
143	Sarnia Airport s	42.983N	82.283W
130	Caribou Island r	47.367N	85.833W
144	Caribou Island s	47.367N	85.833W
131	Copper Harbour r	47.469N	87.867W
145	Copper Harbour s	47.469N	87.867W
132	Isle Royale r	47.896N	89.216W
146	Isle Royale s	47.896N	89.216W

TABLE 3.1.1 (Continued)

No.	Station	Latitude	Longitude
133	Ney Provincial Park r	48.750N	86.567W
147	Ney Provincial Park s	48.750N	86.567W
134	Pinery Provincial Park r	43.233N	81.800W
148	Pinery Provincial Park s	43.233N	81.800W
135	Inverhuron Park r	44.300N	81.567W
149	Inverhuron Park s	44.300N	81.567W
137	Kilbear Provincial Park r	45.350N	80.200W
138	Southampton Buoy	44.325N	81.650W
150	Kilbear Provincial Park s	45.350N	80.200W
201	Porcupine Mountains State Park	46.818N	89.644W
202	Isle Royale	47.896N	89.216W
203	Fort Wilkins State Park	47.497N	87.867W
204	Baraga State Park	46.760N	88.506W
205	Pictured Rocks	46.664N	86.006W
206	Tahquamenon Falls	46.543N	75.035W
207	Mackinac Island State Park	45.858N	84.617W
208	U.S. Coast Guard Station Alpena	45.034N	83.239W
209	U.S. Coast Guard Station Tawas Point	44.254N	83.447W
210	Albert Sleeper State Park	43.977N	83.211W
211	Port Sanilac	43.429N	82.552W
212	Bayfield	46.786N	90.865W
213	Tawas Buoy	44.225N	83.422W
214	Grand Marais	47.735N	90.334W
215	Benton Harbor	42.150N	86.384W
216	Silver Lake	43.608N	86.521W
217	Beaver Island	45.681N	85.508W
218	Two Harbors	47.002N	91.652W
219	Alpena Buoy	45.167N	83.217W
301	Mount Forest - wet	43.164N	80.750W
302	Armstrong -wet	50.283N	88.090W
303	Woodbridge - wet	43.795N	79.552W

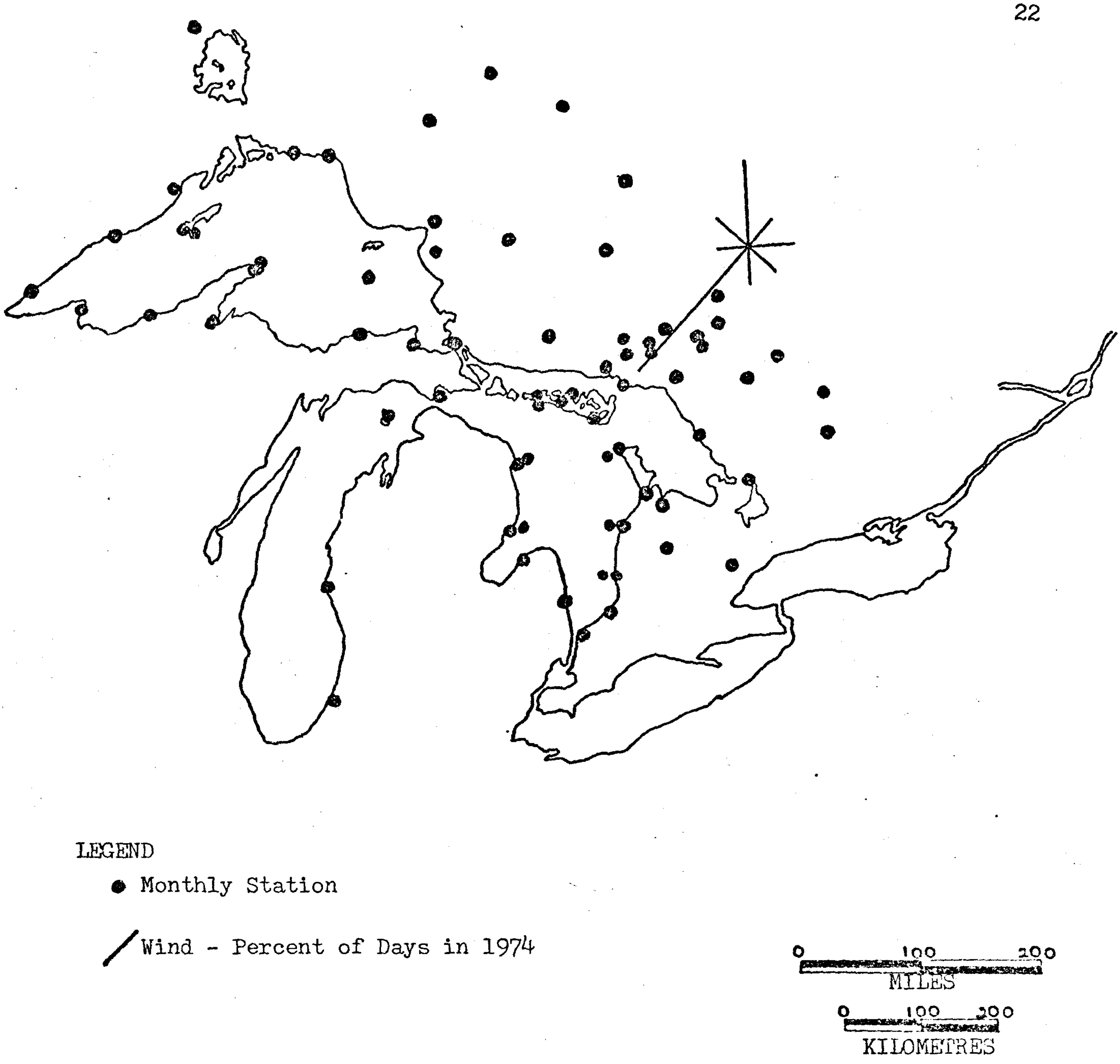


Figure 3.1.1: Precipitation chemistry stations and wind directions in the Upper Great Lakes region

3.2 Codes and Units of Parameters Considered in this Study

Table 3.2.1 shows the parameters used in this study. Not all the parameters listed in this table were used in the six areas considered. Those used in the various areas are given in Appendix B.

TABLE 3.2.1: Codes and units of parameters

Name	Code No.	Definition	Units Concentration
Cd T	101	Total cadmium	micro-g/l
Cu F	106	Filtered copper	micro-g/l
Cu T	107	Total copper	micro-g/l
Fe T	110	Total iron	micro-g/l
Pb T	113	Total lead	micro-g/l
Ni T	116	Total nickel	micro-g/l
Zn F	118	Filtered zinc	micro-g/l
Zn T	119	Total zinc	micro-g/l
SP CON	122	Specific conductivity	umho/cm
pH	123	Hydrogen ion concentration	
SO_4	127	Sulphate	mg/l
T PART	128	Total particulate weight	g/l
P T	240	Total phosphorus	micro-g/l

3.3 Data and Spectra

3.3.1 Sine and cosine functions

Figures 3.3.1 and 3.3.2 show the data and trends for the sine and cosine functions respectively. Note that the trends are not straight lines of zero slopes and zero intercepts. This is due to termination errors which result since a finite number of points are used. Figures 3.3.3 and 3.3.4 show the functions after the trends are removed. As expected, small trends result due to the least squares fit.

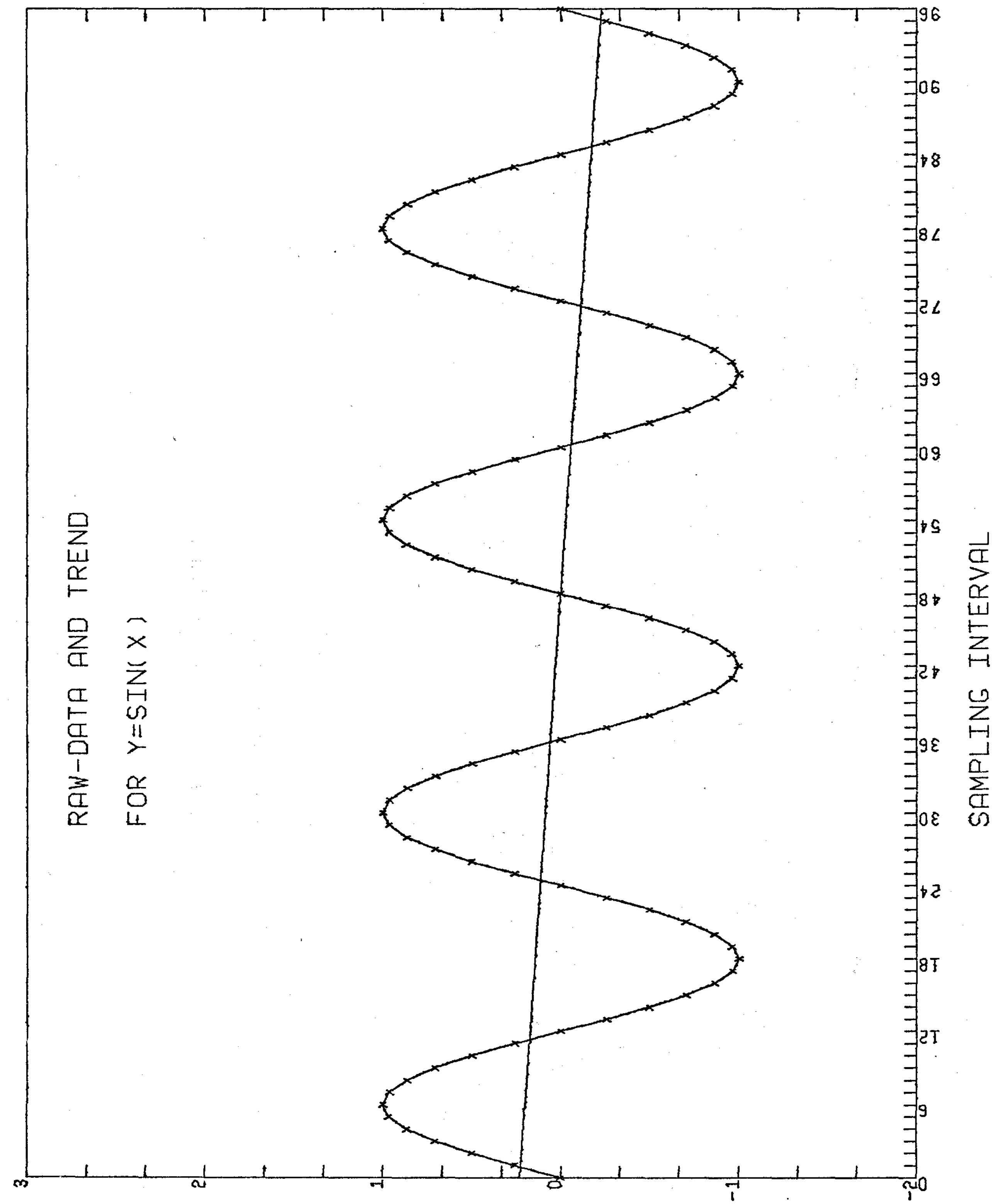


Figure 3.3.1: Sine function and trend for the domain 0 to 8π (24 sampling intervals = 2π)

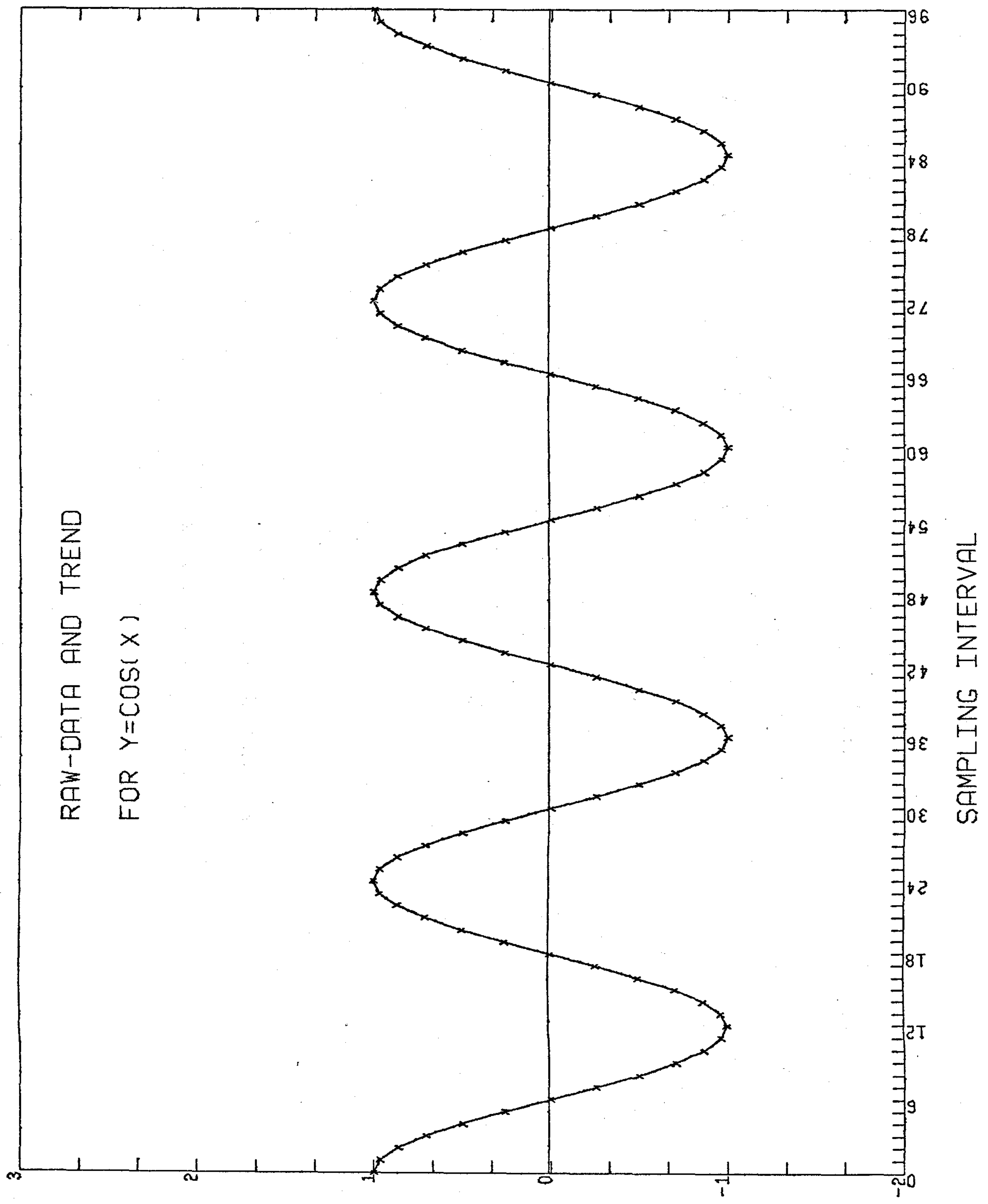


Figure 3.3.2: Cosine function and trend for the domain 0 to 8π (24 sampling intervals = 2π)

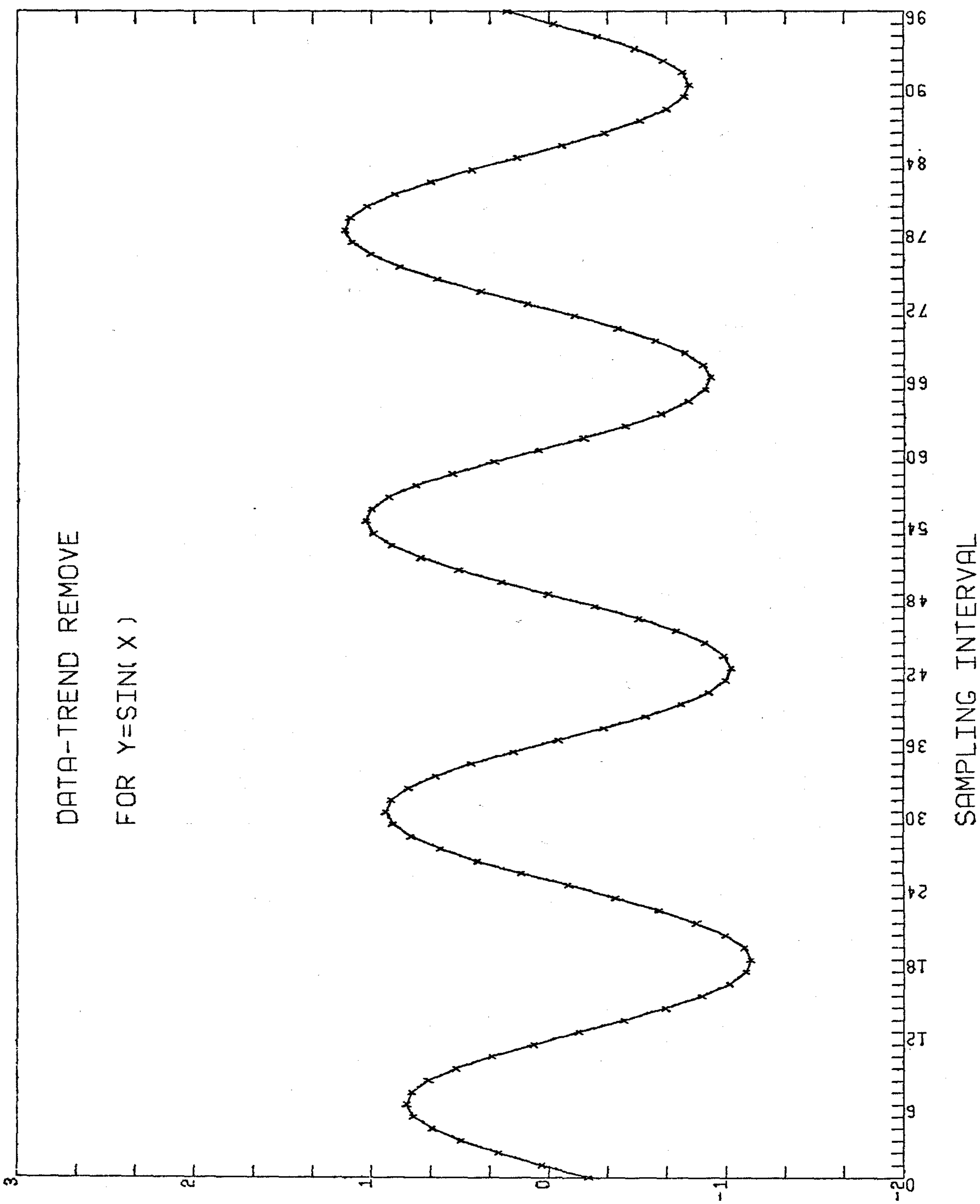


Figure 3.3.3: Sine function of Figure 3.3.1 after trend removed

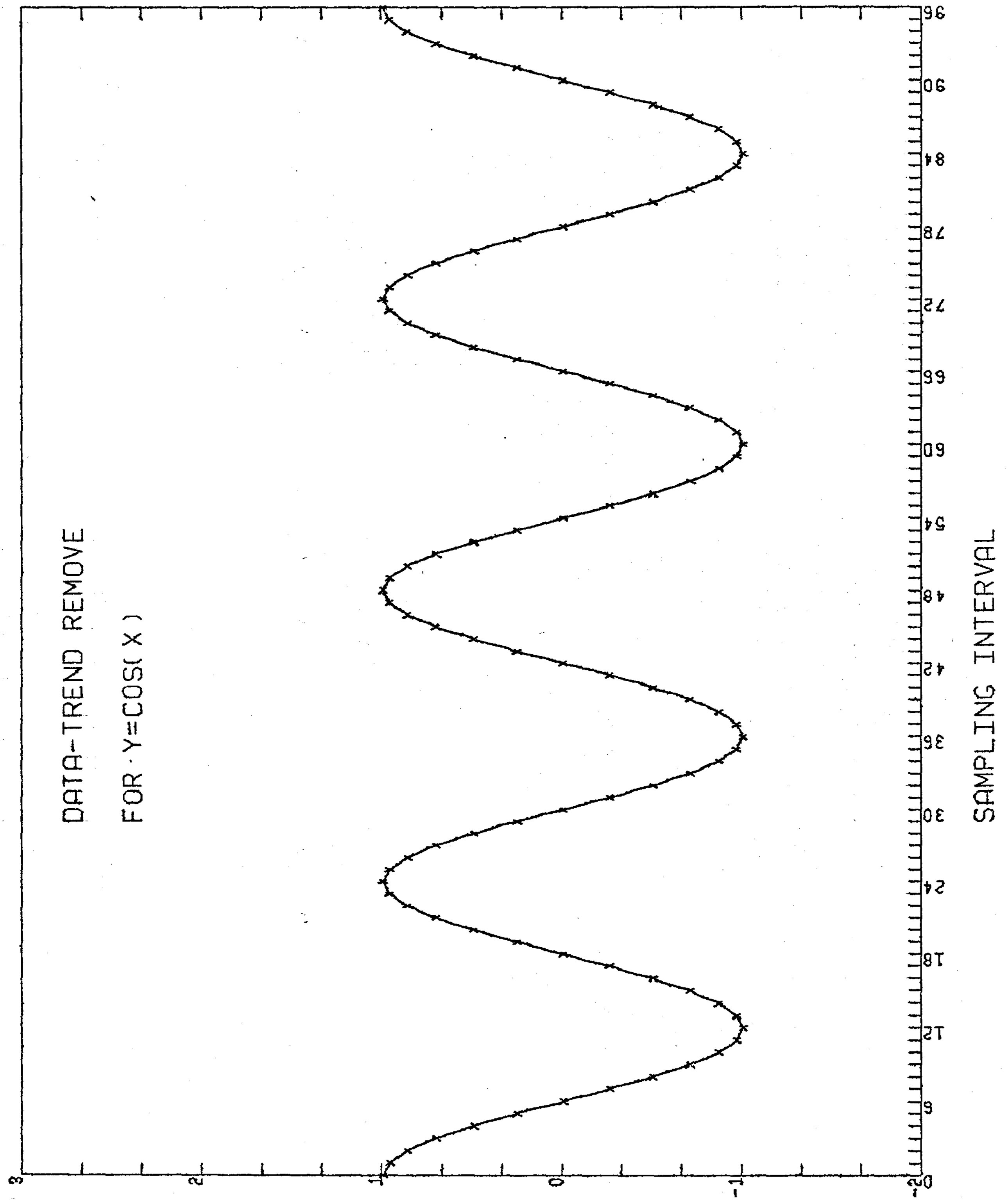


Figure 3.3.4: Cosine function of Figure 3.3.2 after trend removed

The effects of various lags on the power spectrum are shown in Figures 3.3.5 through 3.3.10. The peaks in the spectra indicate the periodicity. The peaks are very close to 2π, which corresponds to 24 sampling intervals. In the spectra, the peak positions are given in terms of lags which has to be converted to sampling intervals.

Figures 3.3.5 and 3.3.6, for which the lags equal 32, show essentially pure spectra. As the lags increase the resolution increases (peaks are sharper) but the spectra become contaminated with "ripples" and the statistical reliability decreases.

To illustrate the fact that any obvious trends in the data must be removed before analysis, a linear trend is added to the sine function (Figure 3.3.11). The power spectrum is shown in Figure 3.3.12. The peak is more or less "washed-out". Hence it is absolutely necessary to remove any trends from the data before using the Fourier analysis.

The phase and squared coherency spectra were not plotted for the trigonometric functions, but the values obtained agree quite well with the expected results. The correlation at the 2π periodicity is 0.99 and the phase difference is 90.0 degrees.

3.3.2 Errors

The peaks in the power spectra do not occur exactly at 24 sampling intervals. Table 3.3.1 gives the periodicity for the various lags and the cross correlation properties. From the spectra, it can be seen that if smooth curves are to be drawn through the points then the periodicity would be very close to the lower values given in the Table. Even though there is some variation in the periodicity, the correlation and phase are quite consistent and very accurate.

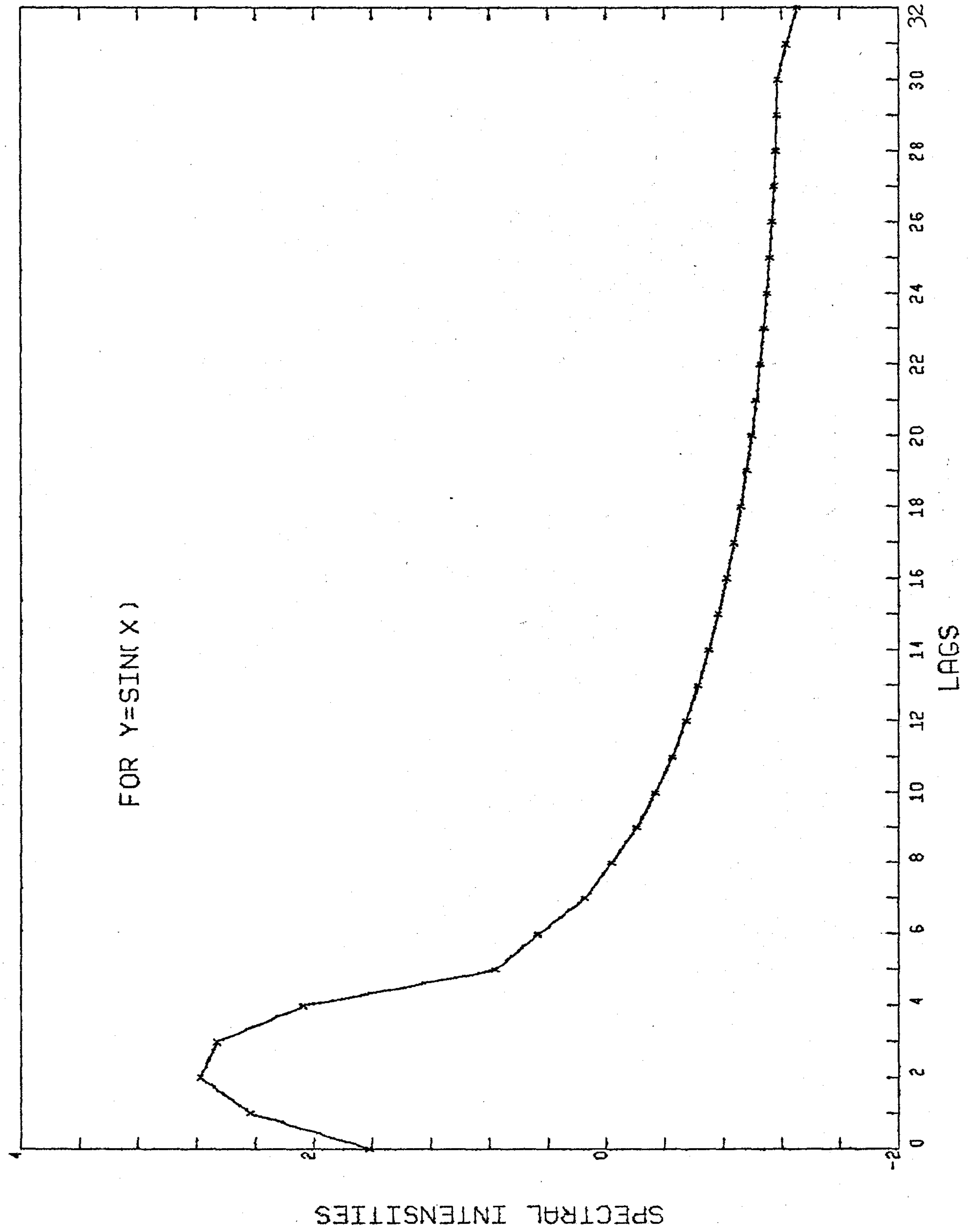


Figure 3.3.5: Power spectrum for data of Figure 3.3.3, lags = 32

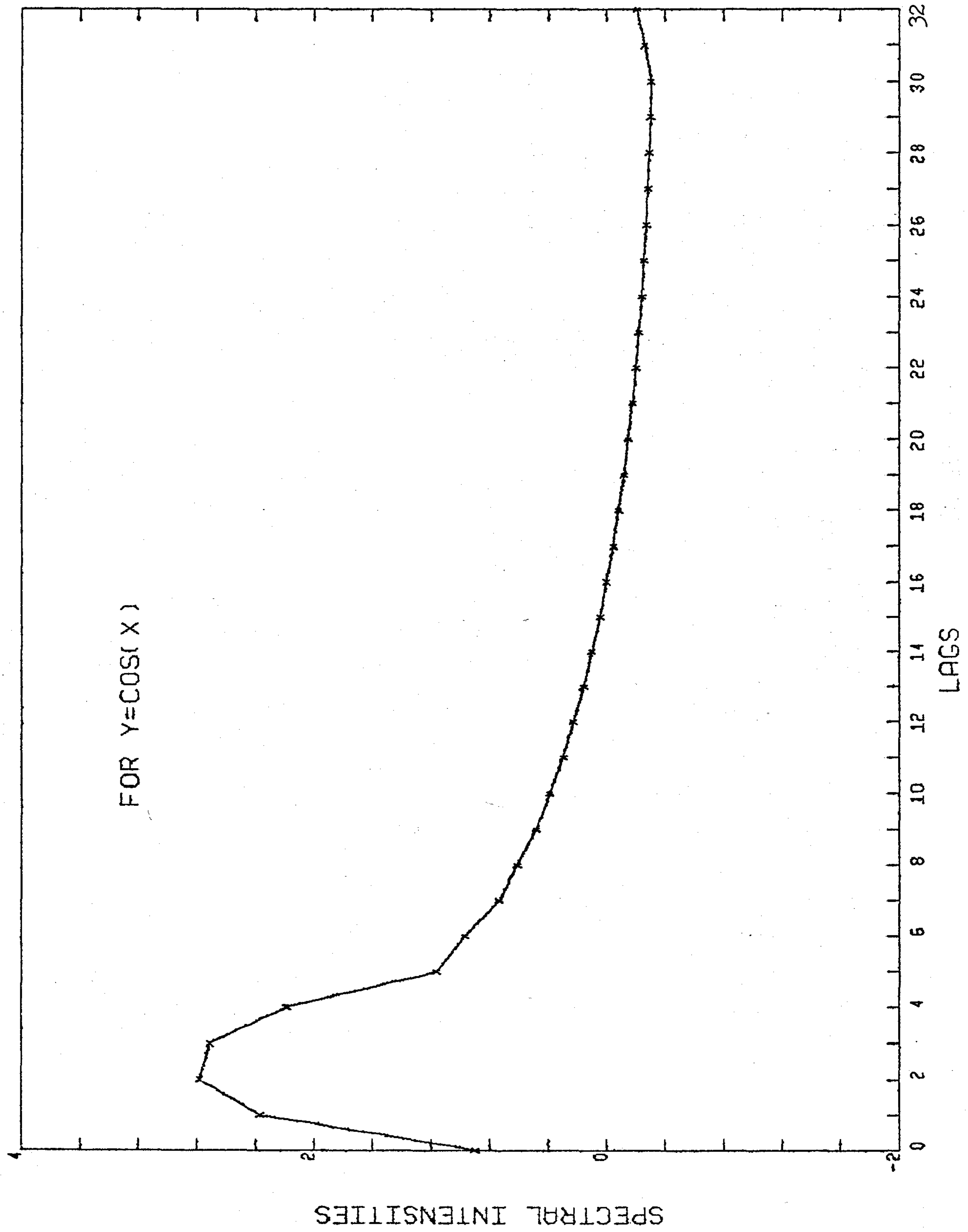


Figure 3.3.6: Power spectrum for data of Figure 3.3.4, lags = 32

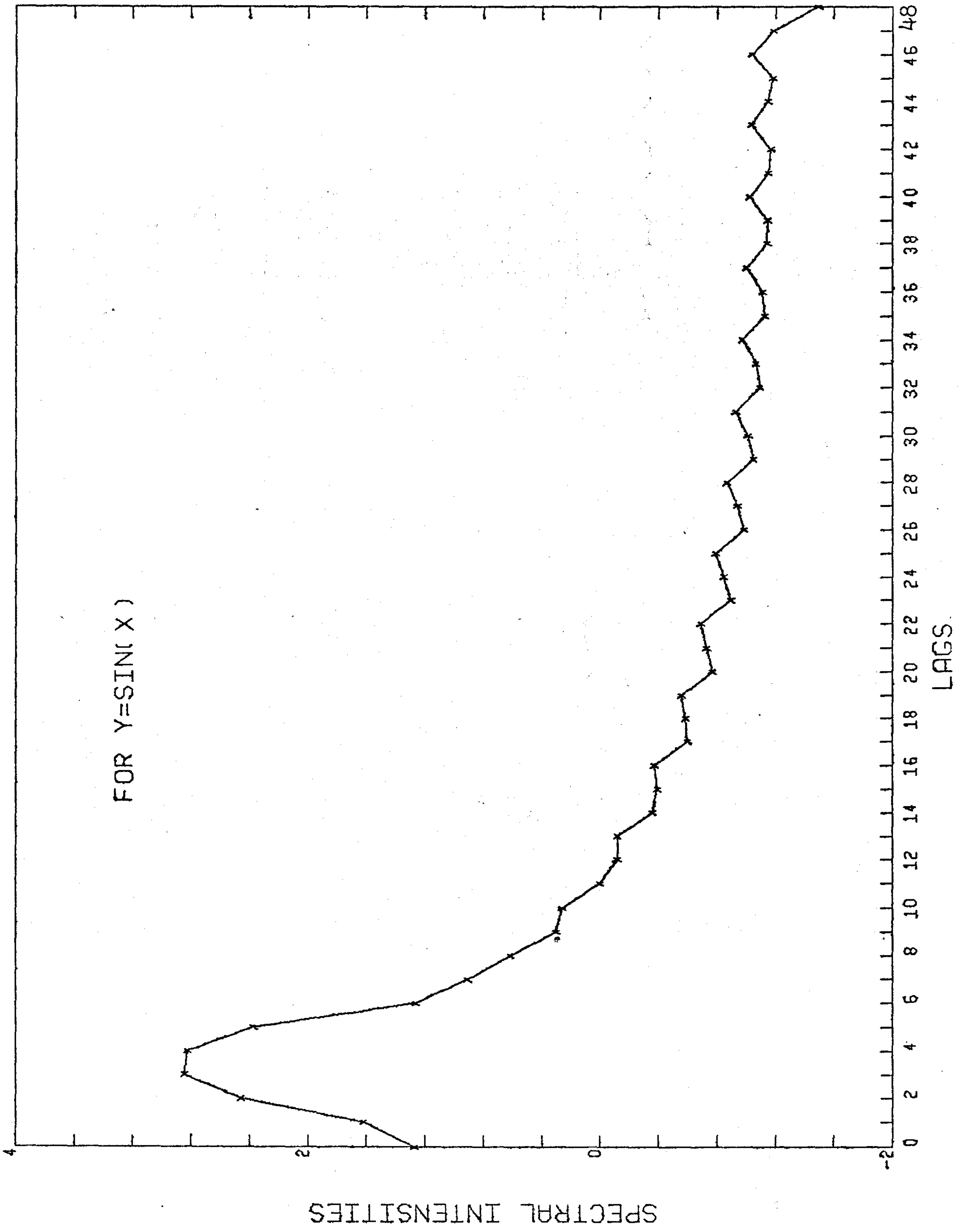


Figure 3.3.7: Power spectrum for data of Figure 3.3.3, lags = 48

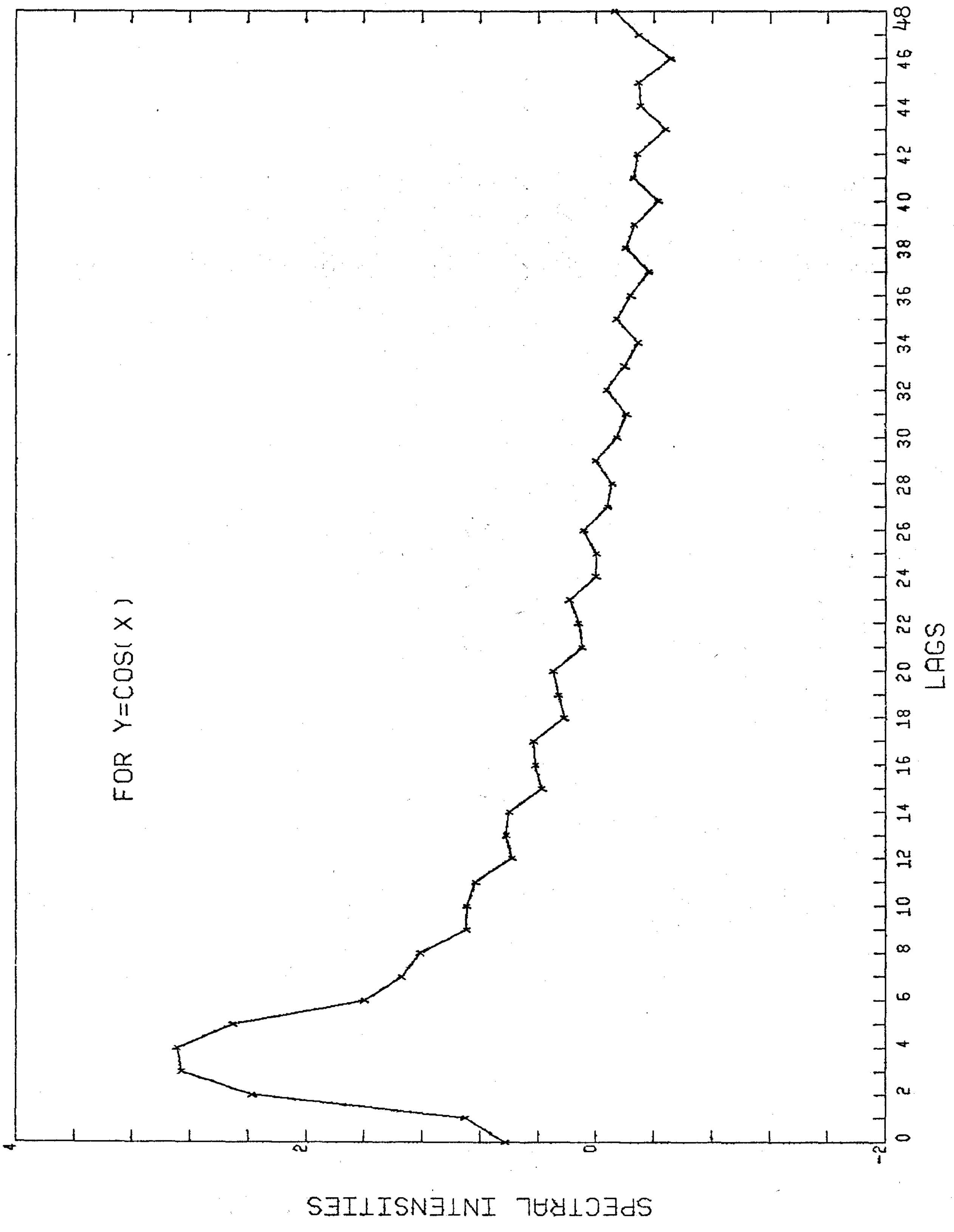


Figure 3.3.8: Power spectrum for data of Figure 3.3.4, lags = 48

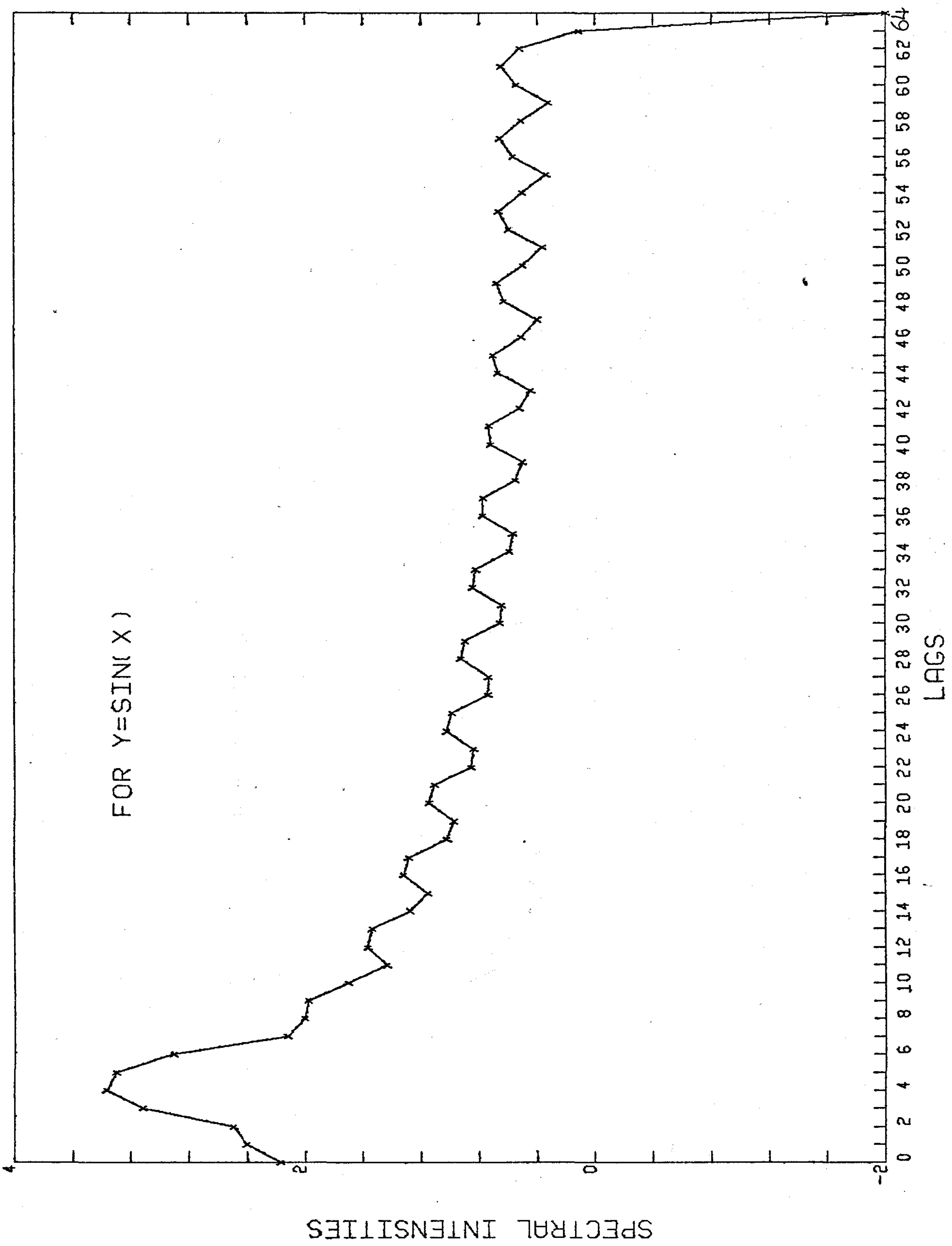


Figure 3.3.9: Power spectrum for data of Figure 3.3.3, lags = 64

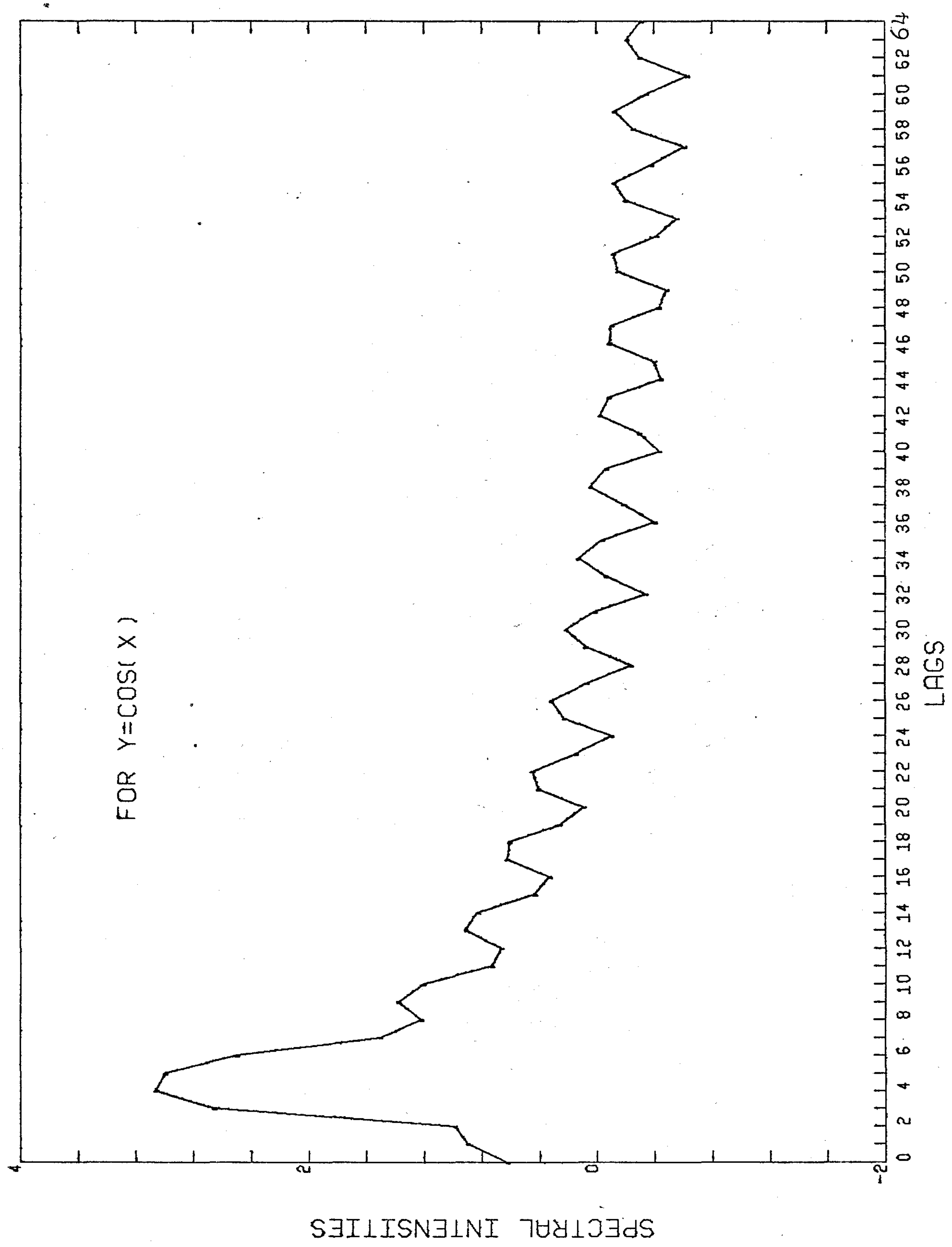


Figure 3.3.10: Power spectrum for data of Figure 3.3.4, lags = 64

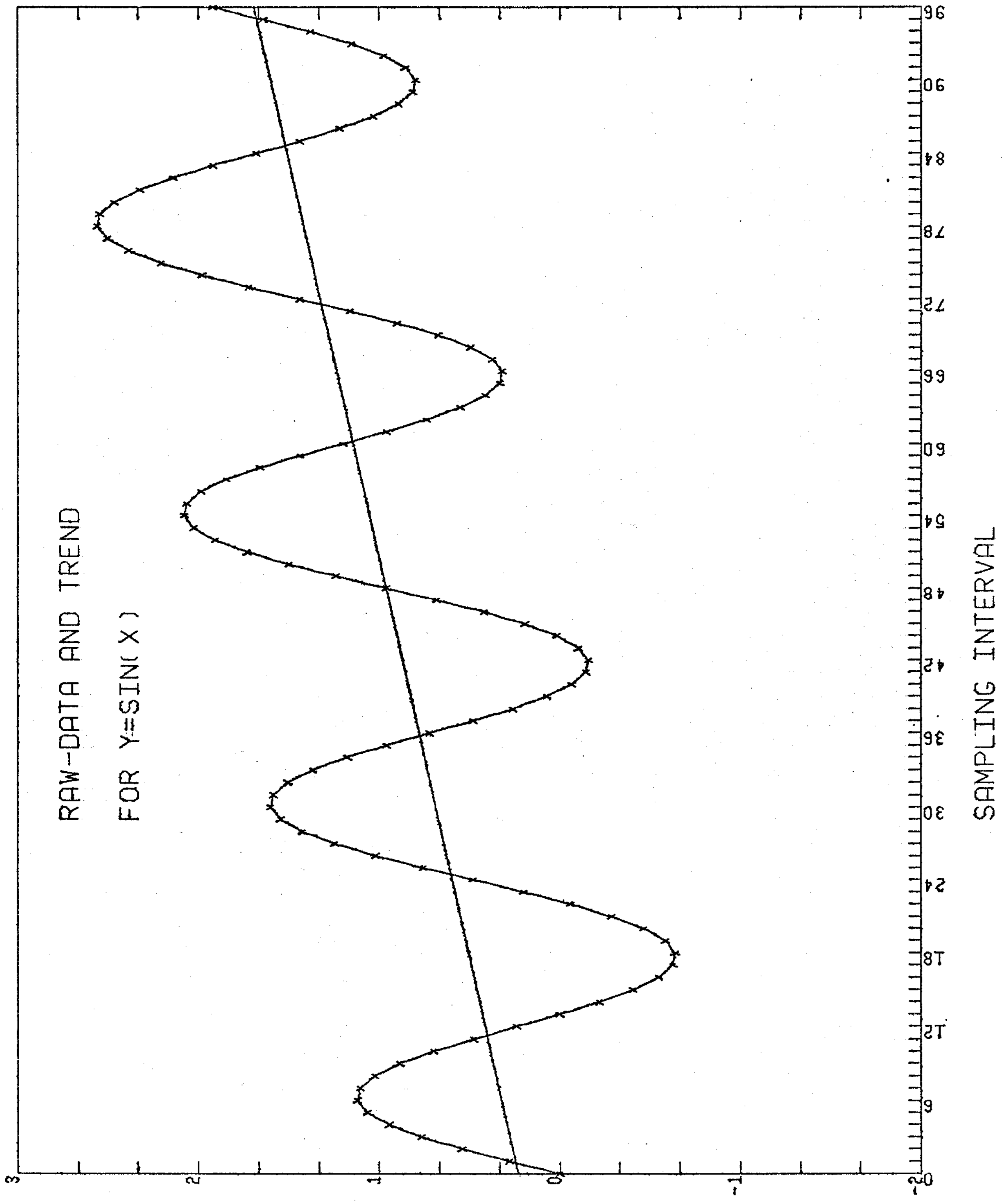


Figure 3.3.11: Data for linear trend added to the sine function

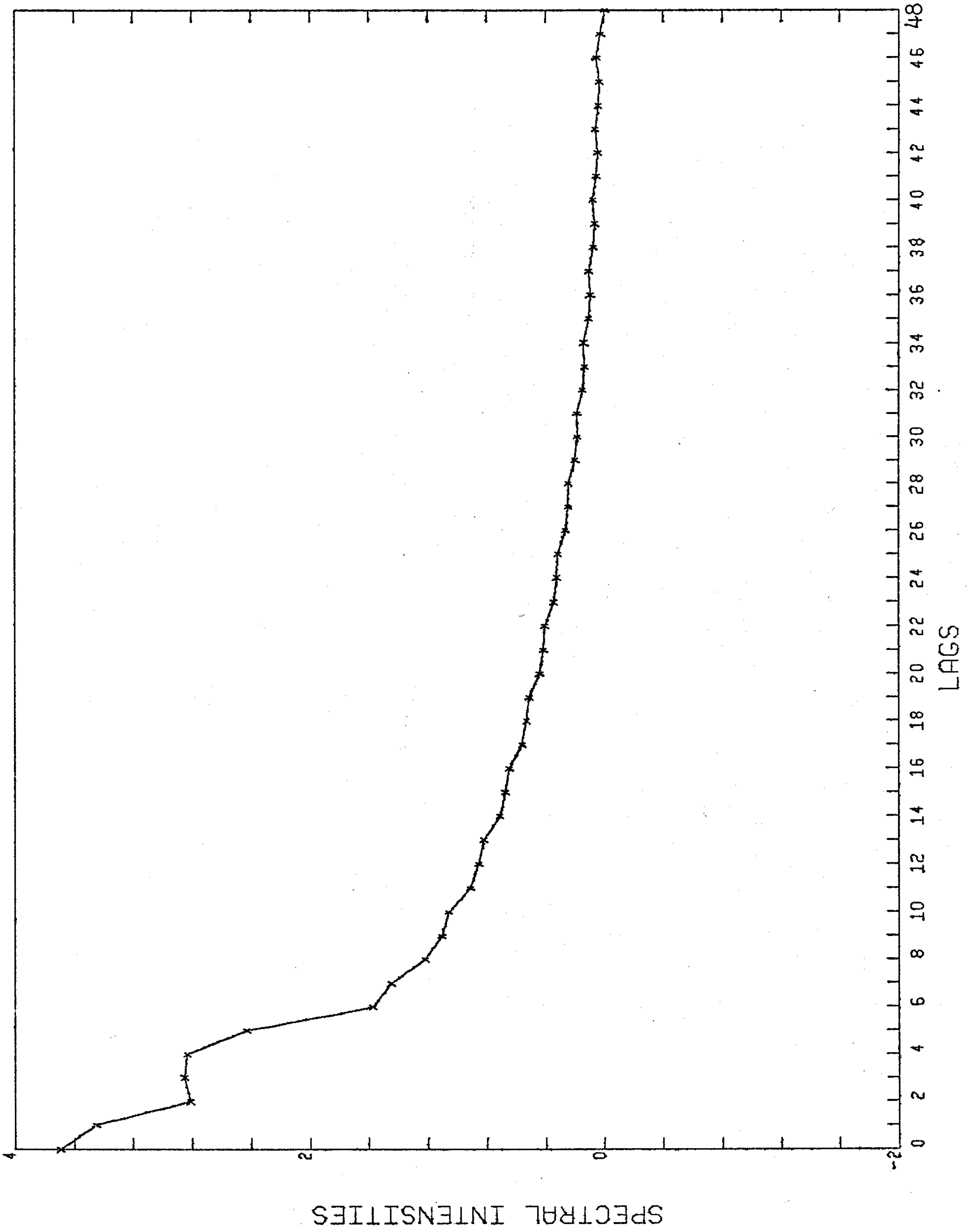


Figure 3.3.12: Power spectrum for data of Figure 3.3.11, lags = 48

TABLE 3.3.1:

Periodic and cross-correlation properties for the trigonometric functions. (24 sampling intervals = 2π).

Lags	Period (sampling intervals) sin x and cos x	Correlation	Phase (degree)
32	21.3 - 32.0	.99	90.0
48	24.0 - 32.0	.99	90.0
64	25.6 - 32.0	.99	90.0

The reason why the periodicity does not occur at exactly 24 sampling intervals is due mainly to the finite amount of points used. The least squares fit is not quite adequate and leads to a small error. Furthermore, there is no definite way to determine the number of lags to be used. However, one must compromise between resolution and statistical reliability. Finally, another source of error is the graphing of the spectra which consist of straight line segments rather than smooth curves.

3.3.3 Precipitation data and spectra

The data for the parameters studied can be displayed in two forms. The first consists of the raw data and trend. The resulting data after the trend is removed is the second form. Note that negative residuals are set equal to zero. The second form is used in the actual analysis. Due to space limitation, only the raw data and trend are given in this work, since they contain more information. However, the second form of data can easily be visualized if one considers the trend line as the time-axis. The data are given in Appendix B.

Three types of data are studied. The first consists of the individual stations 12, 2, 3, and 4. The second consists of data which is the average over eleven stations (2, 3, 4, 5, 6, 8, 9, 10, 11, 12, 13). This will be referred to as the Sudbury area. The third type is the data which is the average of the Upper Great Lakes area. This consists of the stations listed in Table 3.1.1.

The Figures to follow are the spectra of pairs of data sets representing two simultaneously recorded time series. The figures show the parameters, the length of the data and locations. The parameters considered here are listed in Table 3.3.2. Spectra for other parameters are given in Appendix B.

TABLE 3.3.2: Parameters, locations and length of data.

Parameters	Area	Time Interval
Cu T & Ni T	Station 12	7210-7511
Cu T & Ni T	Station 2	7206-7511
Cu T & Ni T	Station 3	7211-7511
Cu T & Ni T	Station 4	7204-7511
Cu T & Ni T	Sudbury area	7208-7511
Cu T & Ni T	U.G.L. area	7201-7511
Cd T & Ni T	Station 12	7210-7511
Cd T & Ni T	Station 2	7206-7510
Cd T & Ni T	Station 3	7211-7511
Cd T & Ni T	Station 4	7204-7507
Cd T & Ni T	Sudbury area	7208-7511
Cd T & Ni T	U.G.L. area	7201-7511
Zn T & Zn F	Sudbury area	7204-7511
Zn T & Zn F	U.G.L. area	7204-7511
Cu T & Cu F	Sudbury area	7204-7511
Cu T & Cu F	U.G.L. area	7204-7511
pH & T PART	Station 12	7206-7511
pH & T PART	Station 2	7206-7511
pH & T PART	U.G.L. area	7204-7511

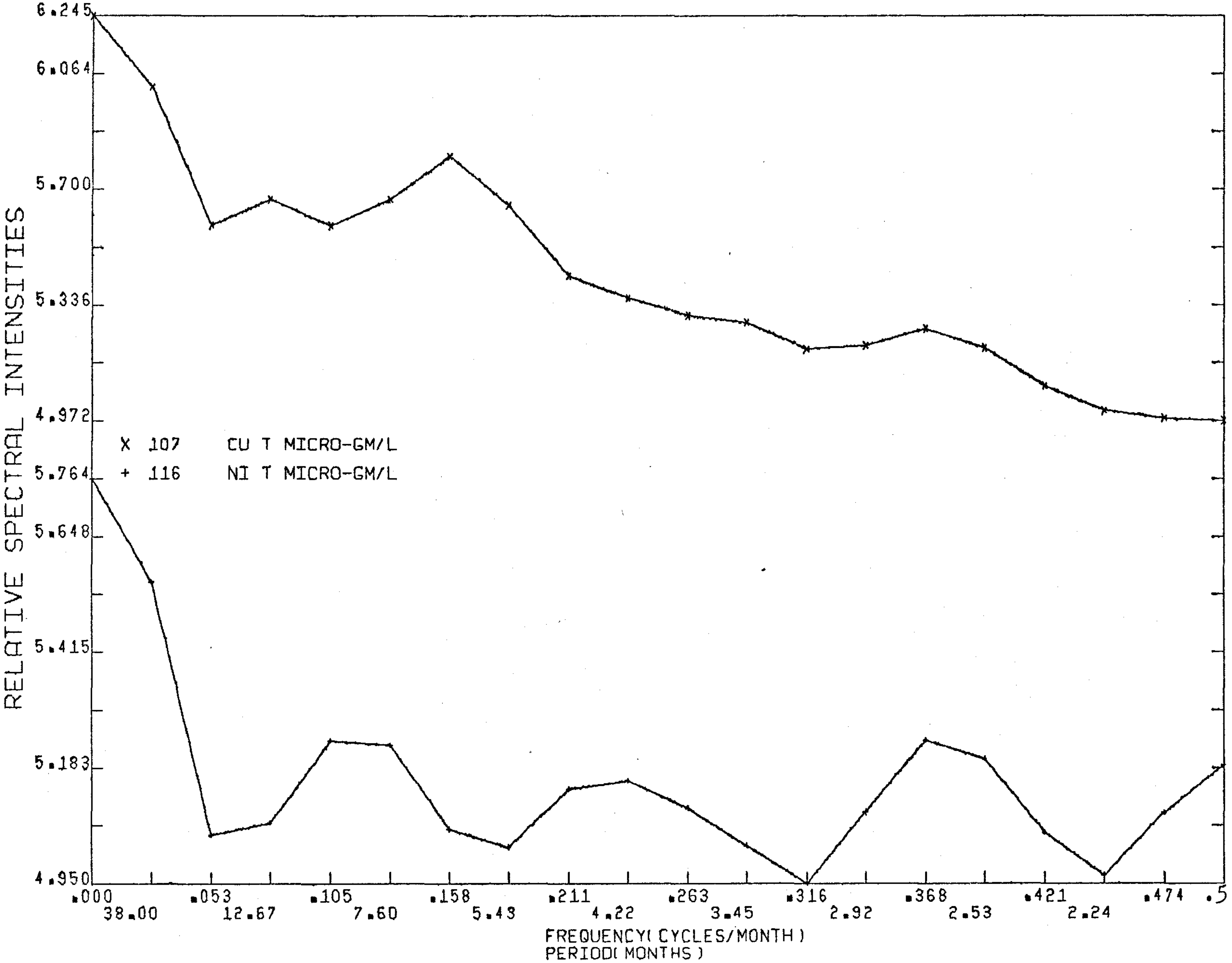


Figure 3.3.13: Power spectrum for Cu T & Ni T, station 12 (7210-7511)

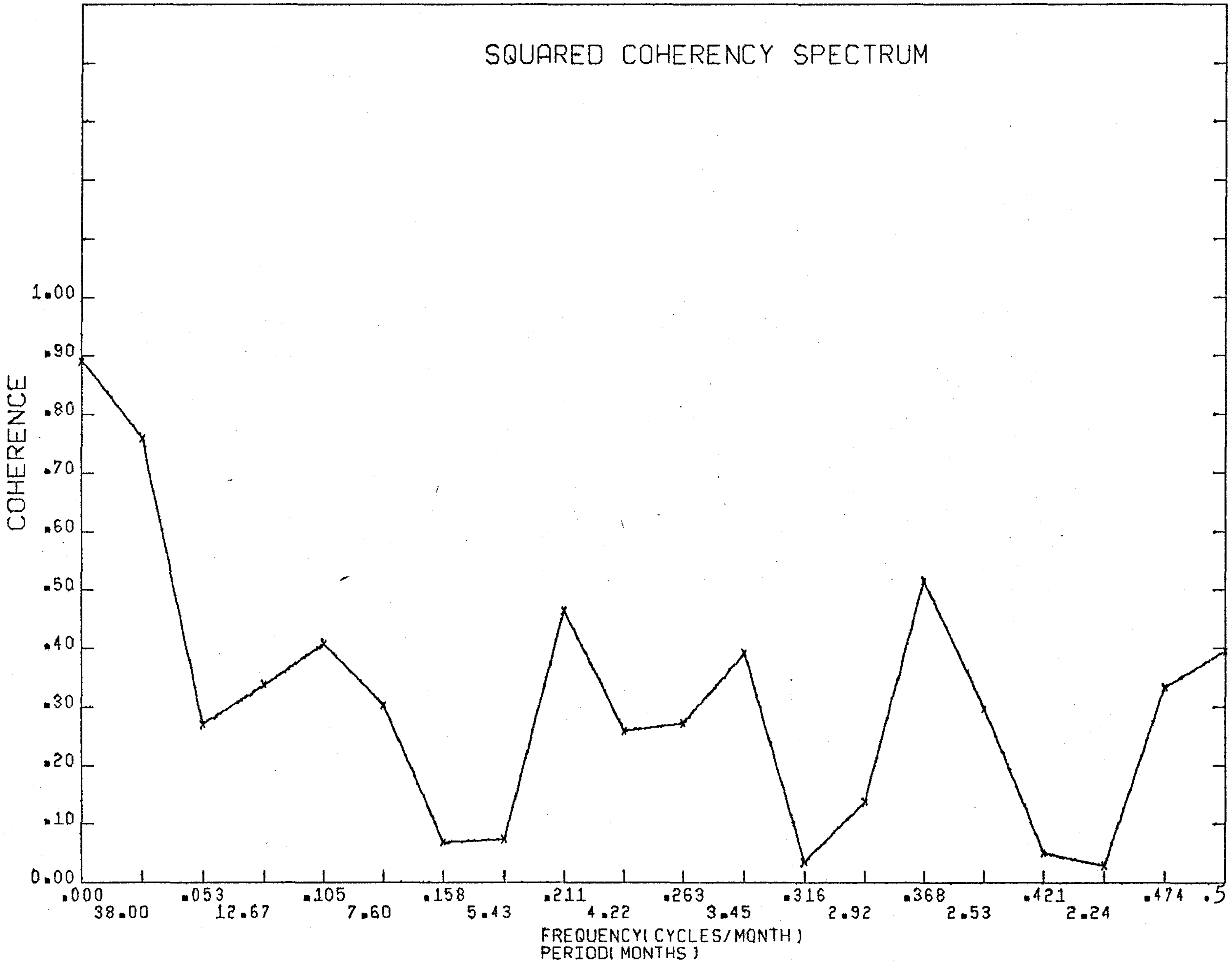


Figure 3.3.14: Coherency spectrum for Cu T & Ni T, station 12 (7210-7511)

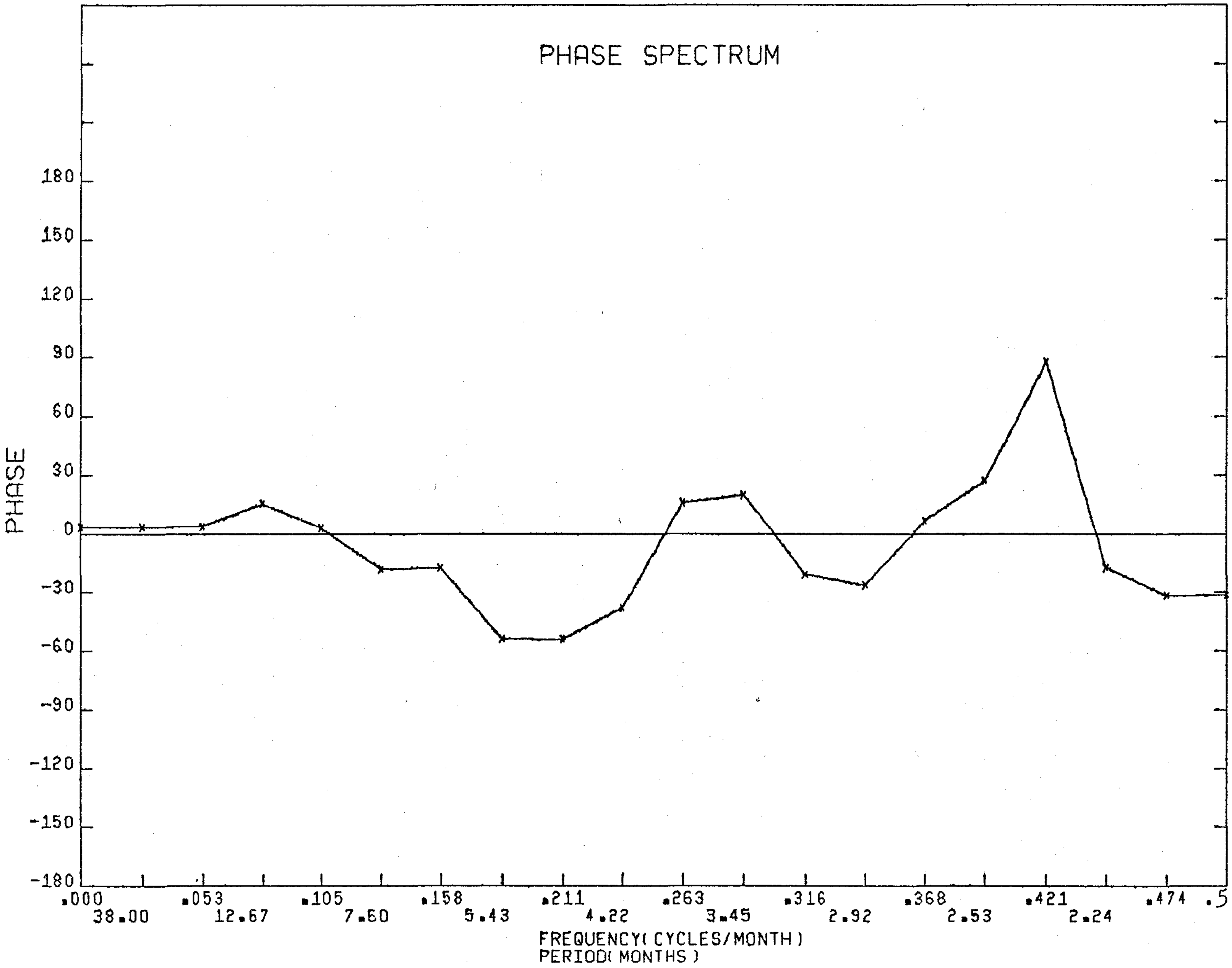


Figure 3.3.15: Phase spectrum for Cu T & Ni T, station 12 (7210-7511)

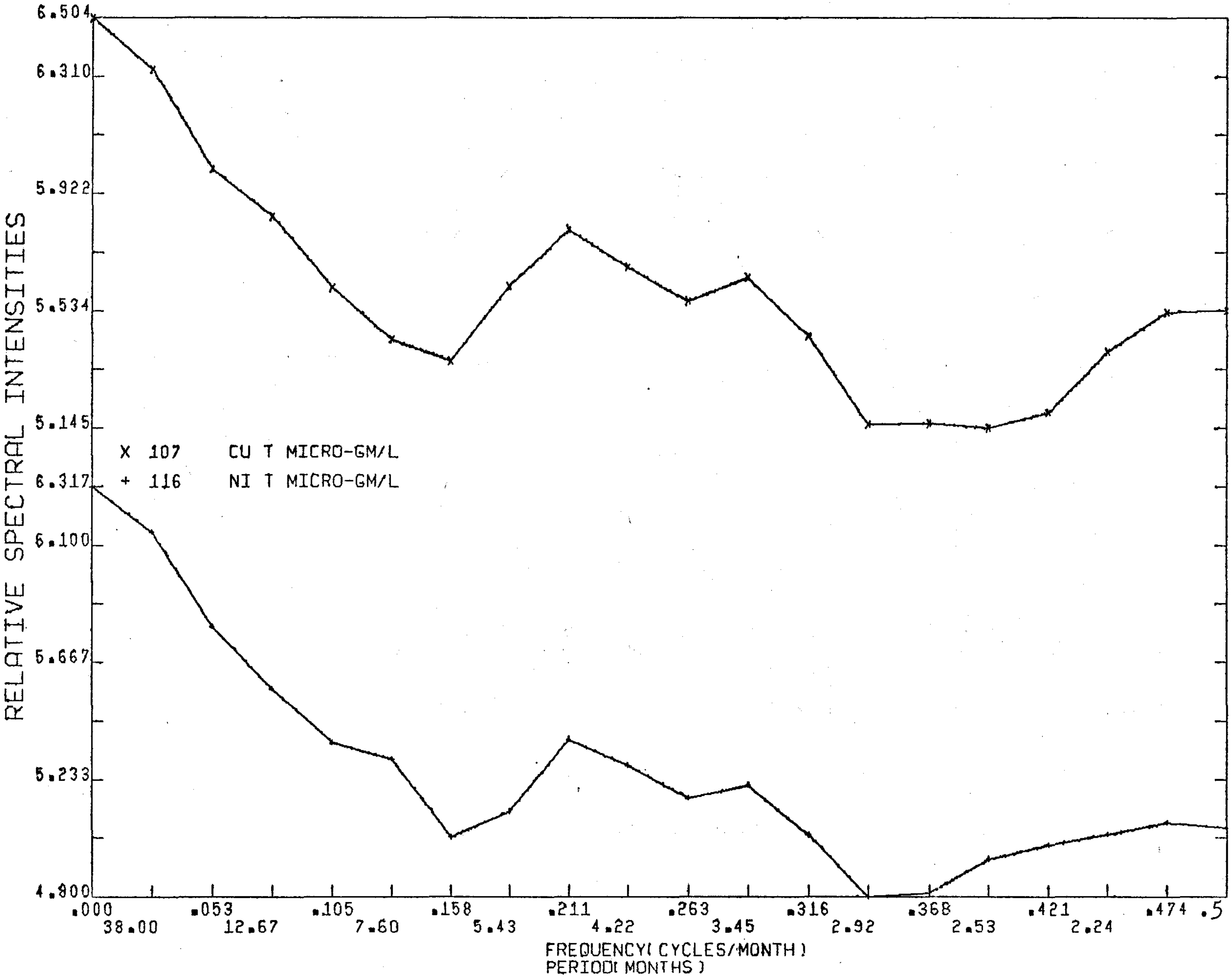


Figure 3.3.16: Power spectrum for Cu T & Ni T, station 2 (7206-7511)

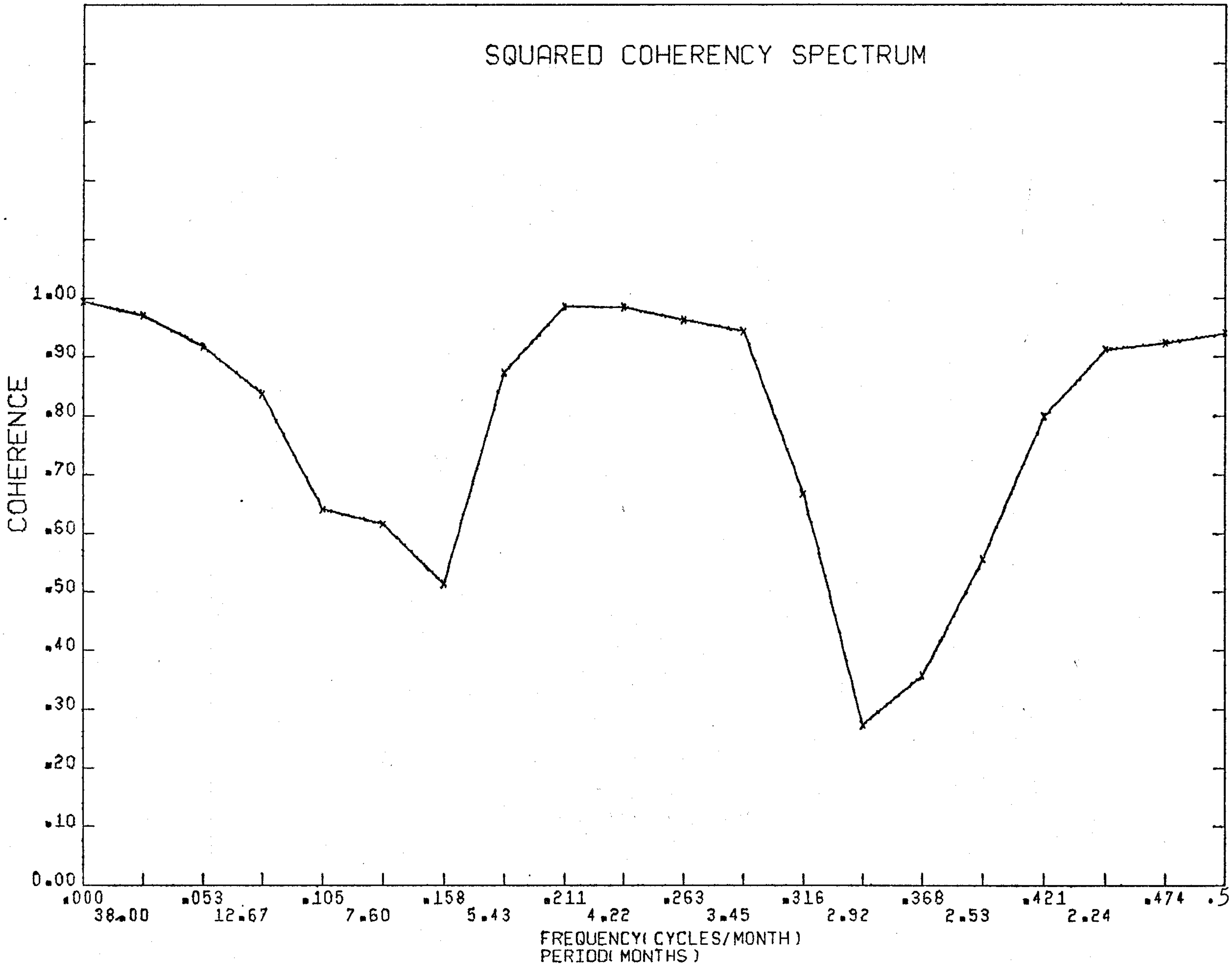


Figure 3.3.17: Coherency spectrum for Cu T & Ni T, station 2 (7206-7511)

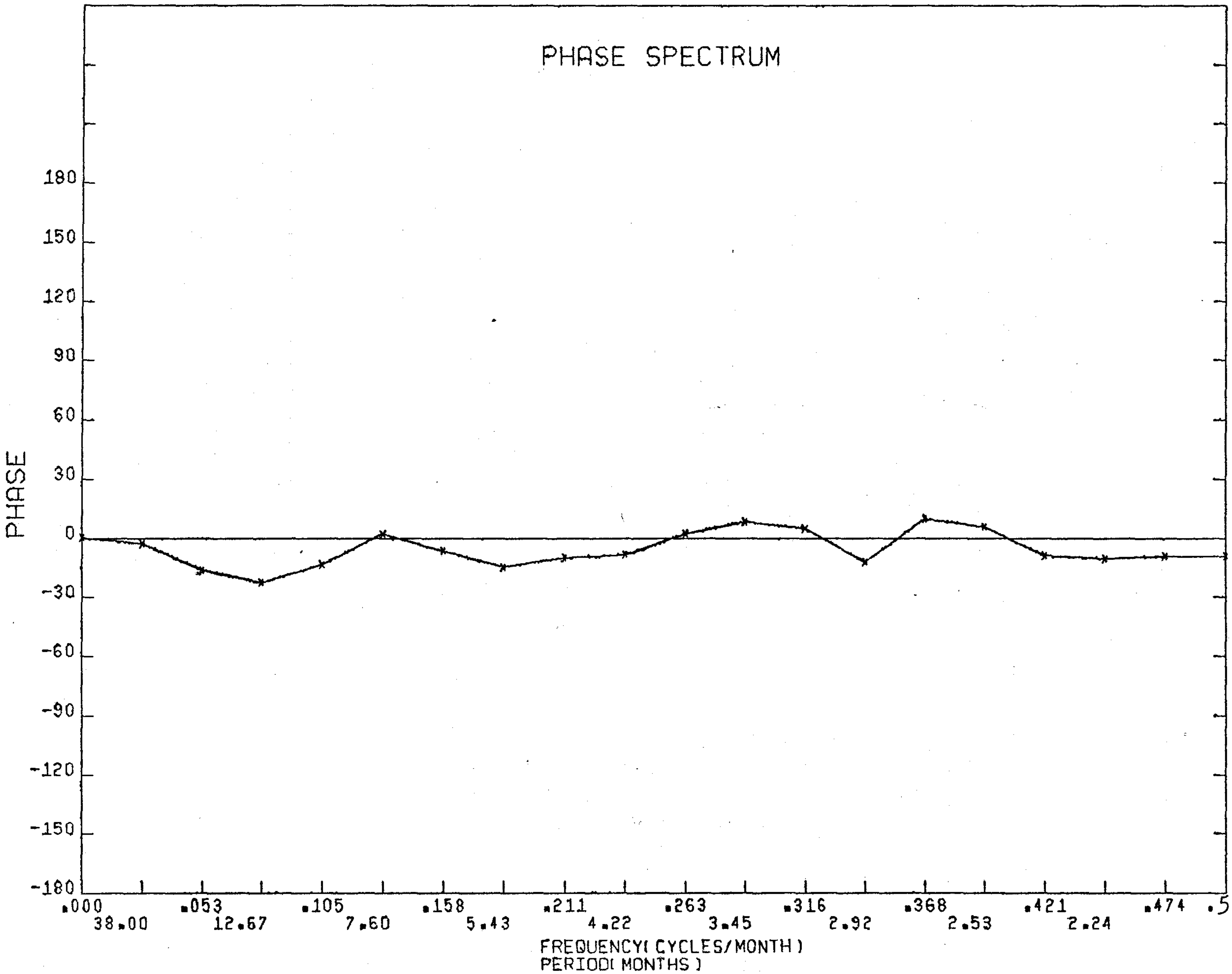


Figure 3.3.18: Phase spectrum for Cu T & Ni T, station 2 (7206-7511)

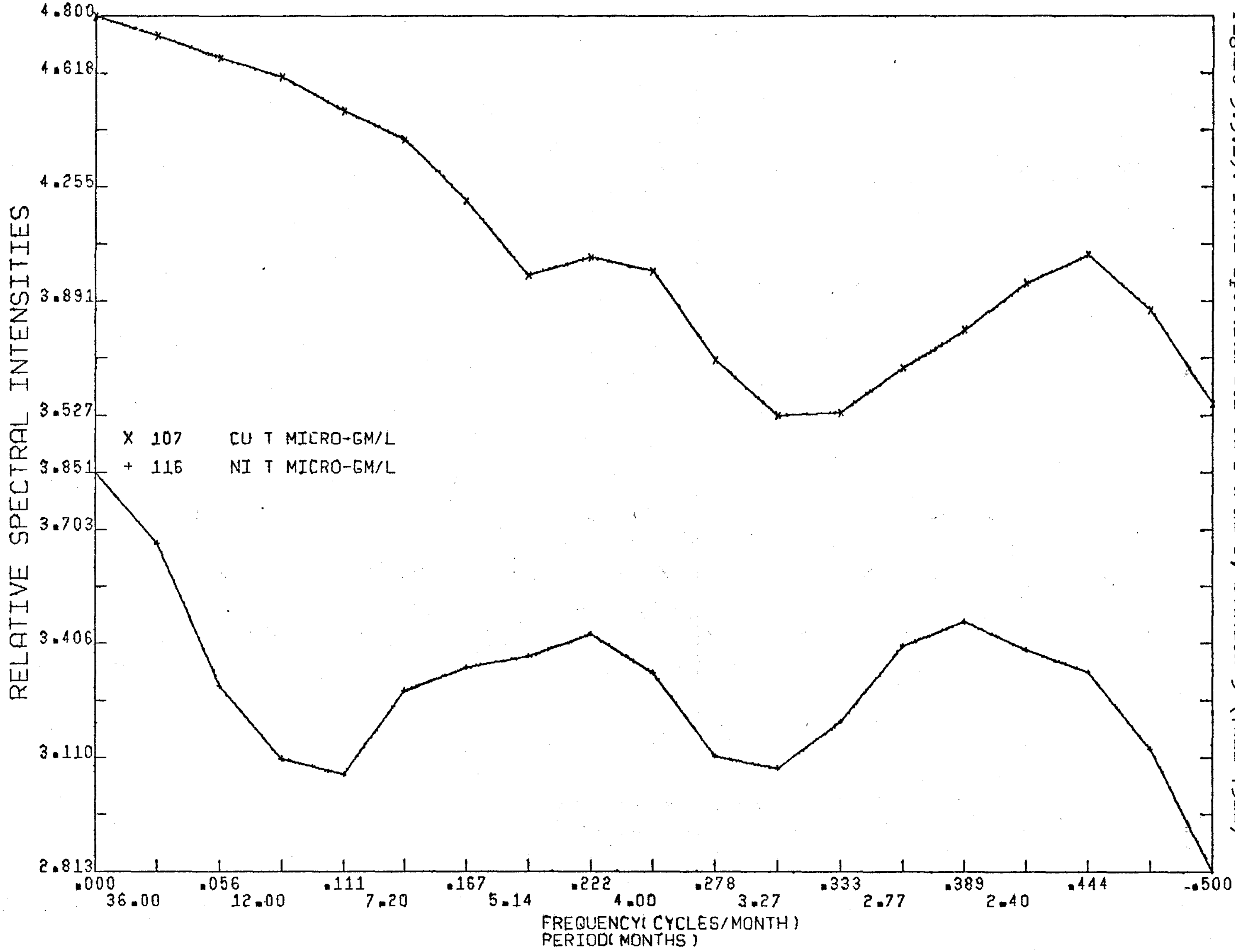
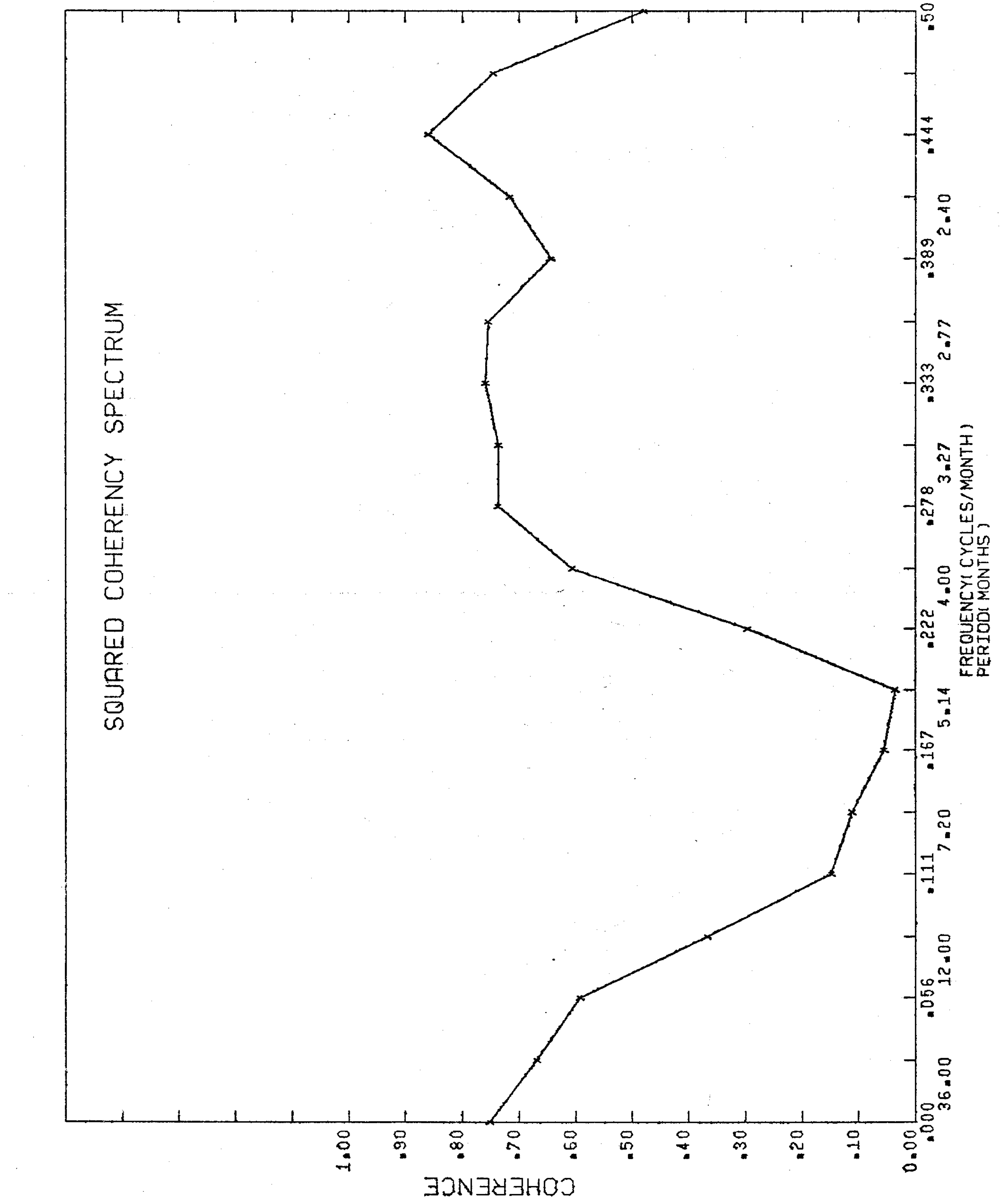


Figure 3.3.19: Power spectrum for Cu T & Ni T, station 3 (7211-7511)

Figure 3.3.20: Coherency spectrum for Cu T & Ni T, station 3 (7211-7511)



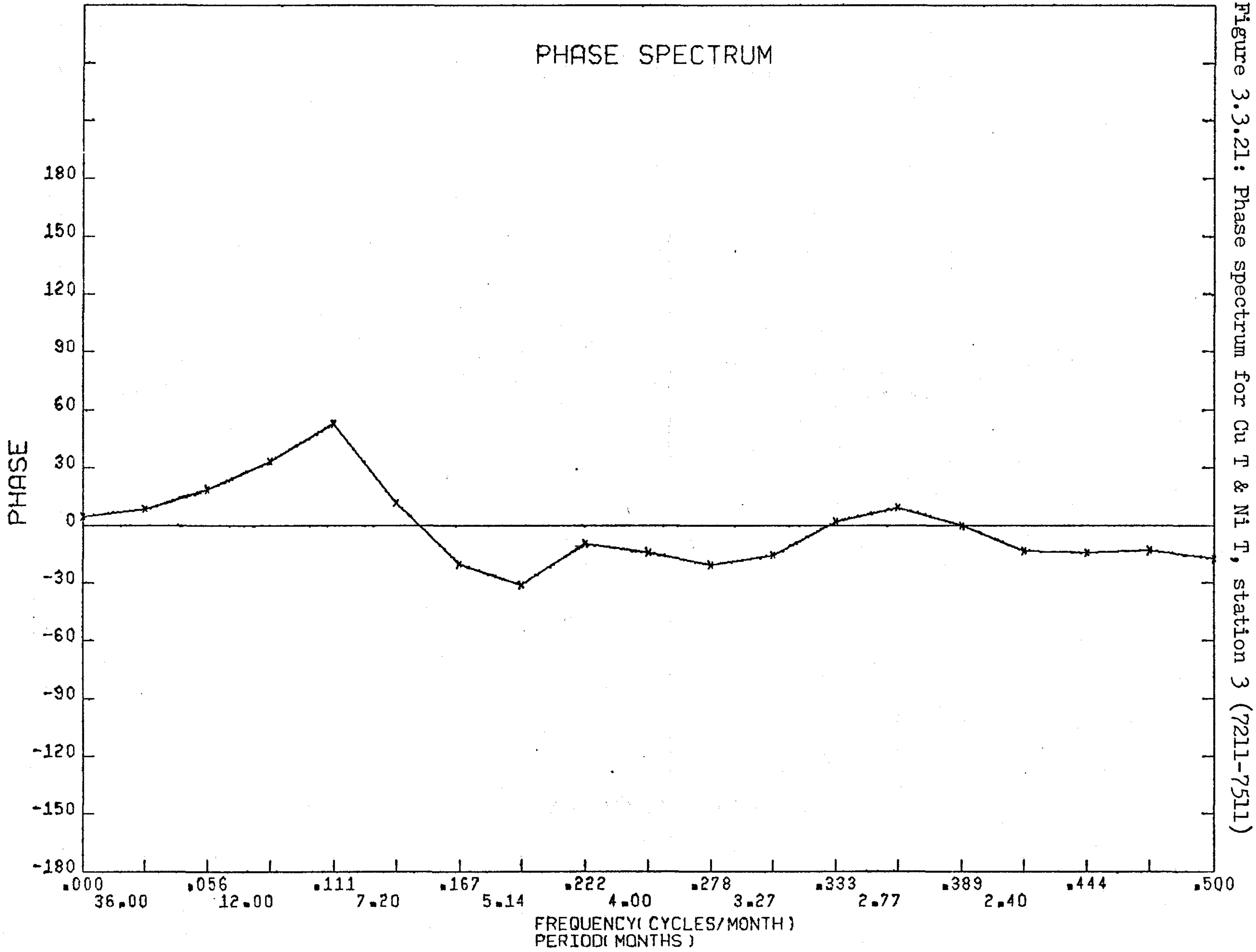


Figure 3.3.21: Phase spectrum for Cu T & Ni T, station 3 (7211-7511)

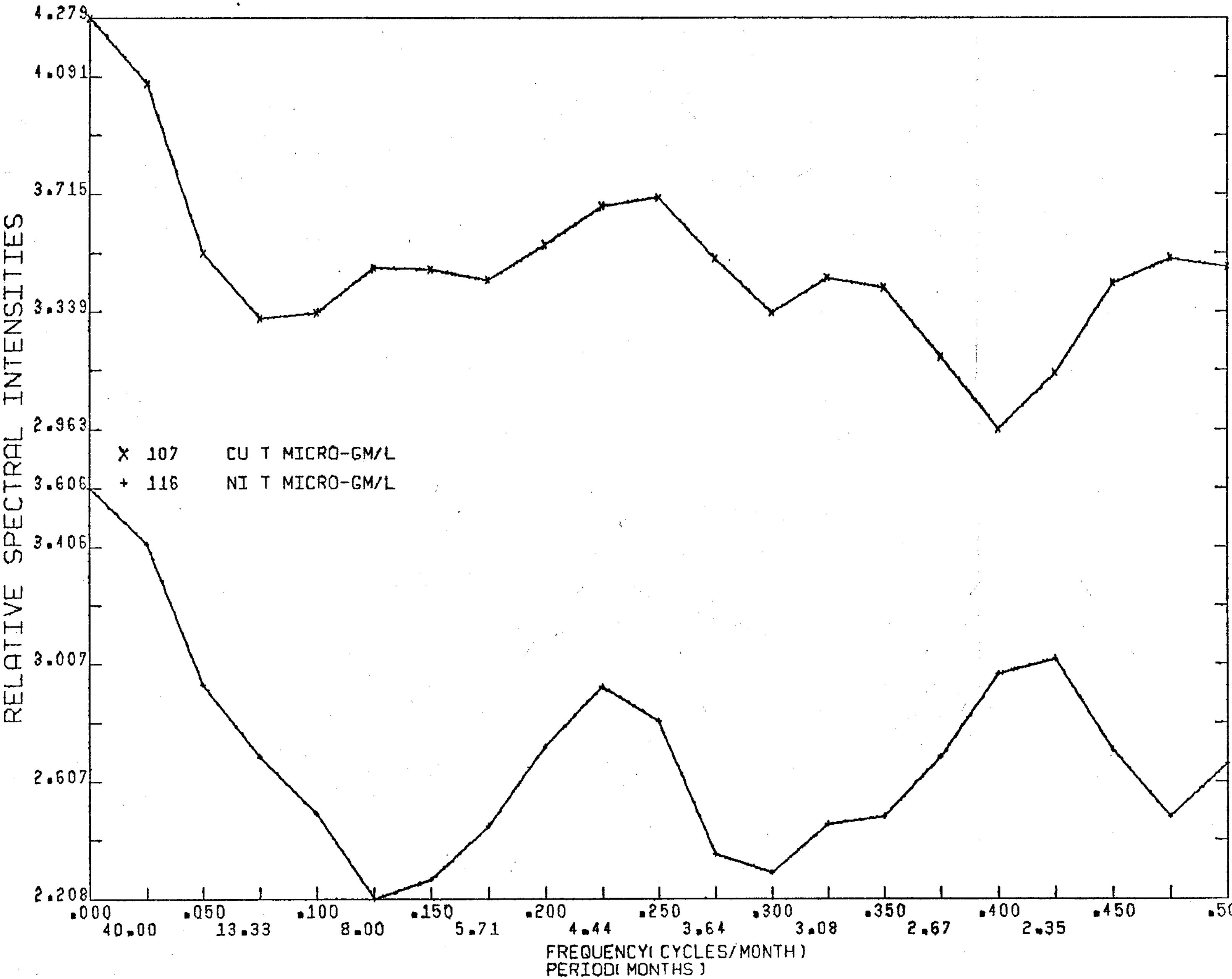


Figure 3.3.22: Power spectrum for Cu T & Ni T, station 4 (7204-7511)

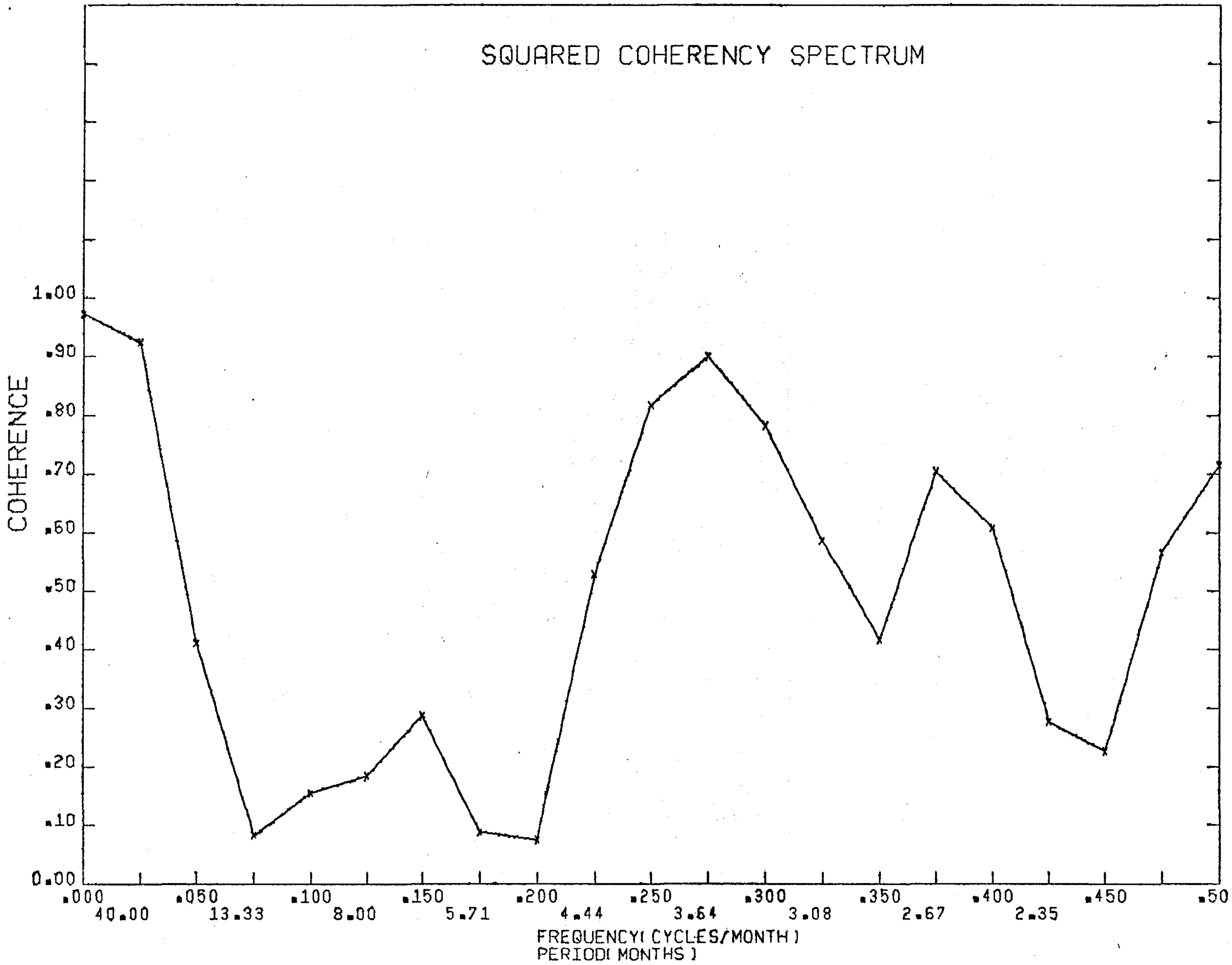


Figure 3.3.23: Coherency spectrum for Cu T & Ni T, station 4 (7204-7511)

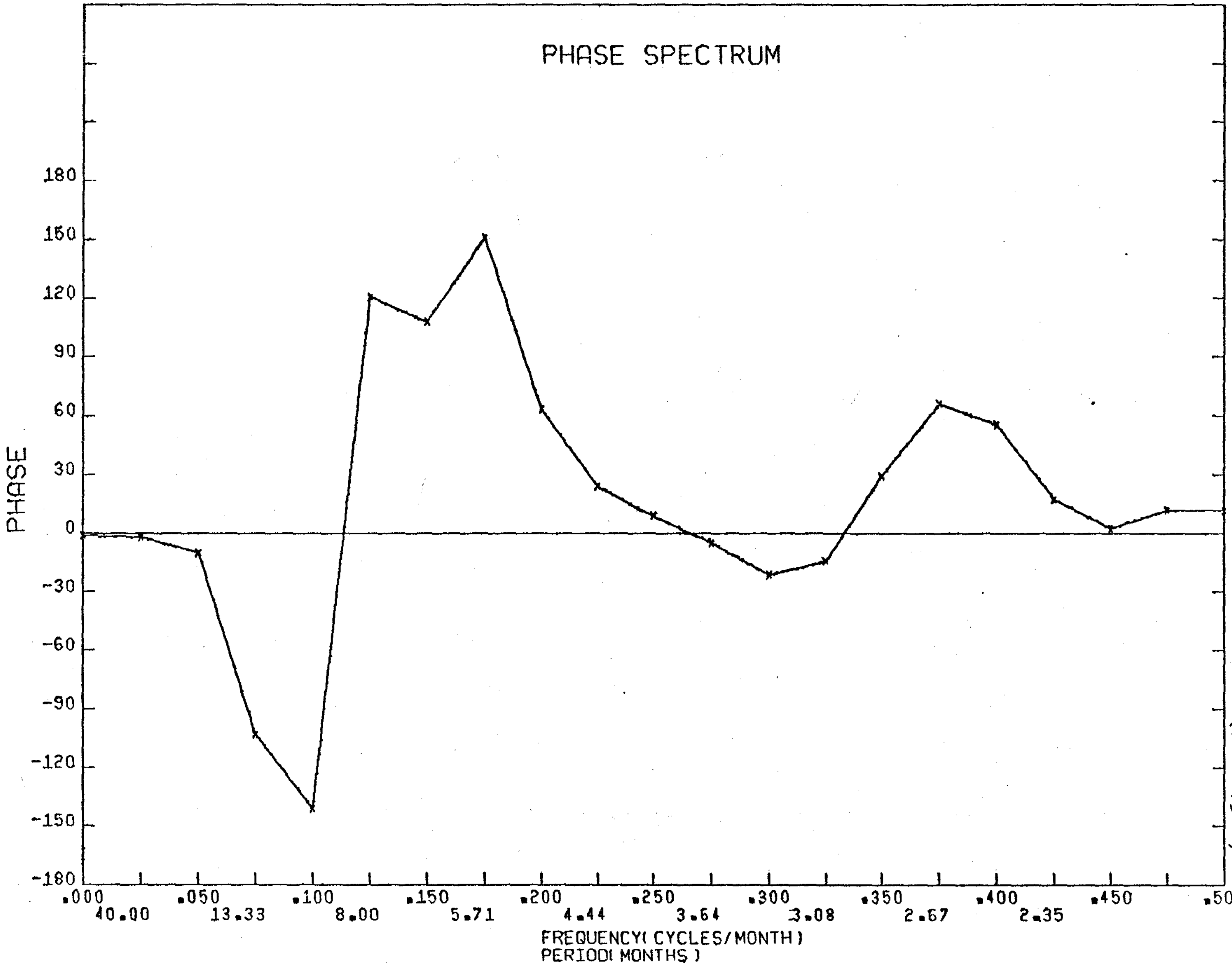


Figure 3.3.24: Phase spectrum for Cu T & Ni T, station 4 (7204-7511)

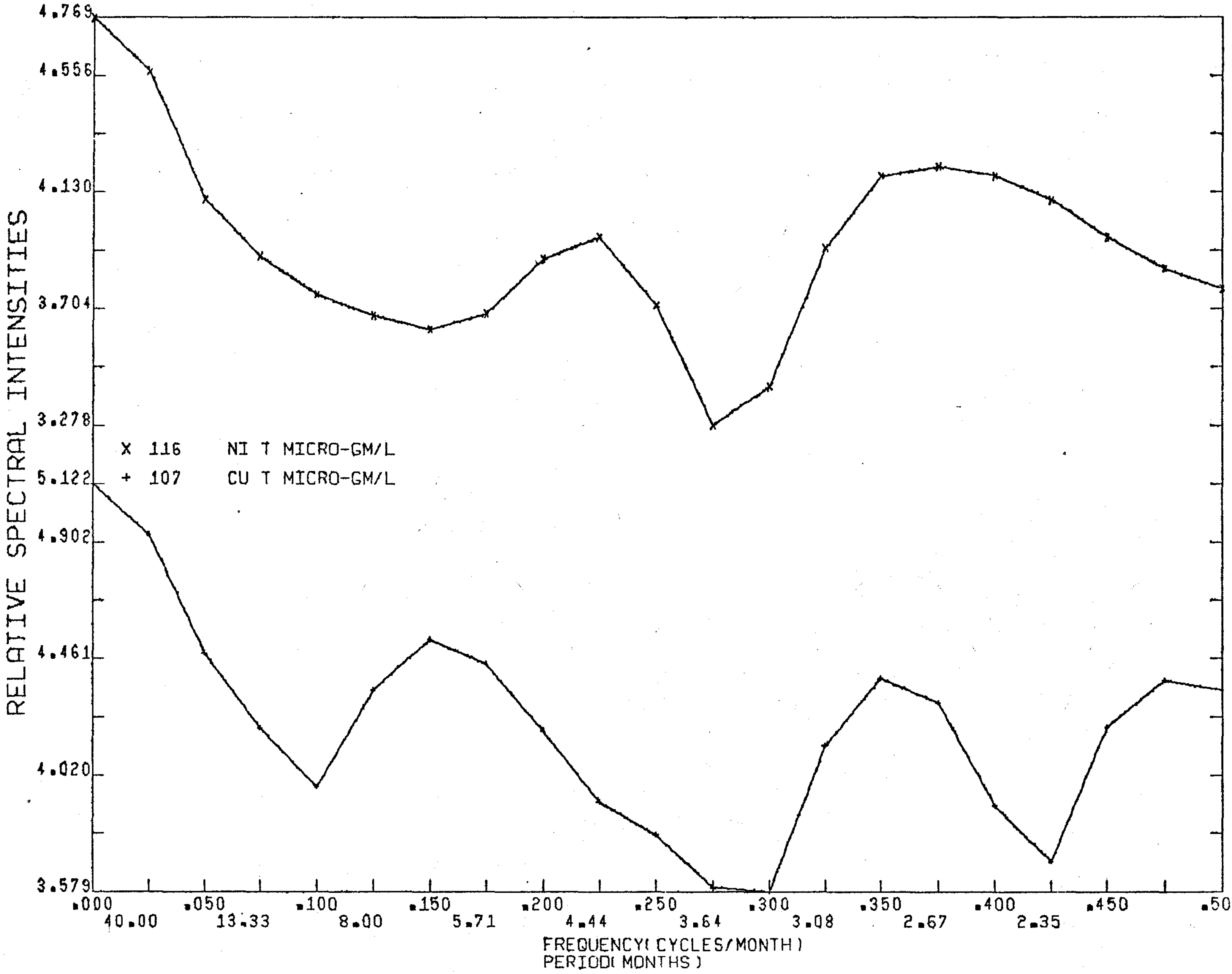


Figure 3.3.25: Power spectrum for Cu T & Ni T, Sudbury area (7208-7511)

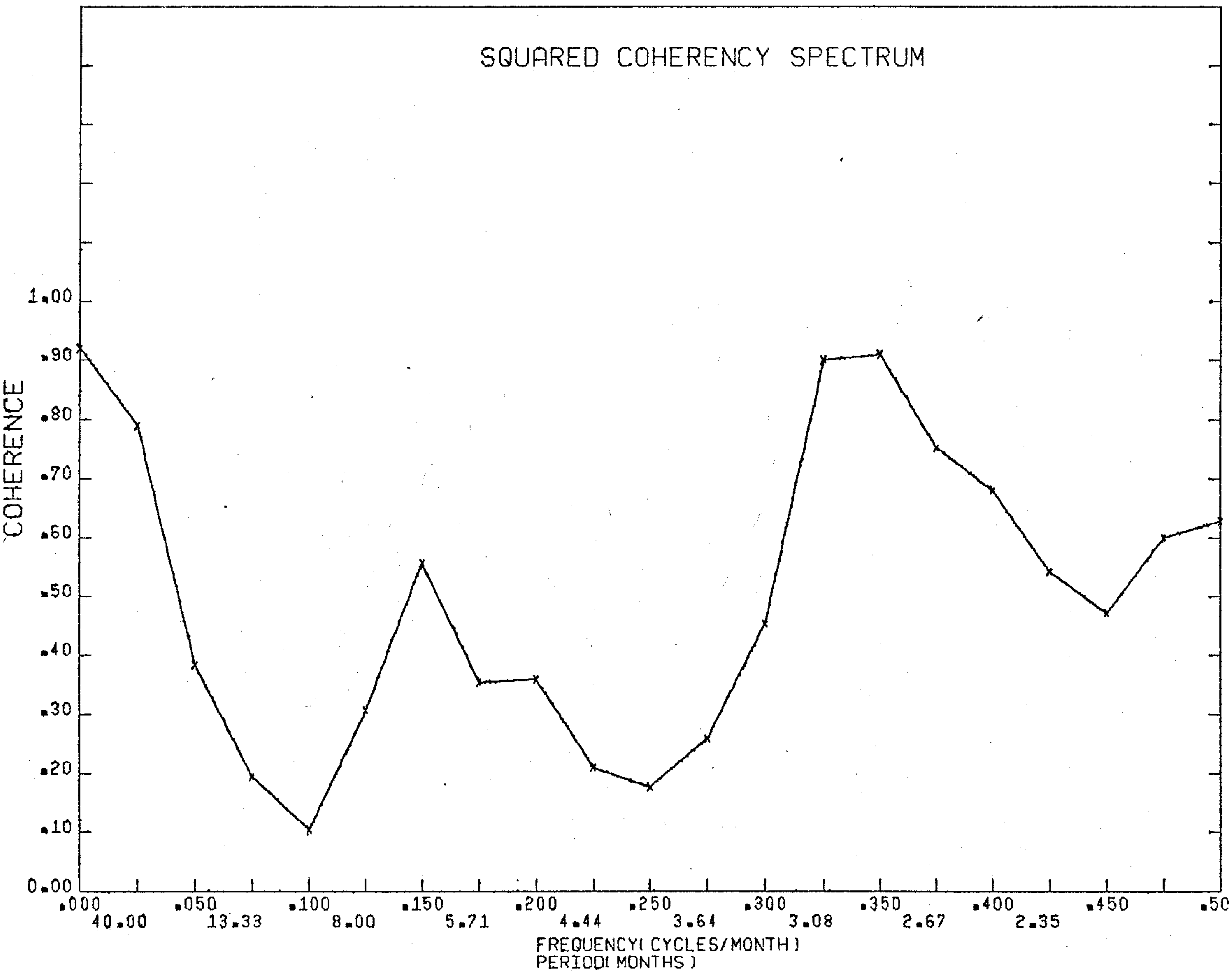


Figure 3.3.26: Coherency spectrum for Cu T & Ni T, Sudbury area (7208-7511)

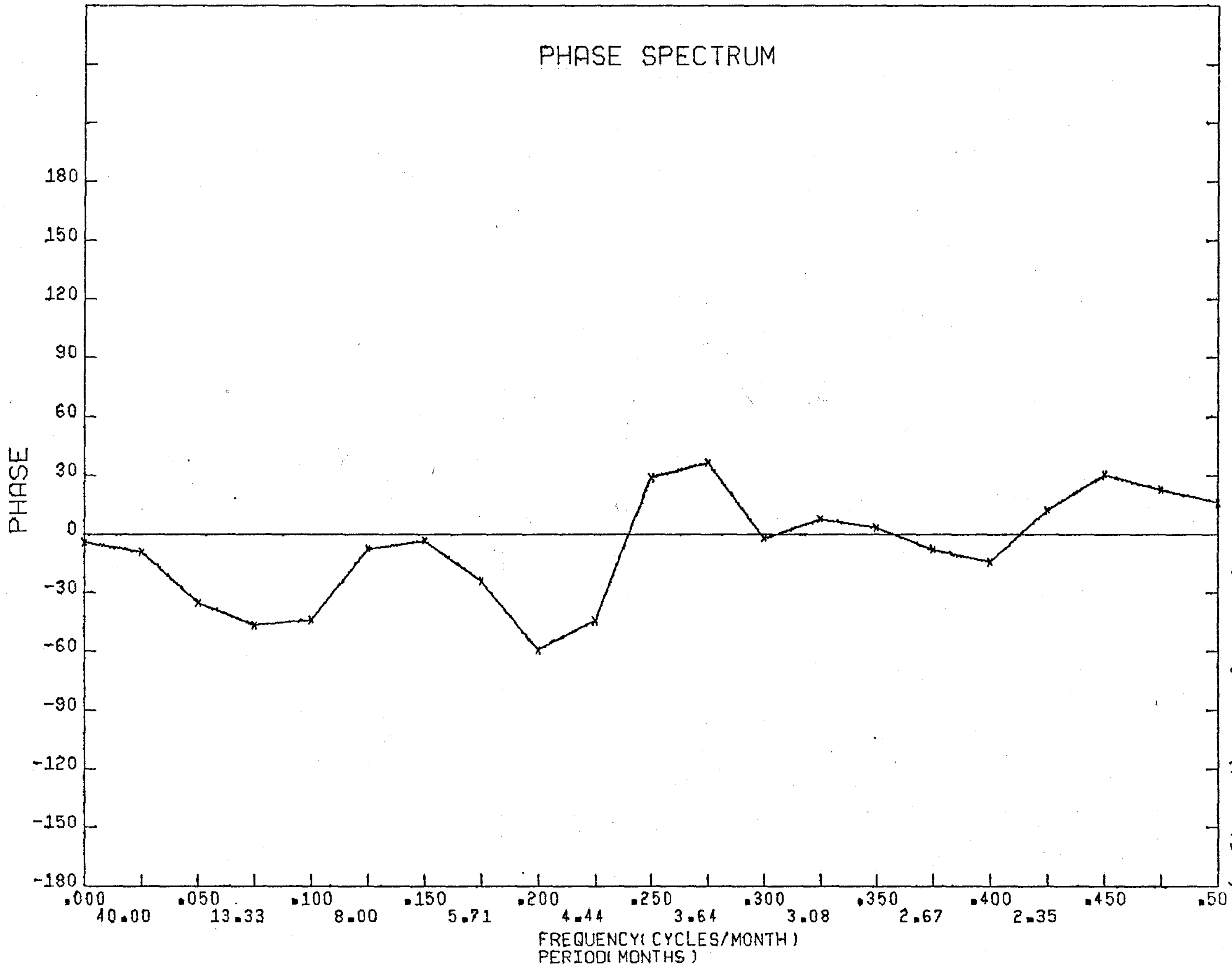


Figure 3.3.27: Phase spectrum for Cu T & Ni T, Sudbury area (7208-7511)

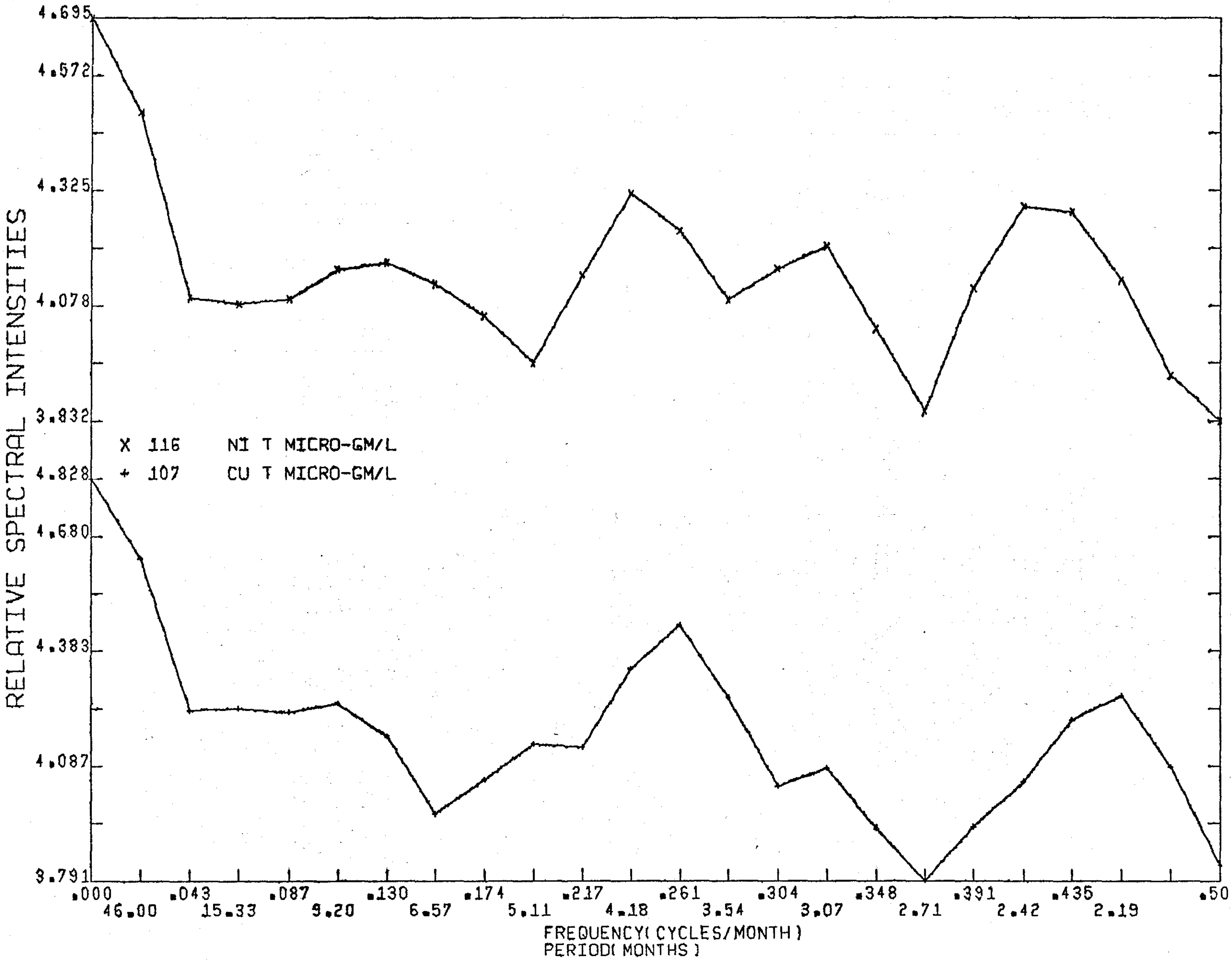


Figure 3.3.28: Power spectrum for Cu T & Ni T, U.G.L. area (7201-7511)

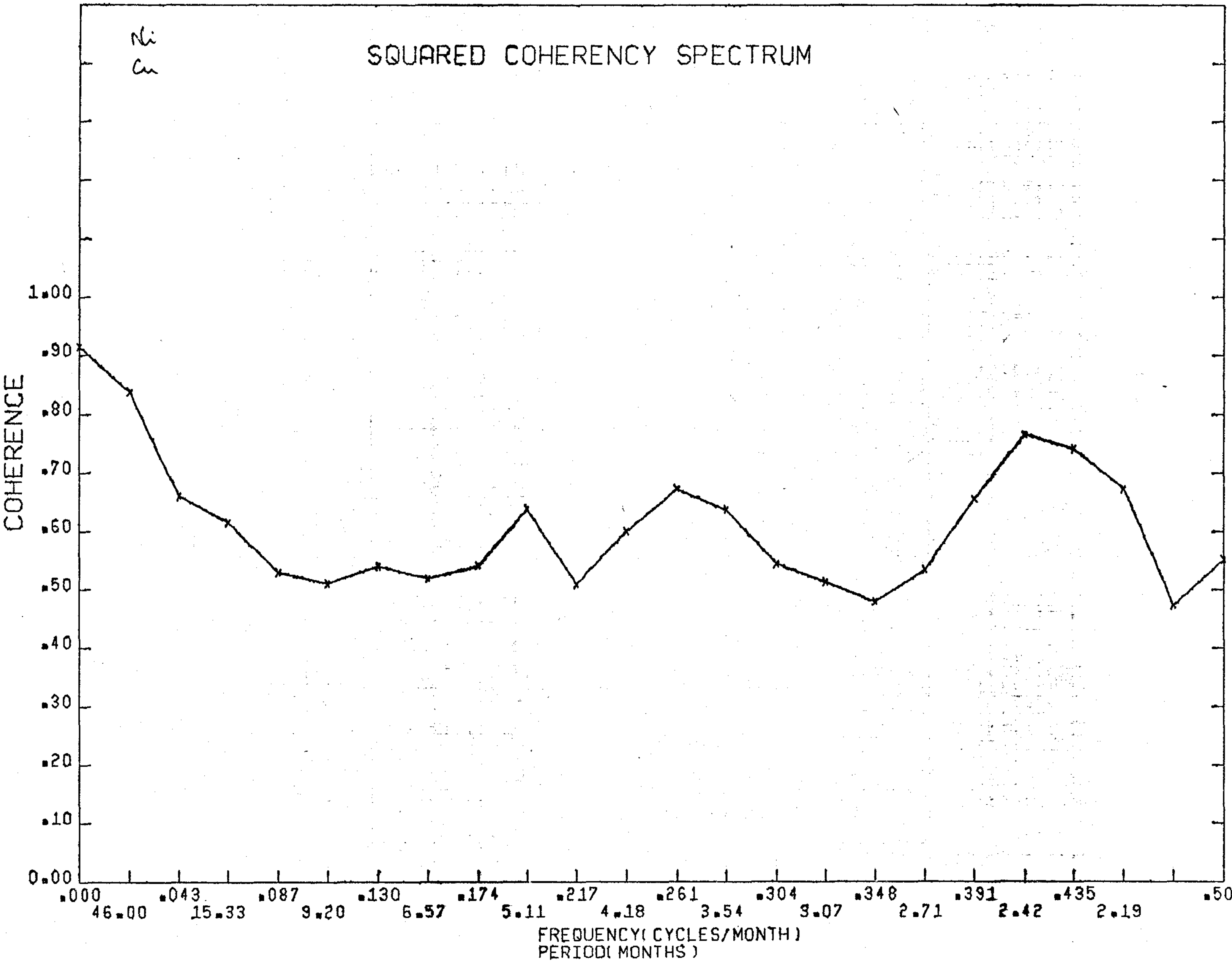


Figure 3.3.29: Coherency spectrum for Cu T & Ni T, U.G.L. area (7201-7511)

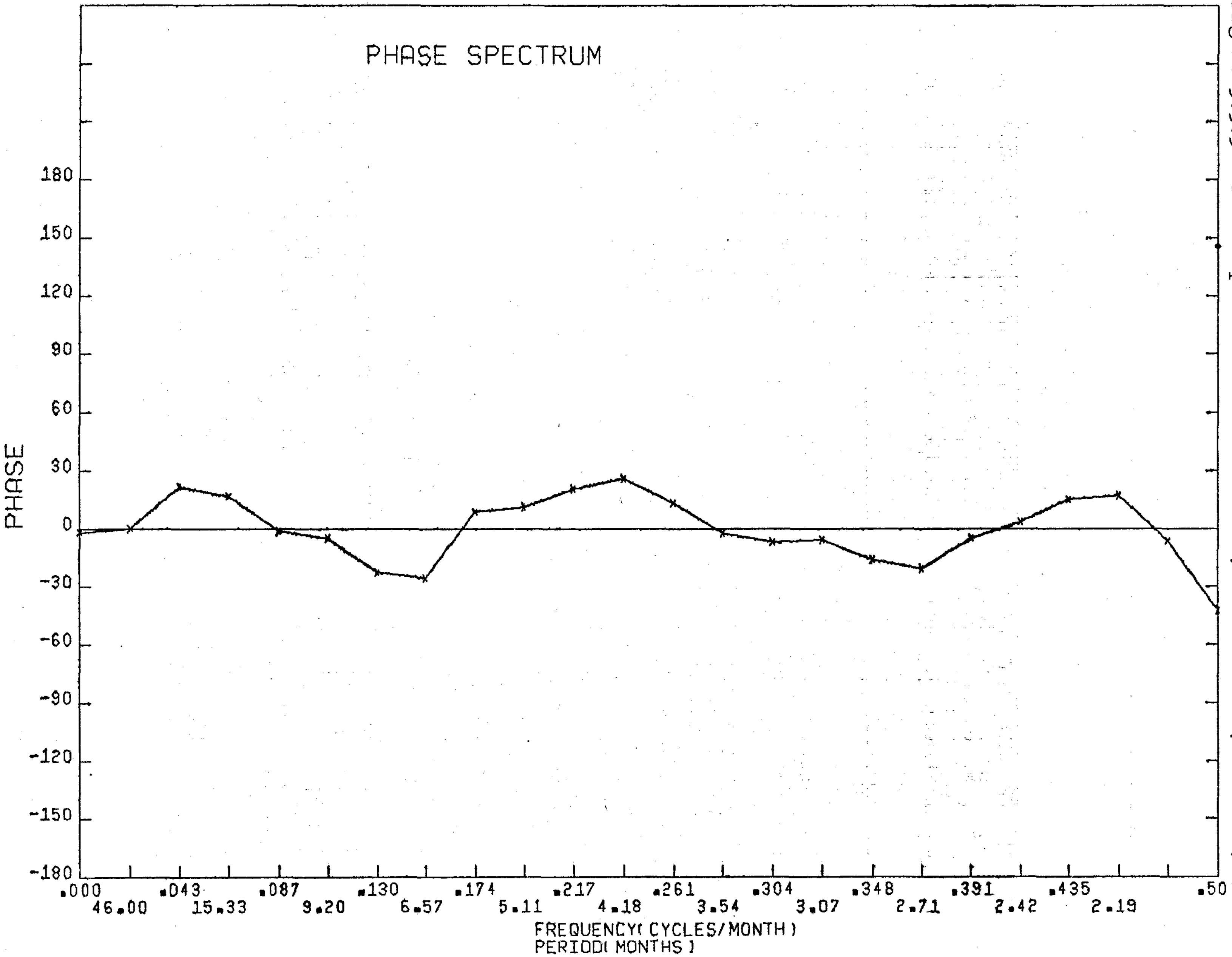


Figure 3.3.30: Phase spectrum for Cu T & Ni T, U.G.L. area (7201-7511)

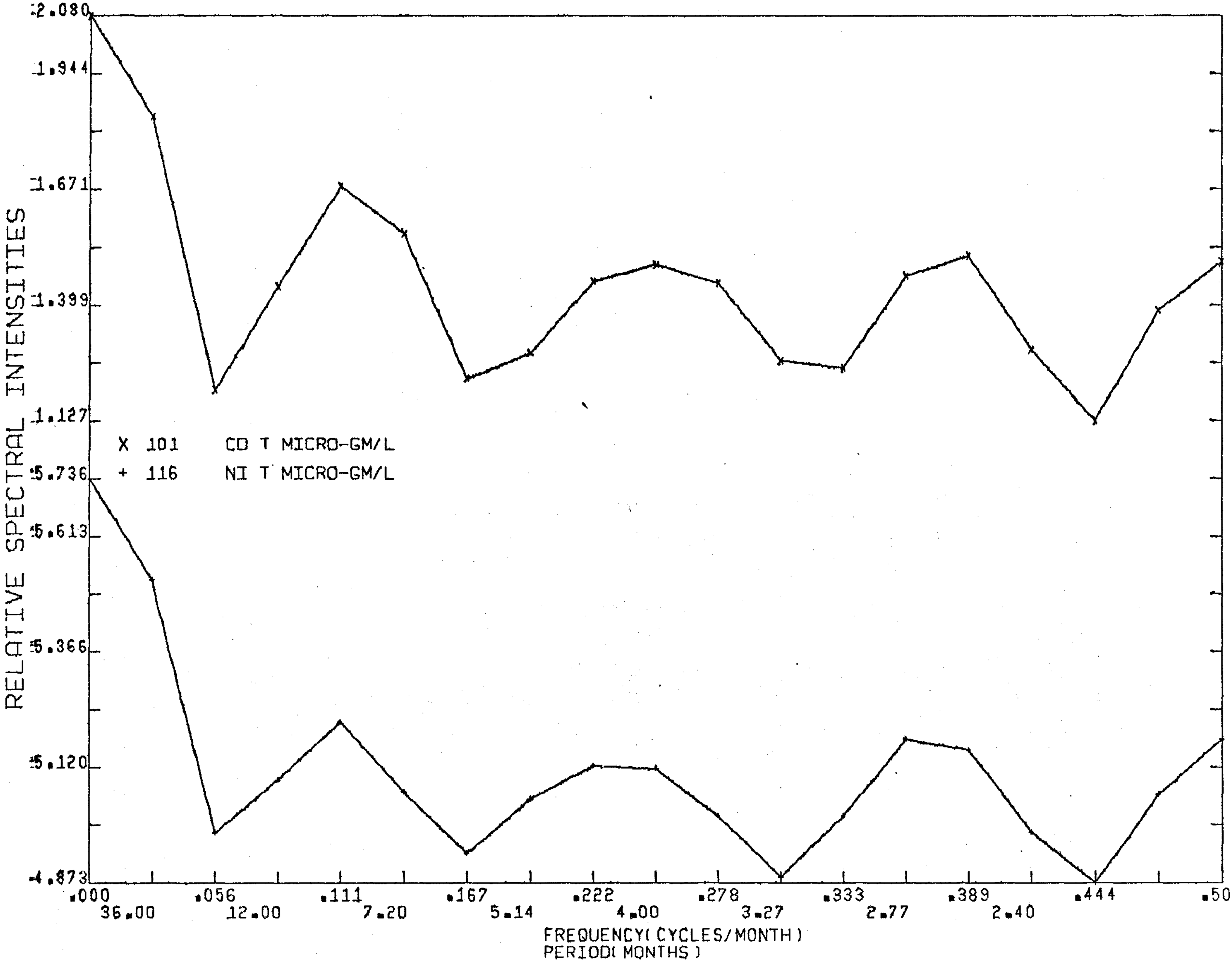


Figure 3.3.31: Power spectrum for Cd T & Ni T, station 12 (7210-7511)

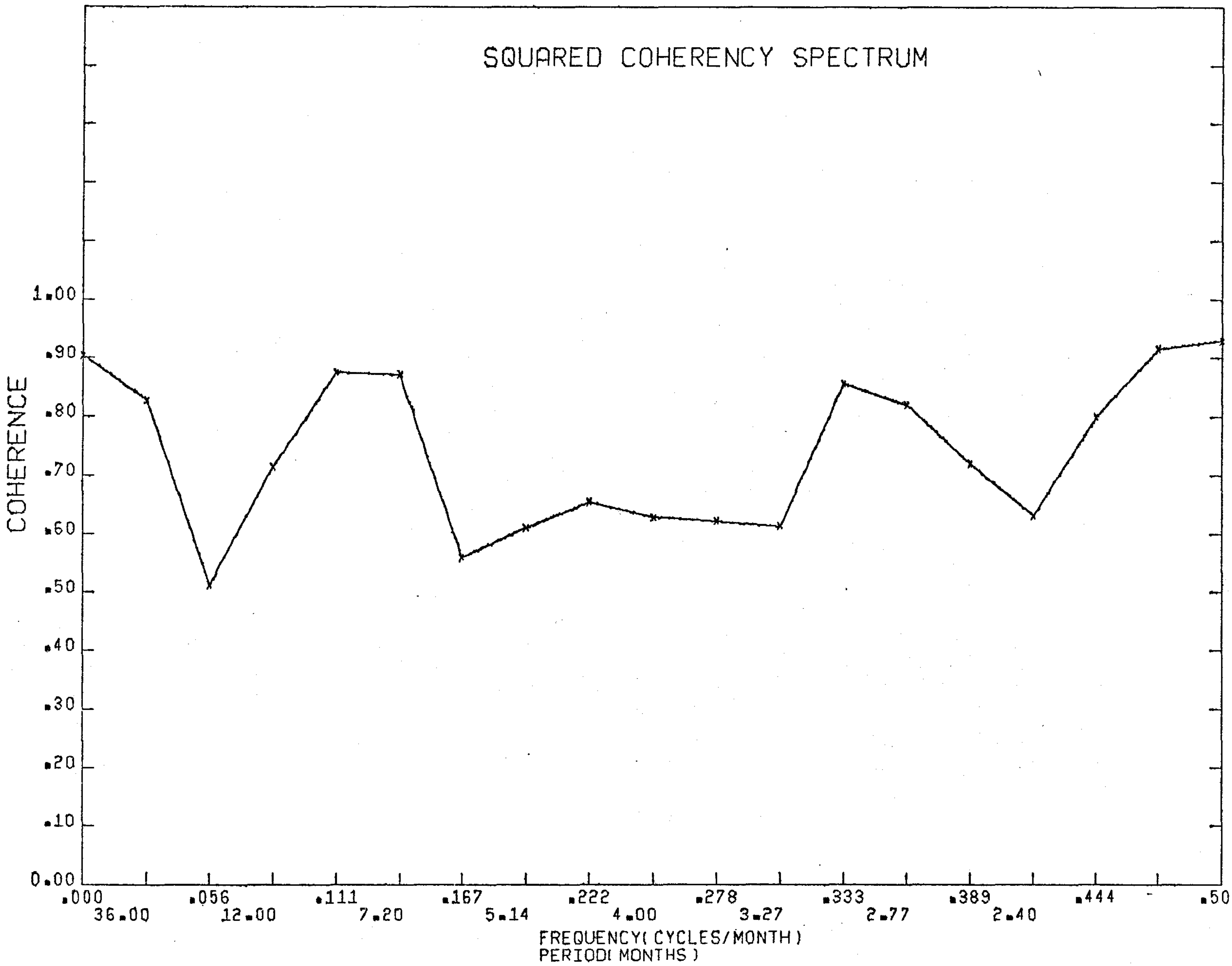


Figure 3.3.32: Coherency spectrum for Cd T & Ni T, station 12 (7210-7511)

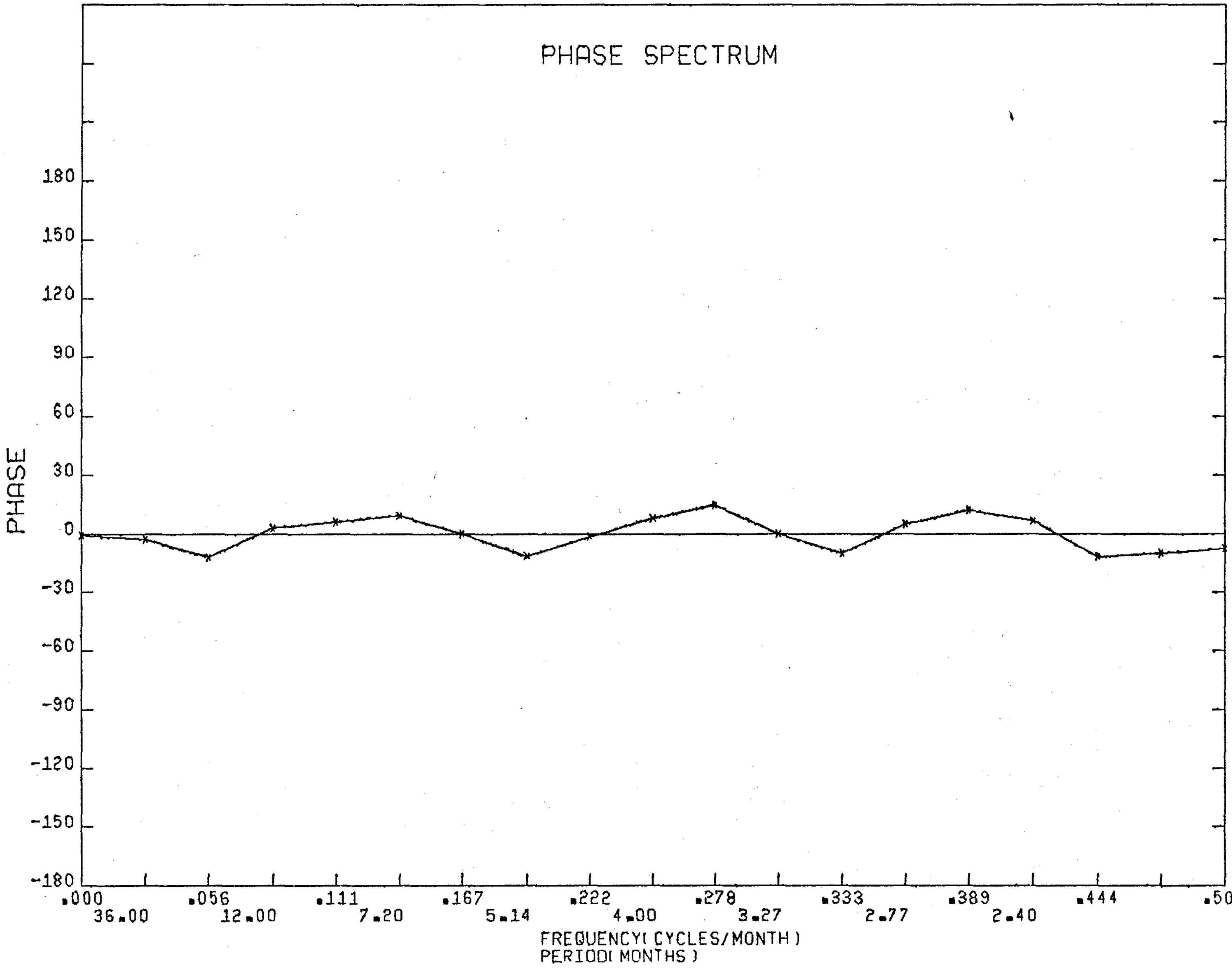


Figure 3.3.33: Phase spectrum for Cd T & Ni T, station 12 (7210-7511)

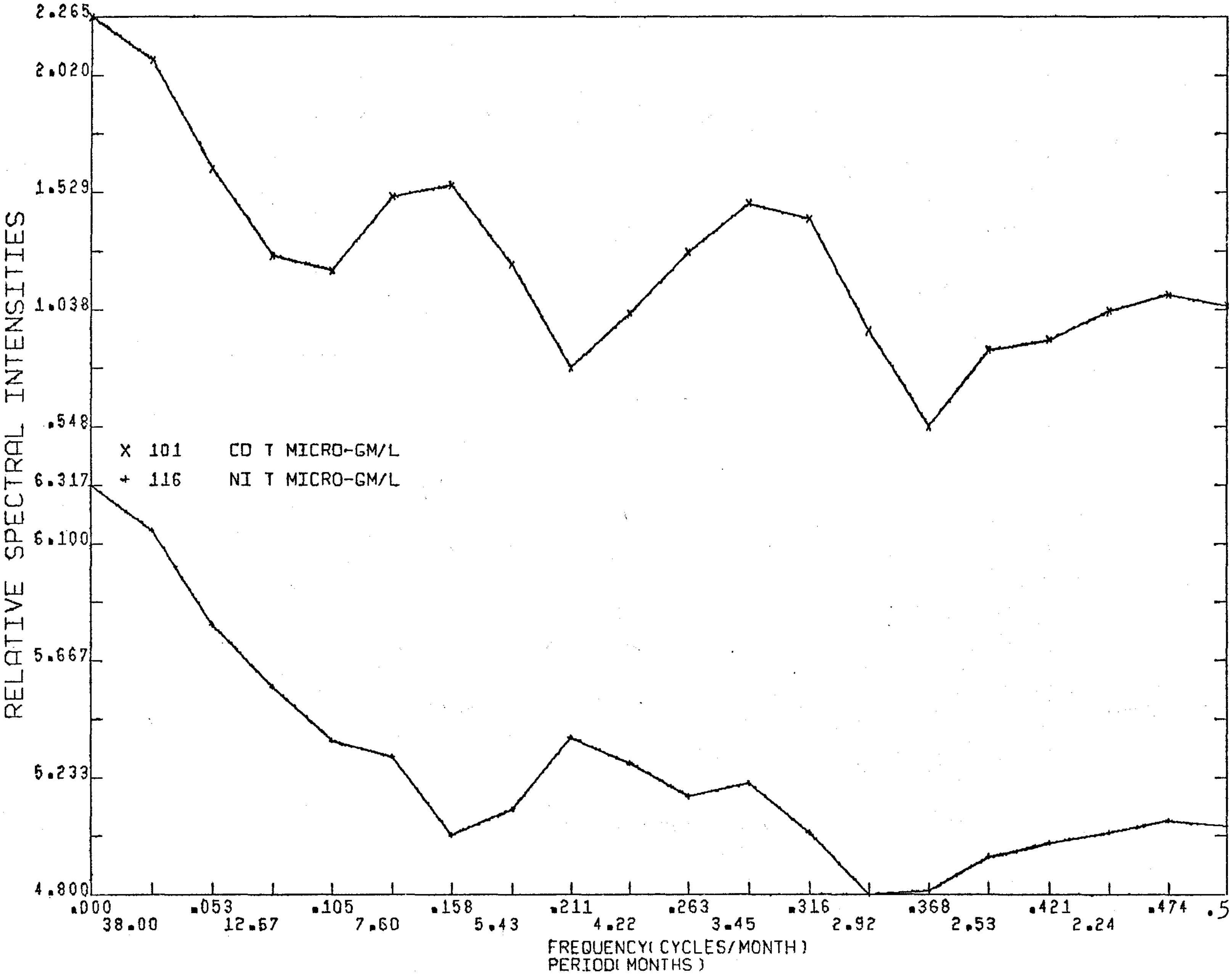


Figure 3.3.34: Power spectrum for Cd T & Ni T, station 2 (7206-7510)

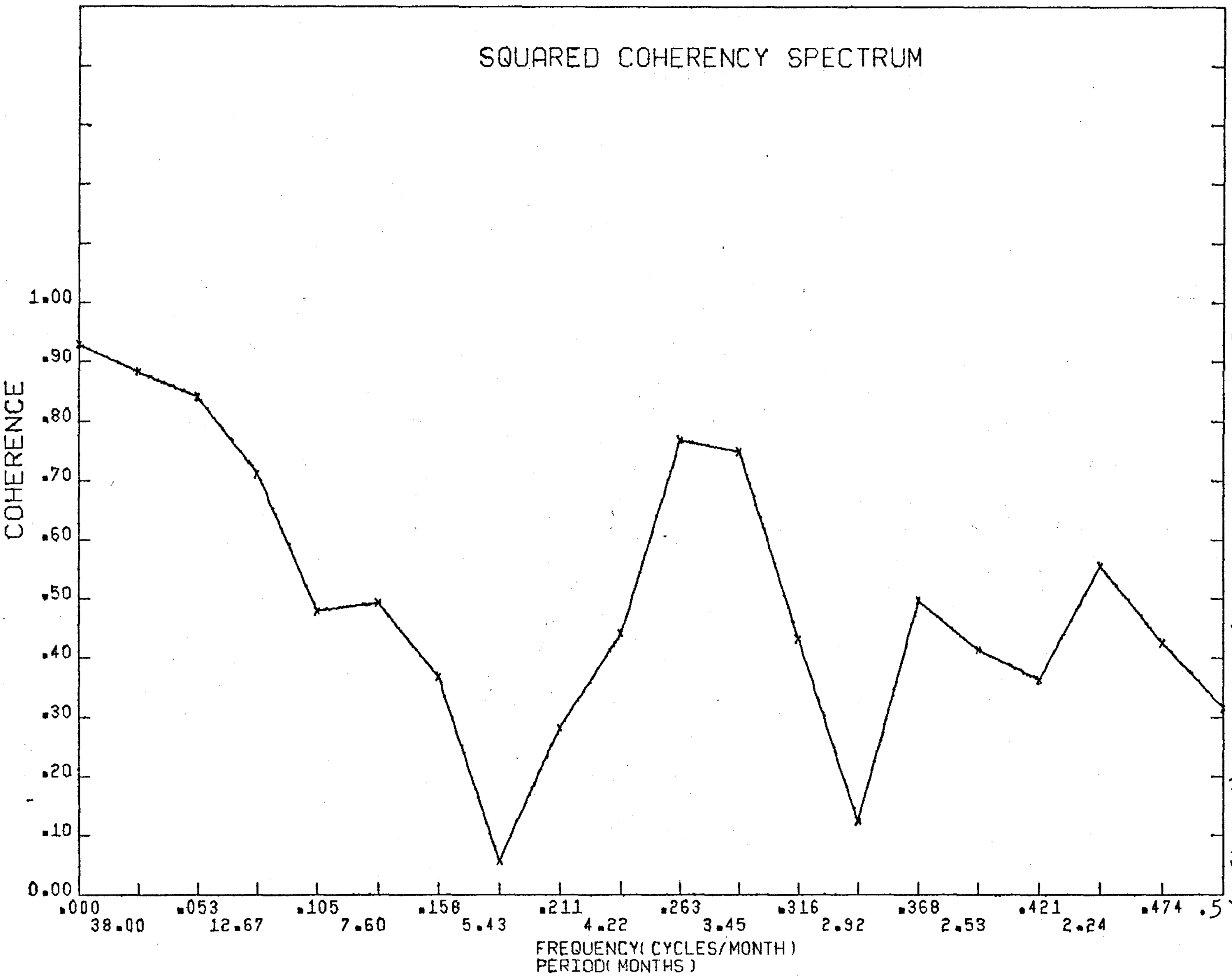


Figure 3.3.35: Coherency spectrum for Cd T & Ni T, station 2 (7206-7511)

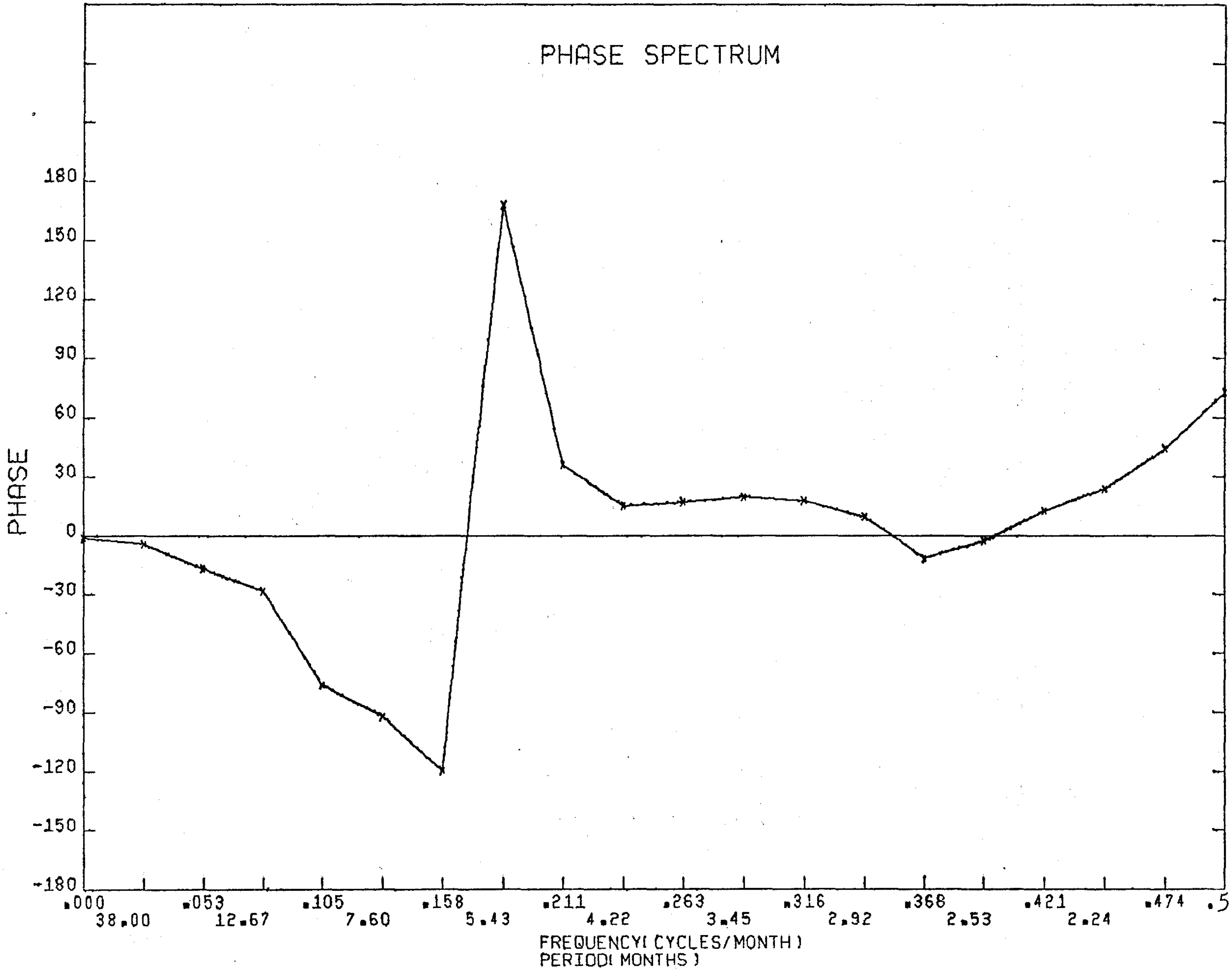


Figure 3.3.36: Phase spectrum for Cd T & Ni T, station 2 (7206-7511)

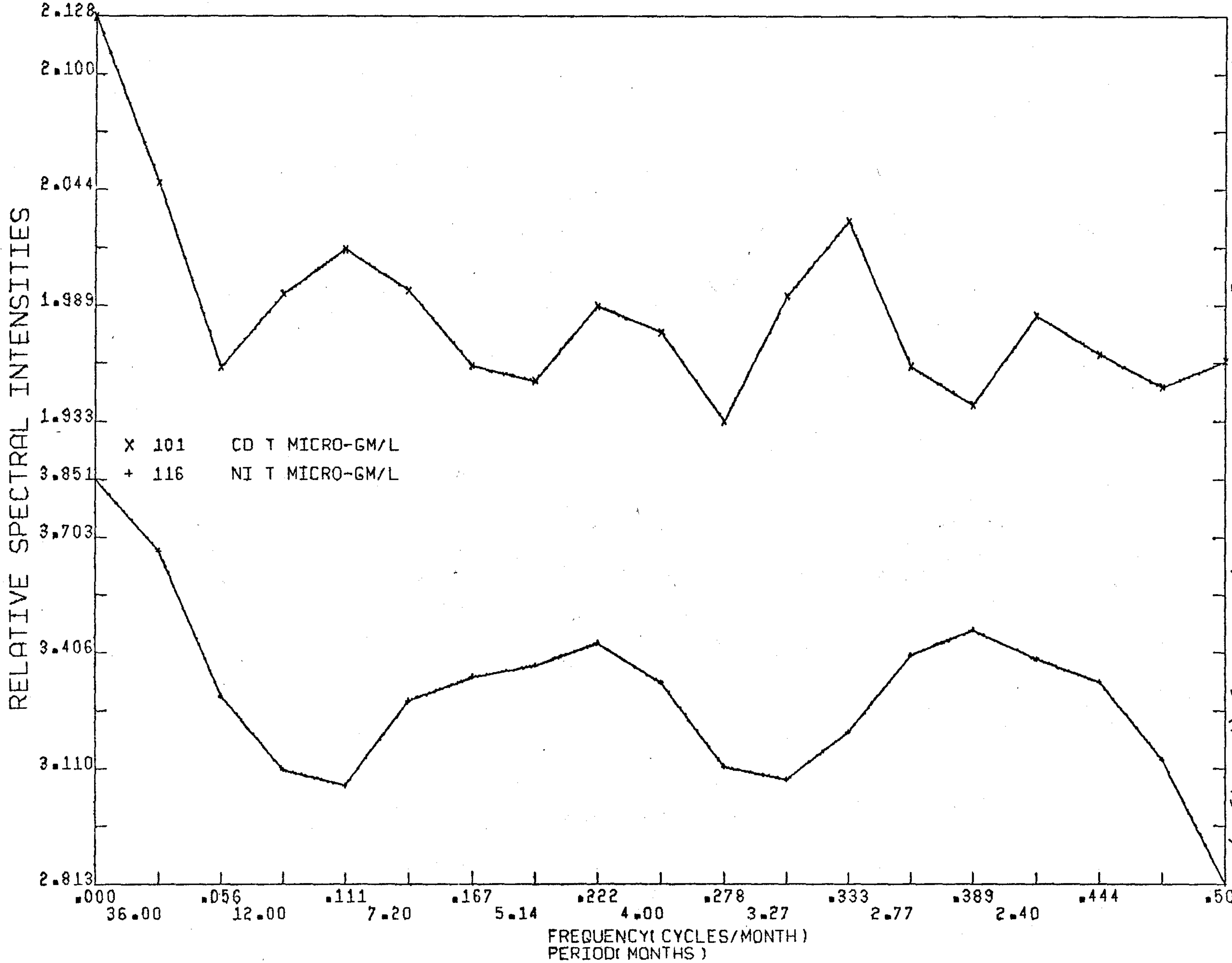


Figure 3.3.37: Power spectrum for Cd T & Ni T, station 3 (7211-7511)

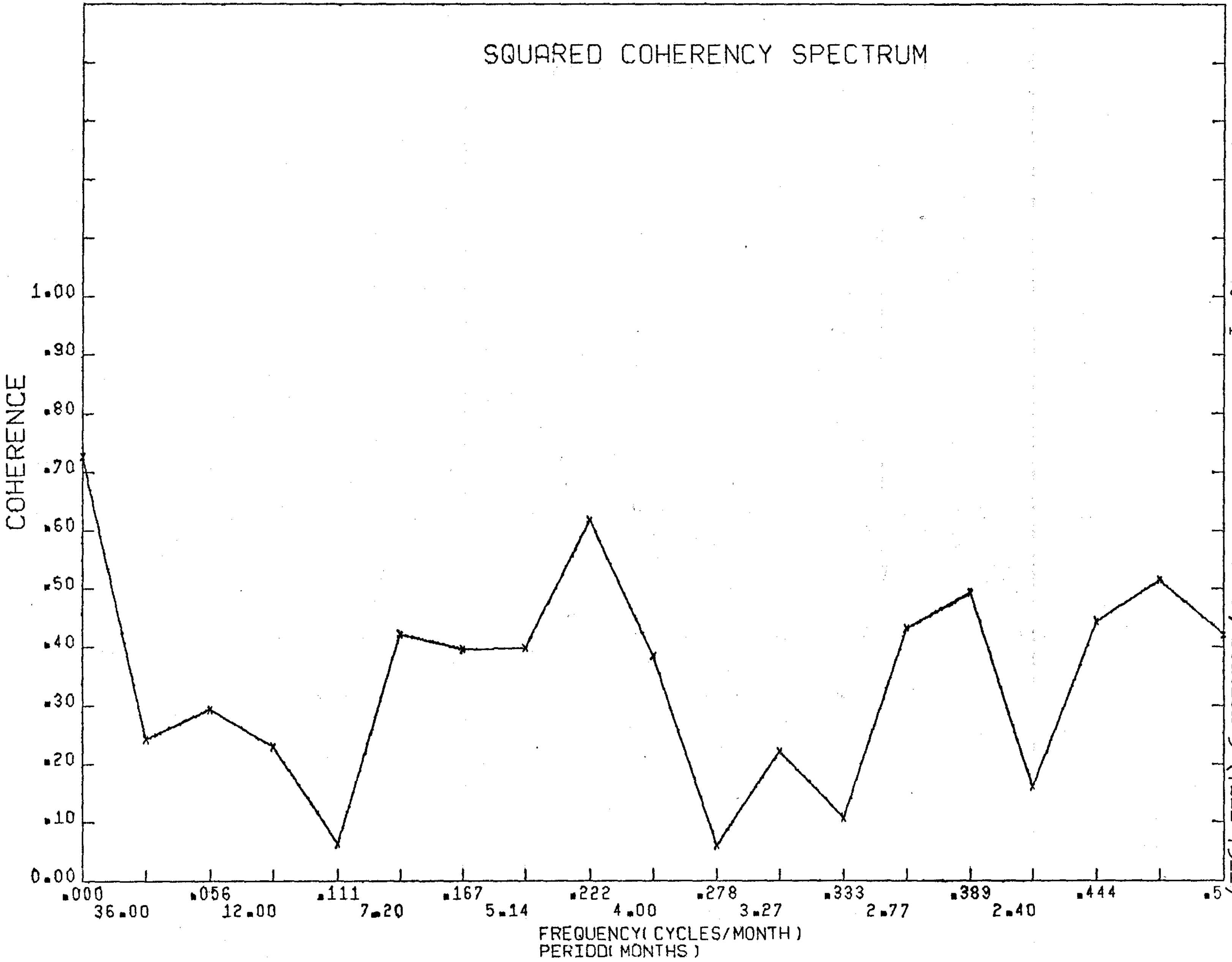


Figure 3.3.38: Coherency spectrum for Cd T & Ni T, station 3 (7211-7511)

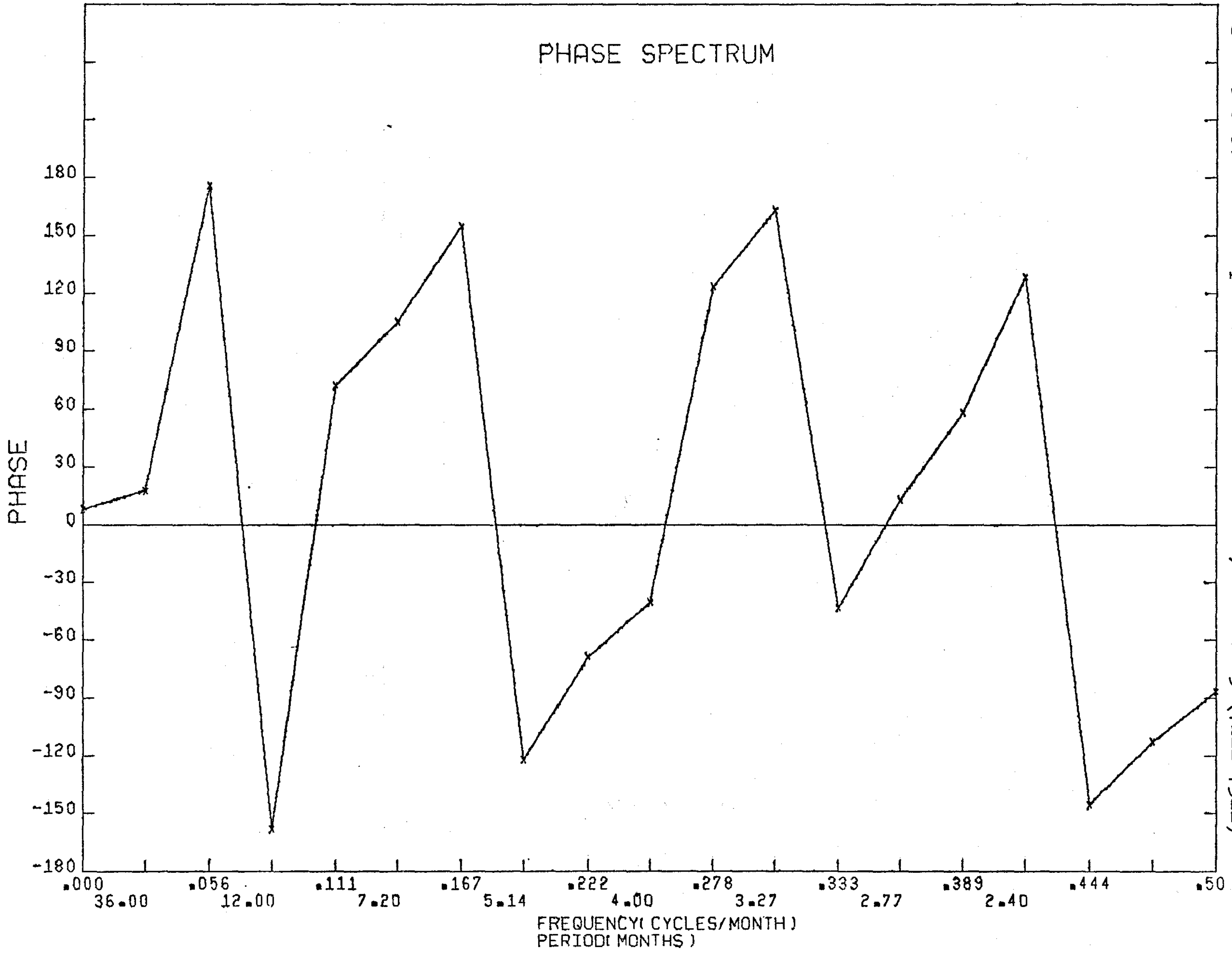


Figure 3.3.39: Phase spectrum for Cd T & Ni T, station 3 (7211-7511)

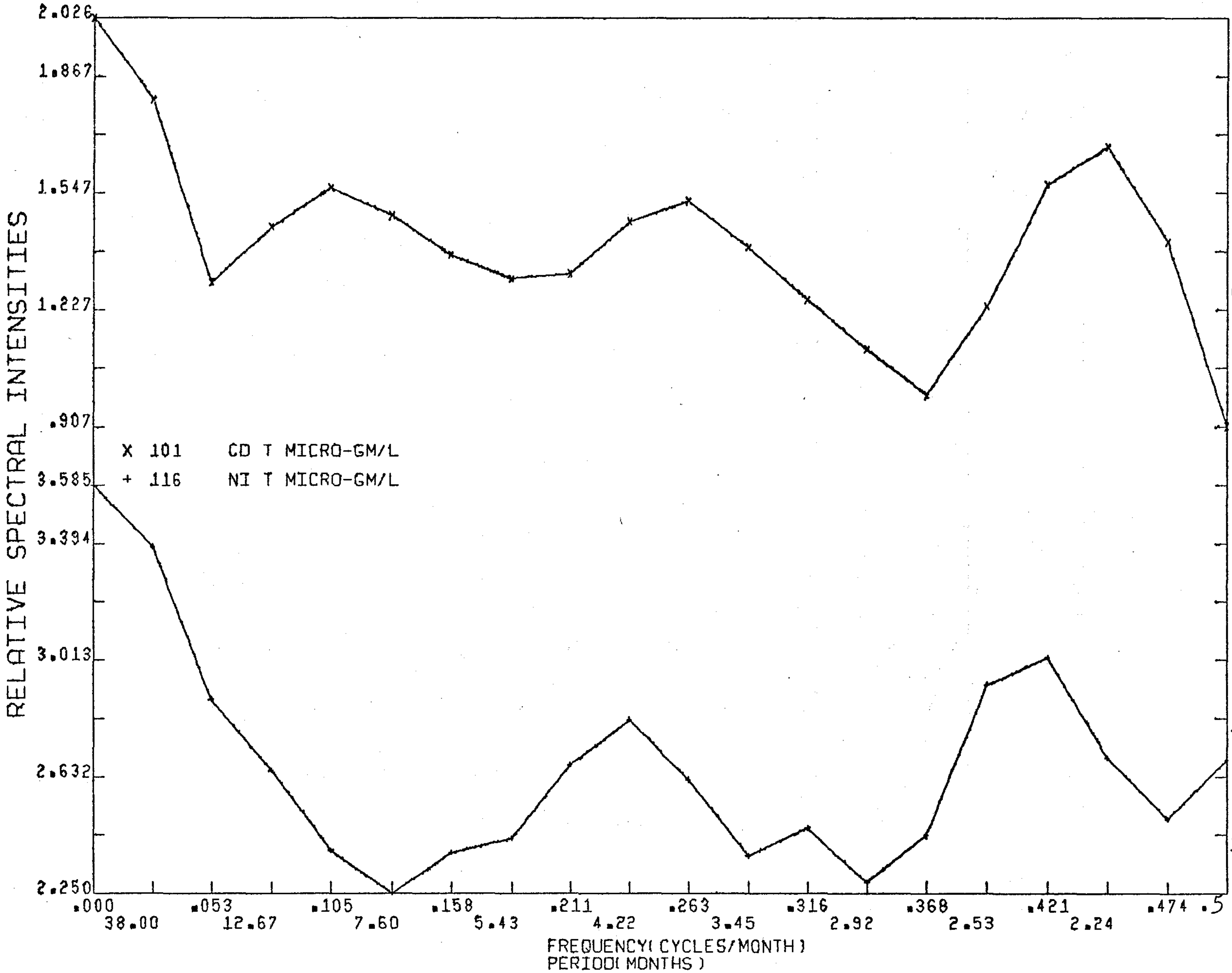


Figure 3.3.40: Power spectrum for Cd T & Ni T, station 4 (7204-7507)

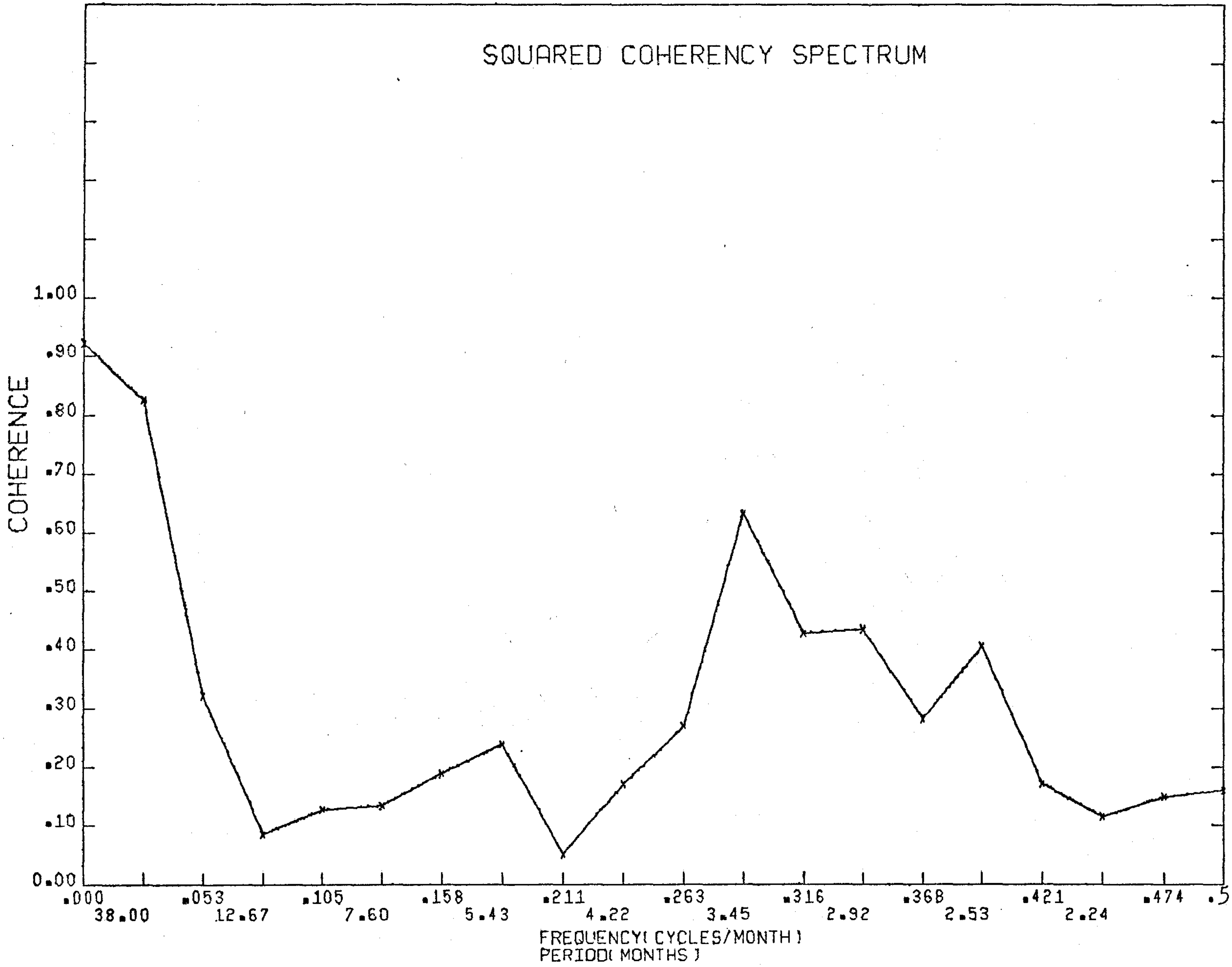


Figure 3.3.41: Coherency spectrum for Cd T & Ni T, station 4 (7204-7507)

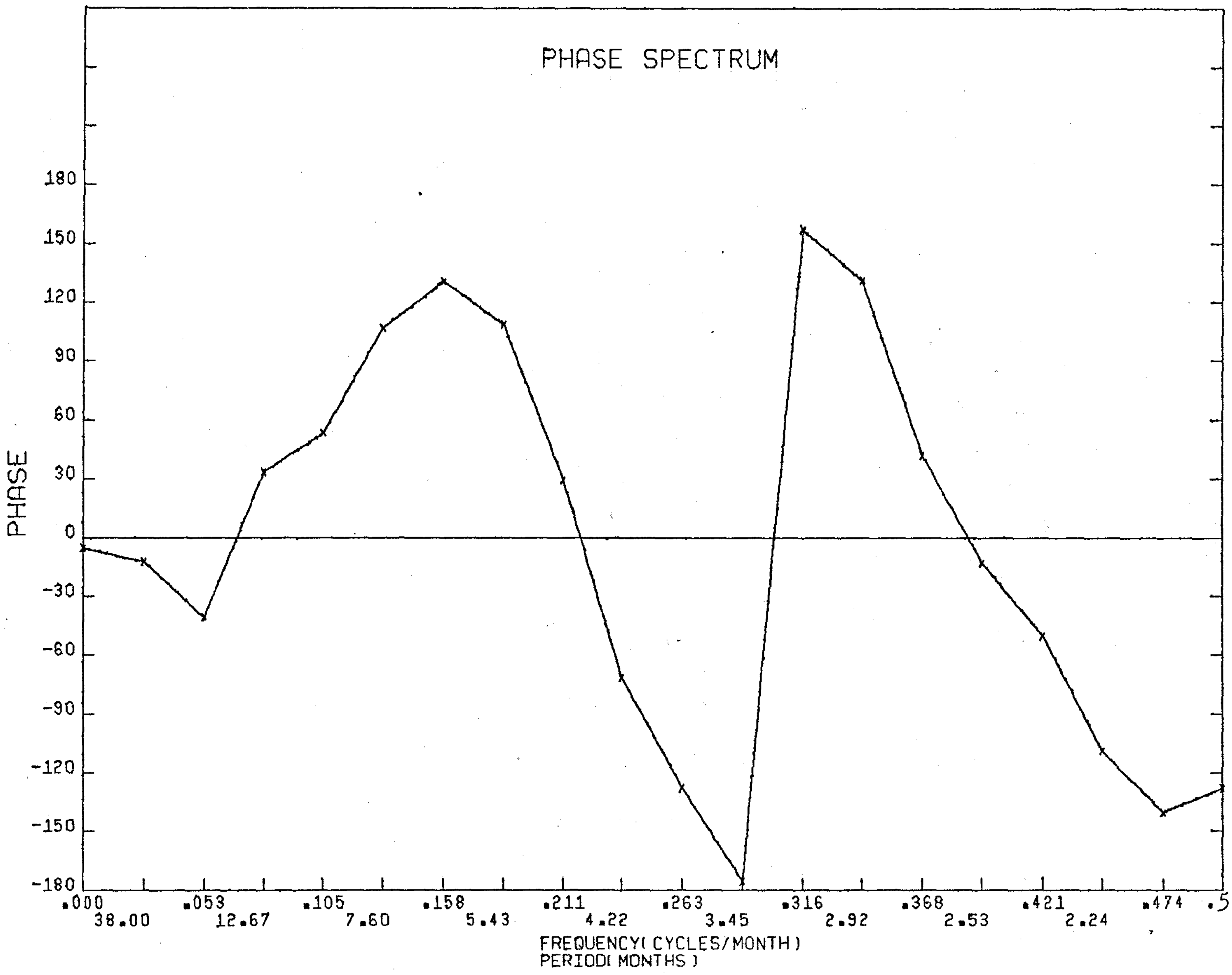


Figure 3.3.42: Phase spectrum for Cd T & Ni T, station 4 (7204-7507)

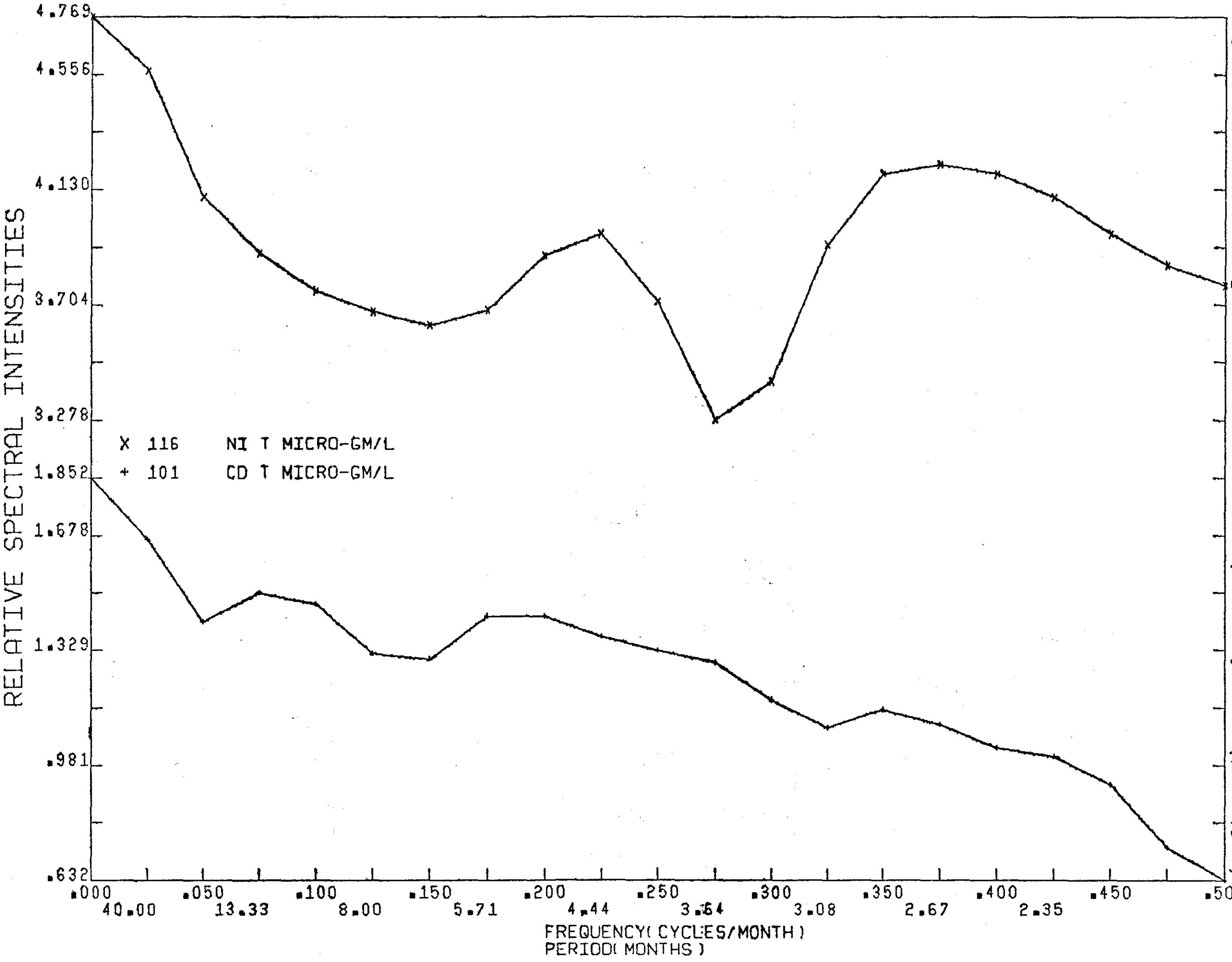


Figure 3.3.43: Power spectrum for Cd T & Ni T, Sudbury area (7208-7511)

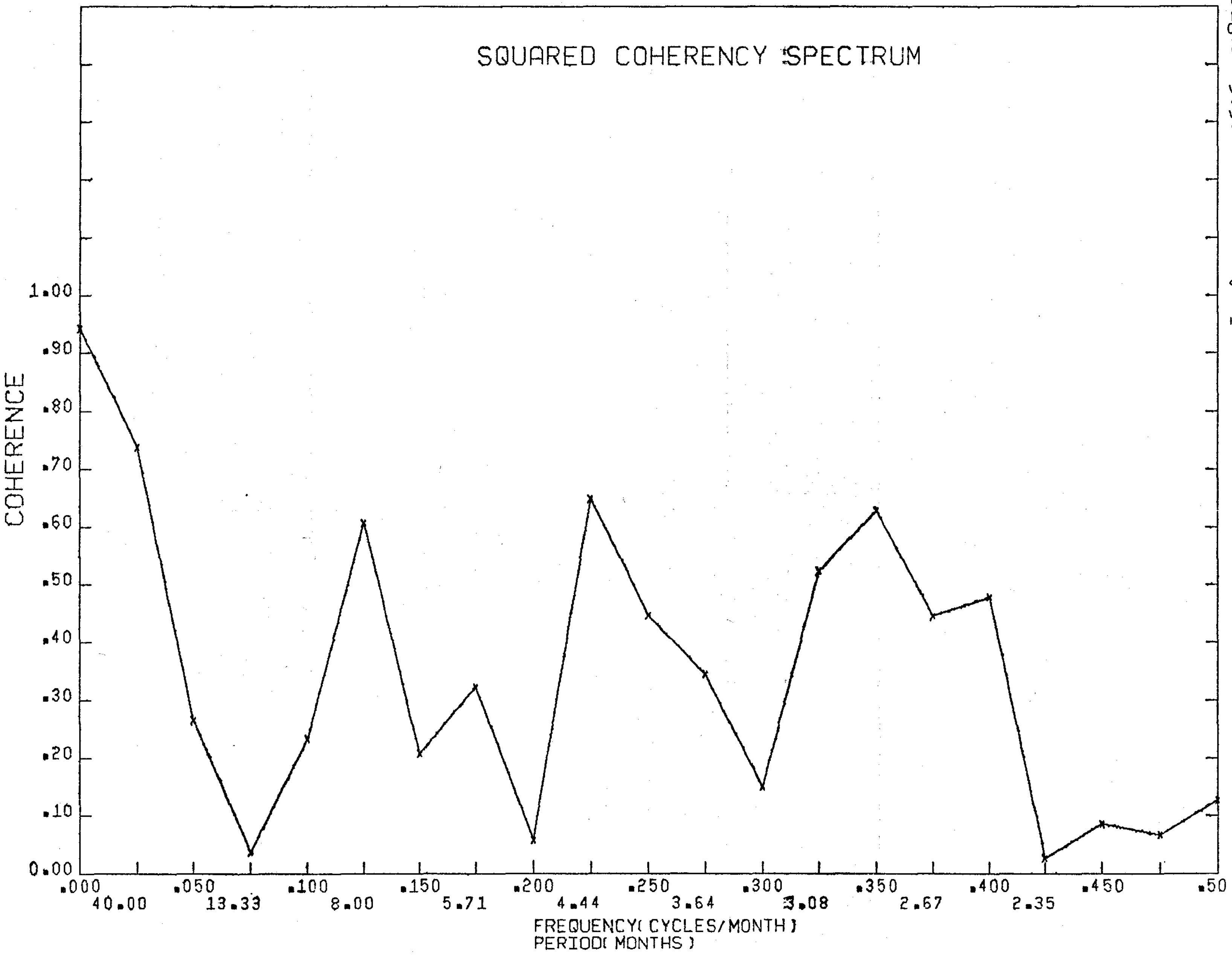


Figure 3.3.44: Coherency spectrum for Cd T & Ni T, Sudbury area (7208-7511)

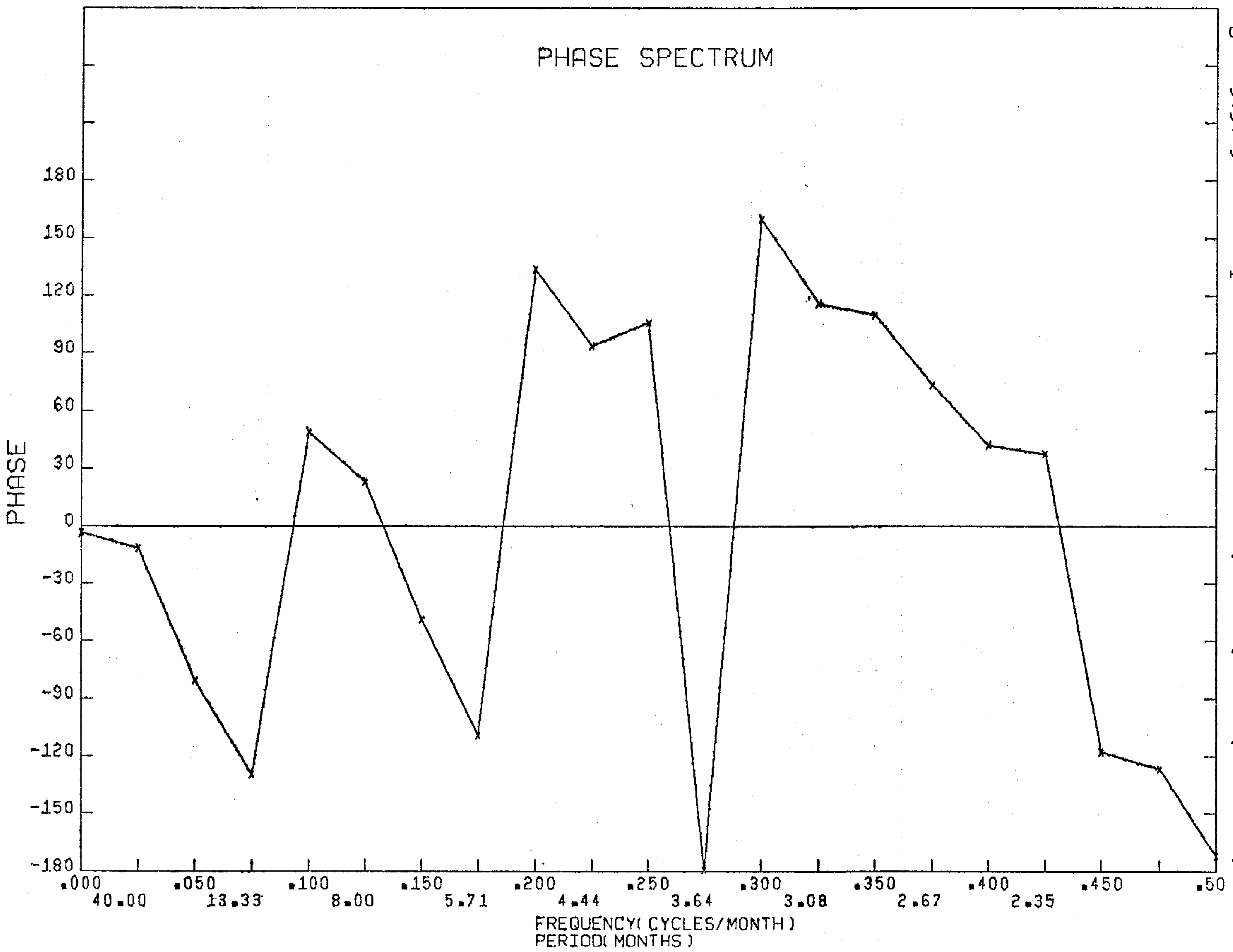


Figure 3.3.45: Phase spectrum for Cd T & Ni T, Sudbury area (7208-7511)

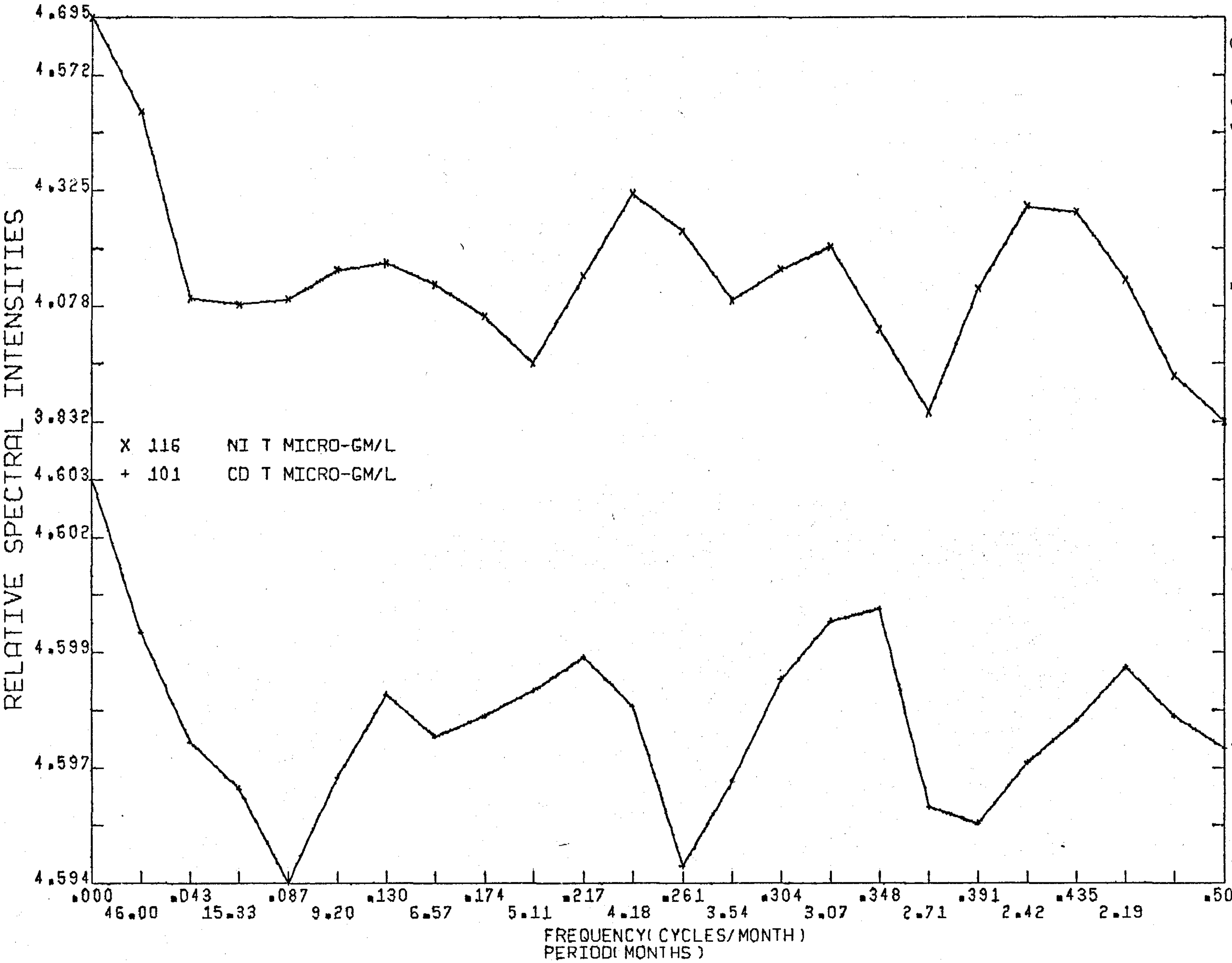


Figure 3.3.46: Power spectrum for Cd T & Ni T, U.G.L. area (7201-7511)

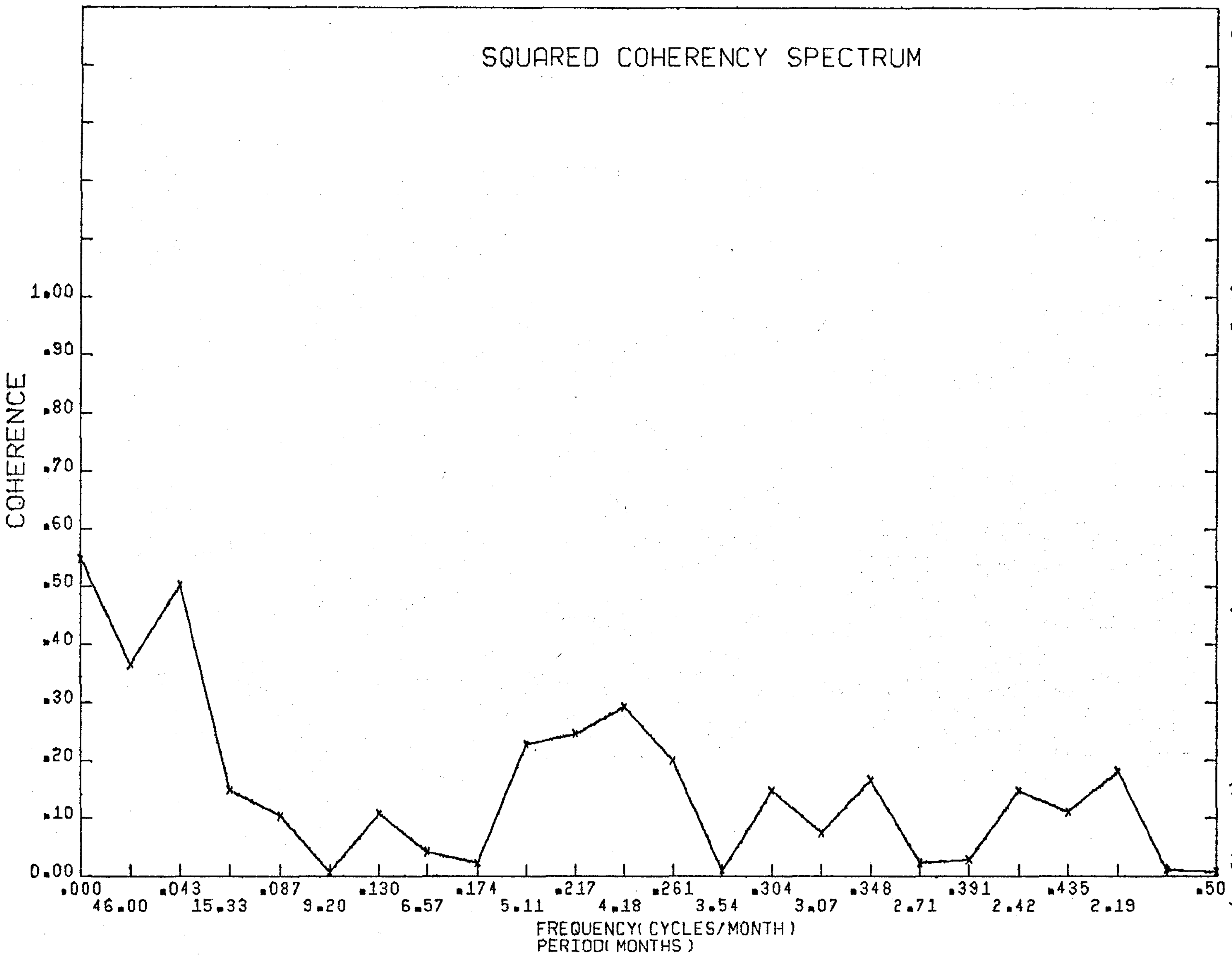


Figure 3.3.47: Coherency spectrum for Cd T & Ni T, U.G.L. area (7201-7511)

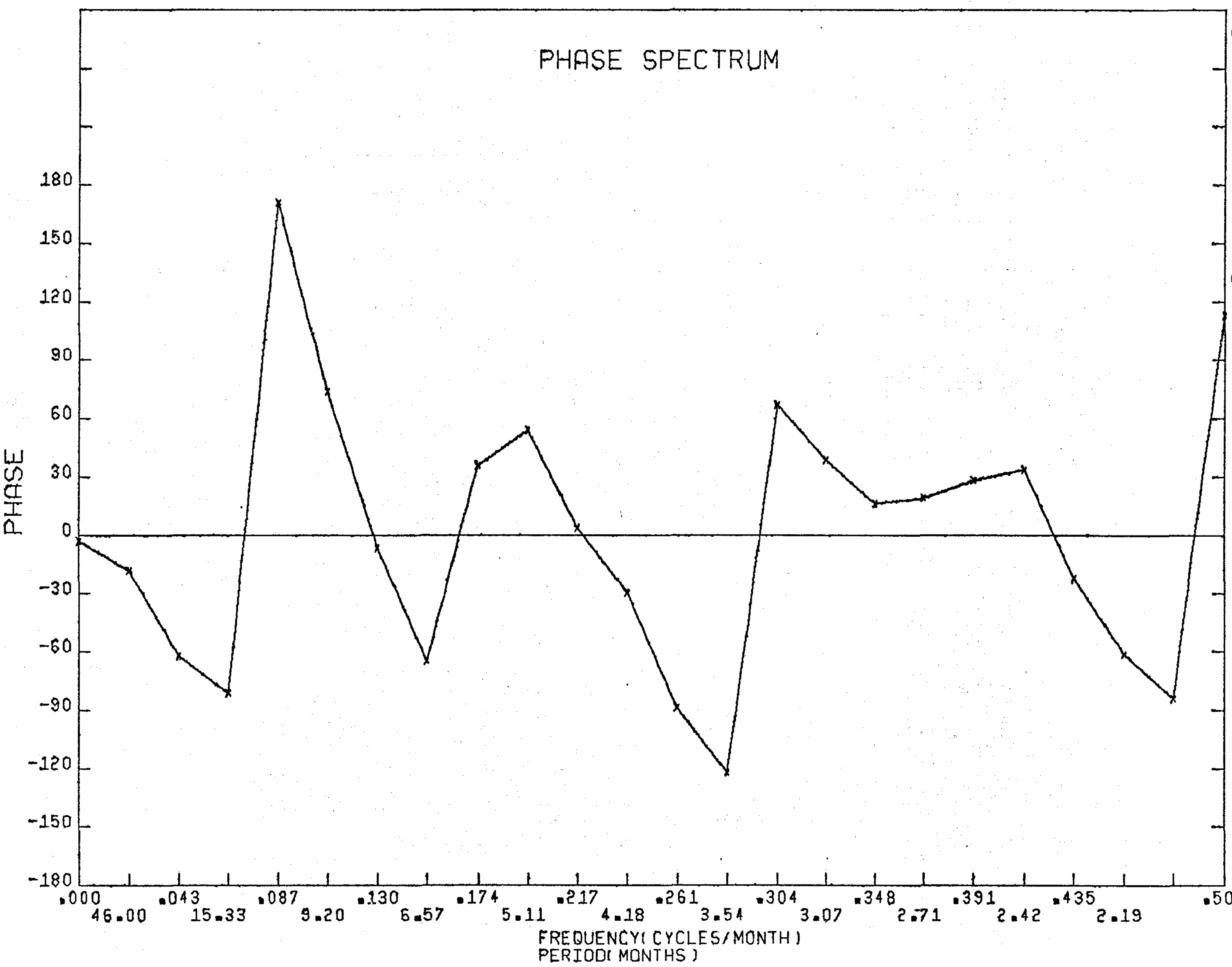


Figure 3.3.48: Phase spectrum for Cd T & Ni T, U.G.L. area (7201-7511)

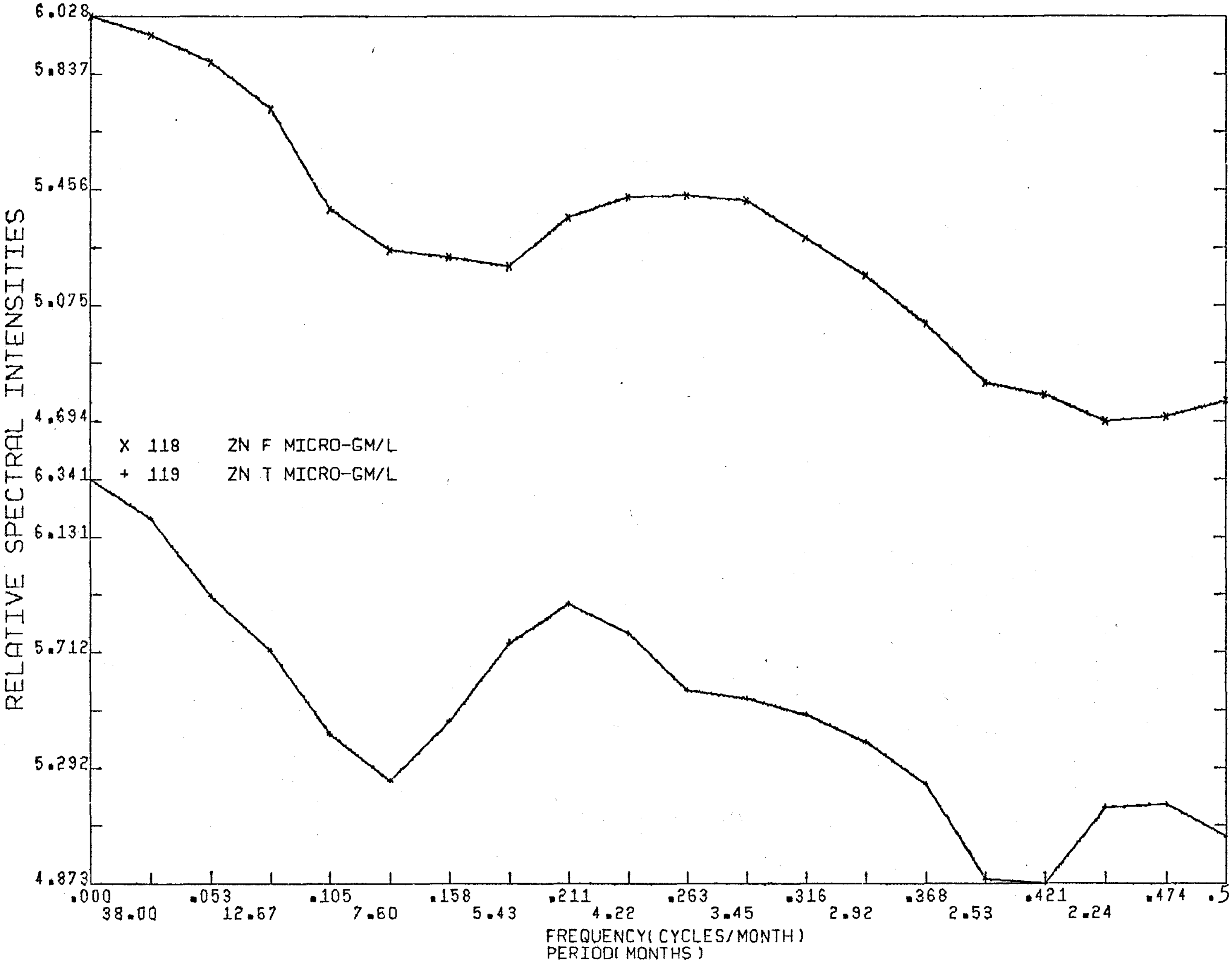


Figure 3.3.49: Power spectrum for Zn T & Zn F, Sudbury area (7204-7511)

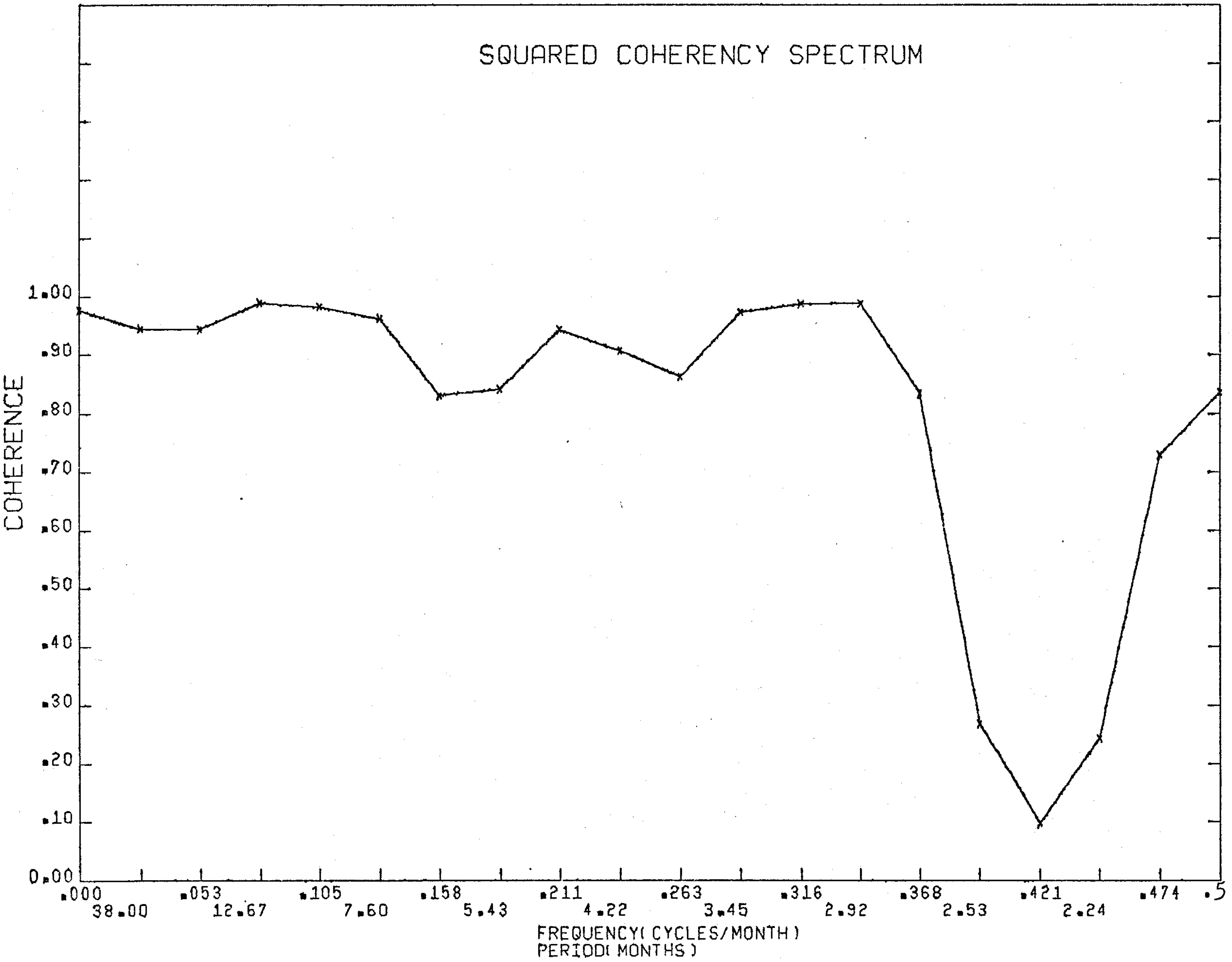


Figure 3.3.50: Coherency spectrum for Zn T & Zn F, Sudbury area (7204-7511)

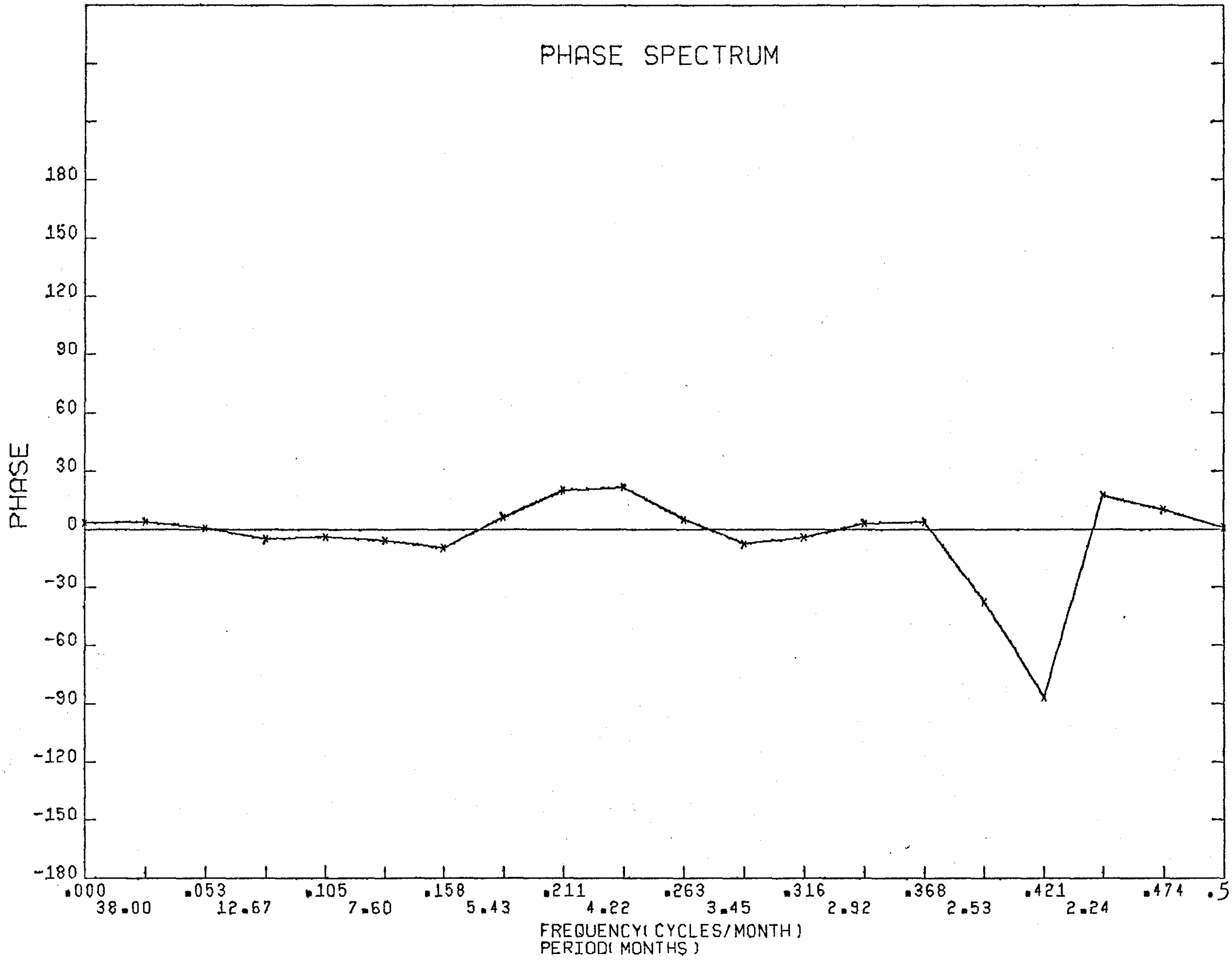


Figure 3.3.51: Phase spectrum for Zn T & Zn F, Sudbury area (7204-7511)

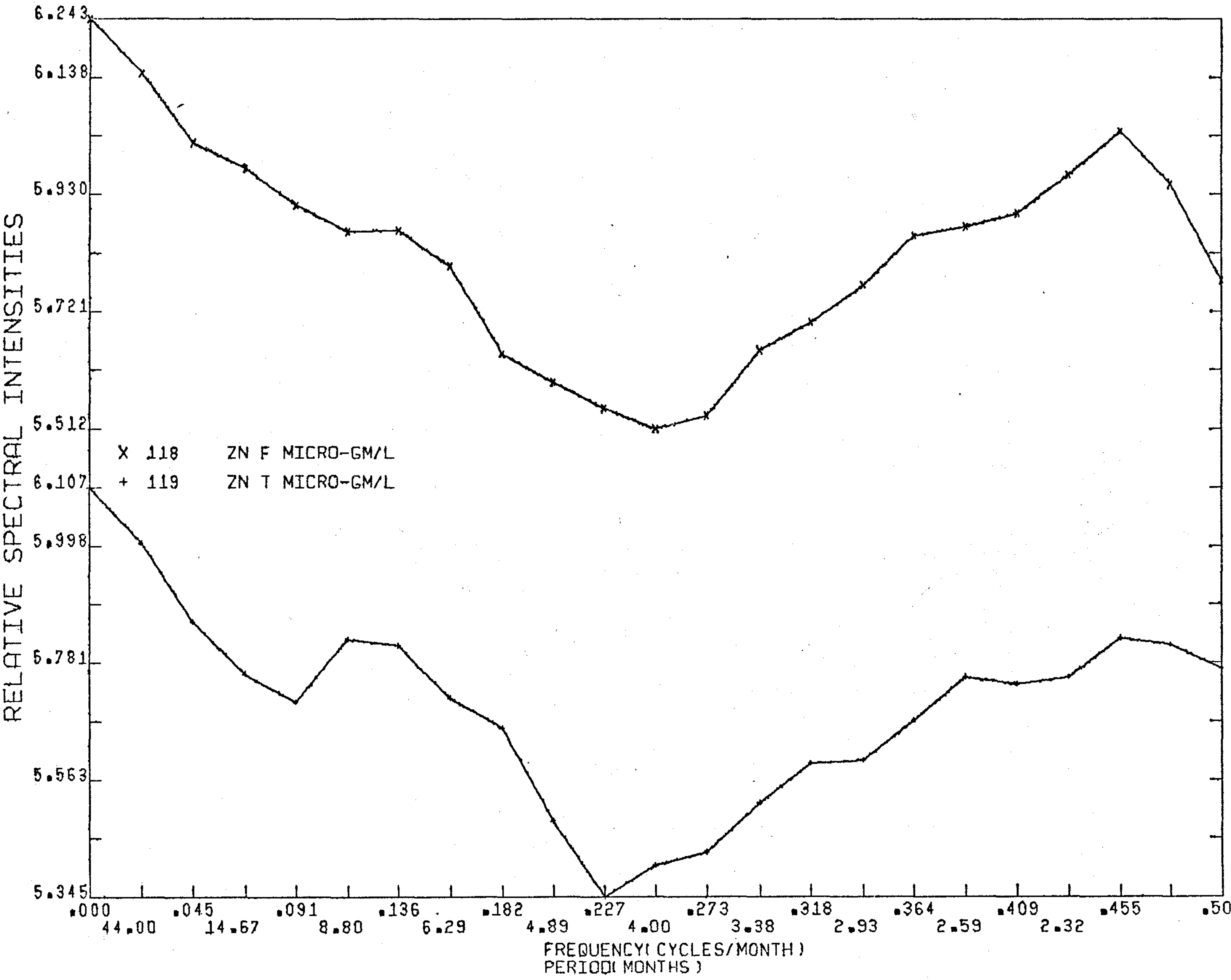


Figure 3.3.52: Power spectrum for Zn T & Zn F, U.G.L. area (7204-7511)

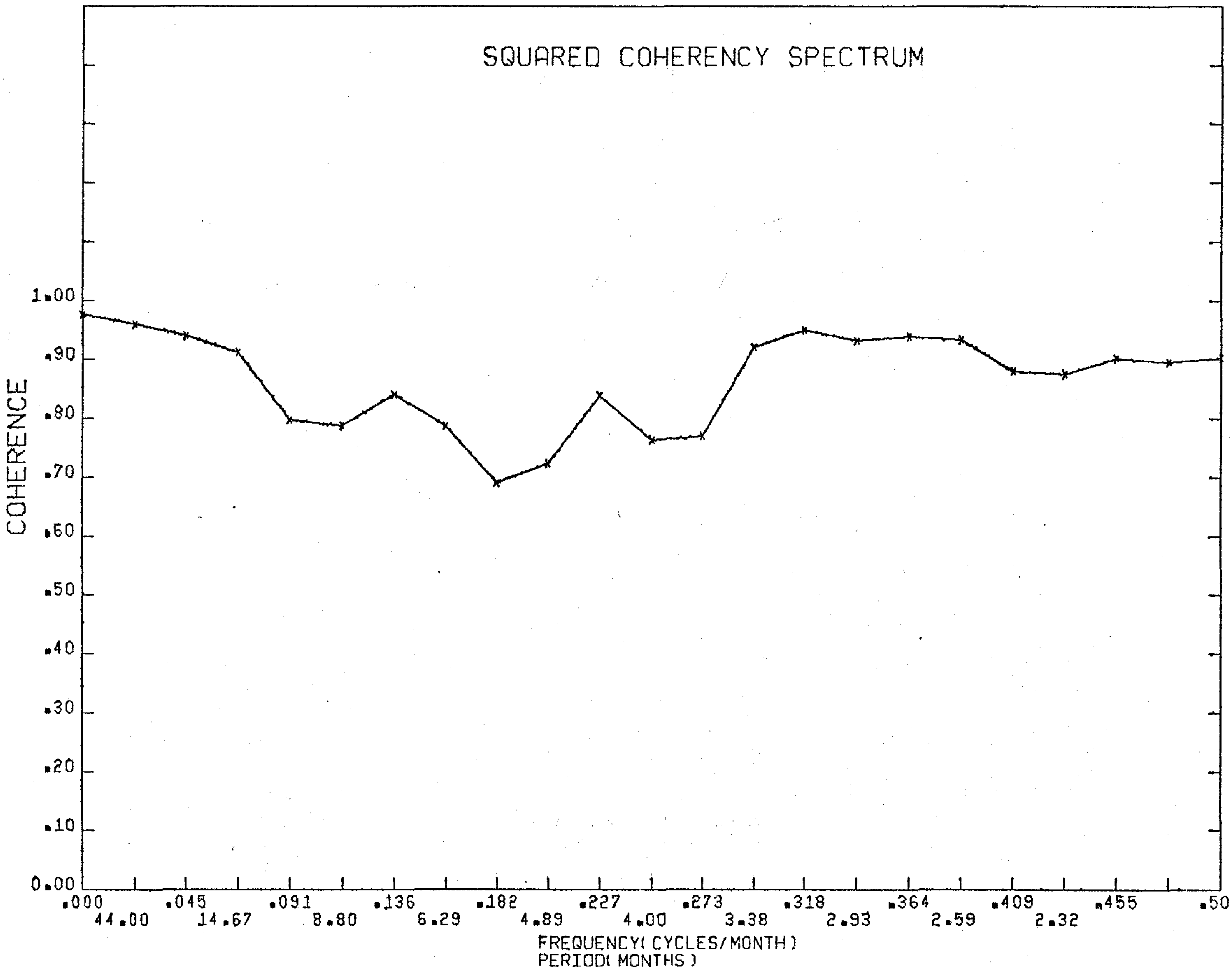


Figure 3.3.53: Coherency spectrum for Zn T & Zn F, U.G.L. area (7204-7511)

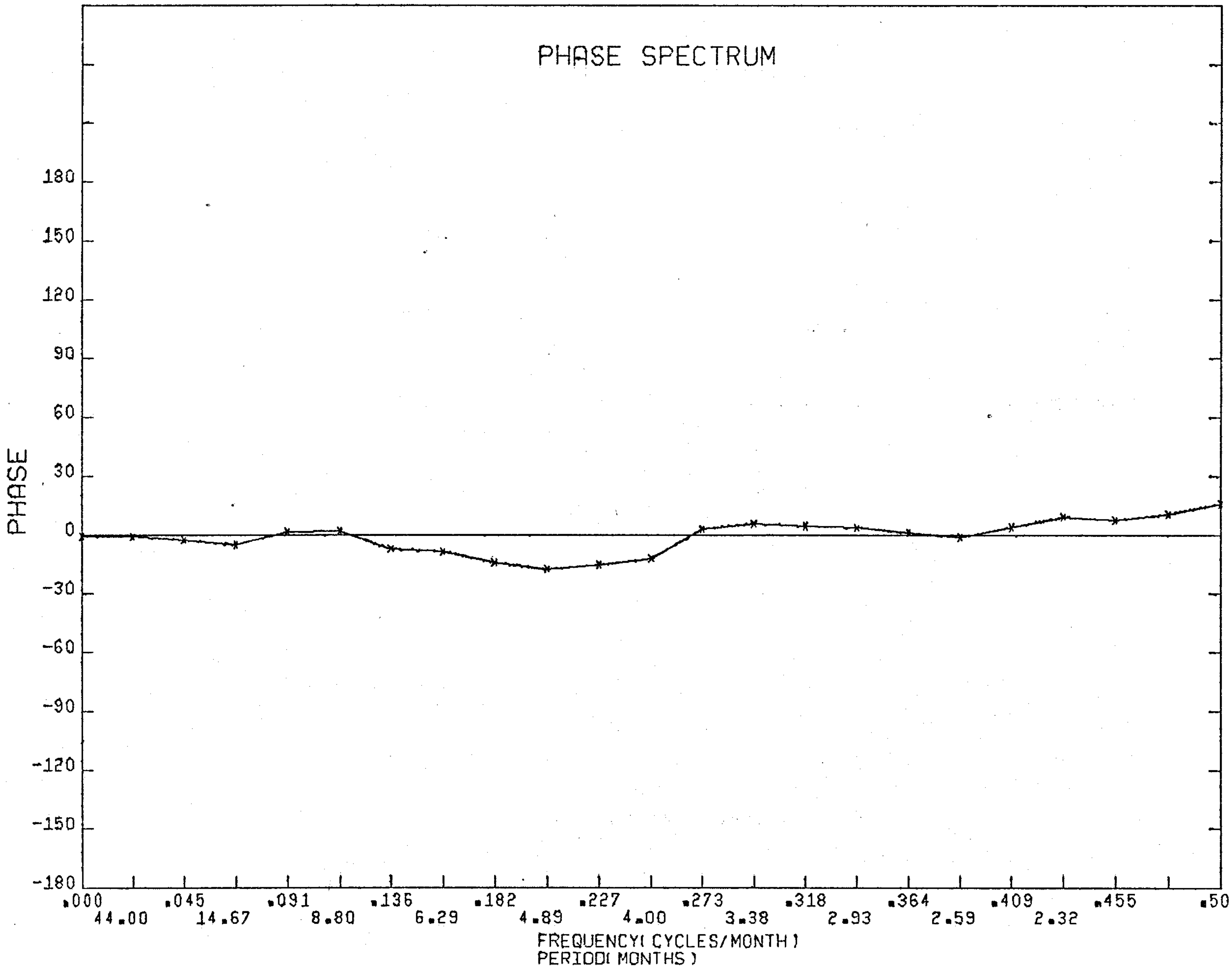


Figure 3.3.54: Phase spectrum for Zn T & Zn F, U.G.L. area (7204-7511)

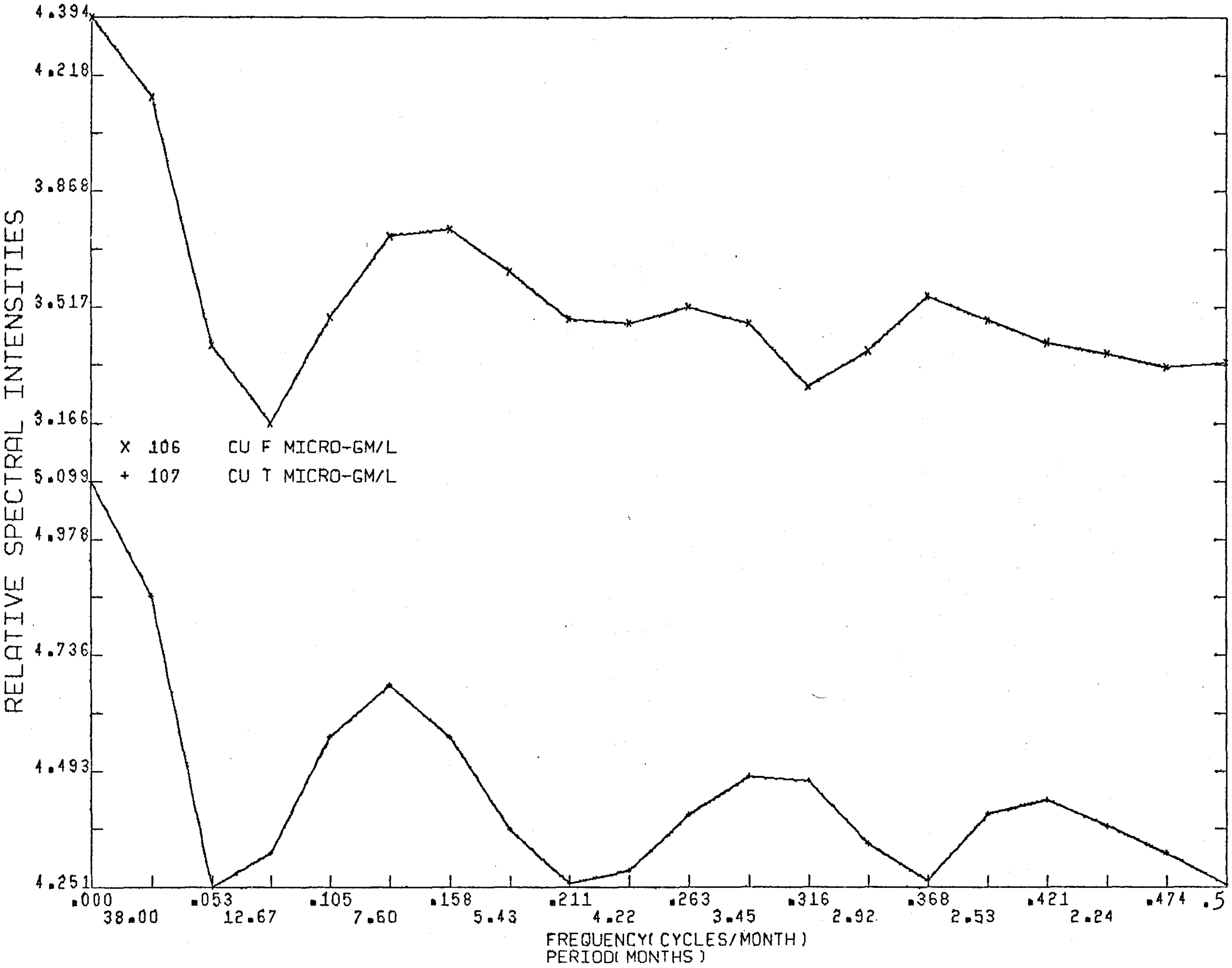


Figure 3.3.55: Power spectrum for Cu T & Cu F, Sudbury area (7204-7511)

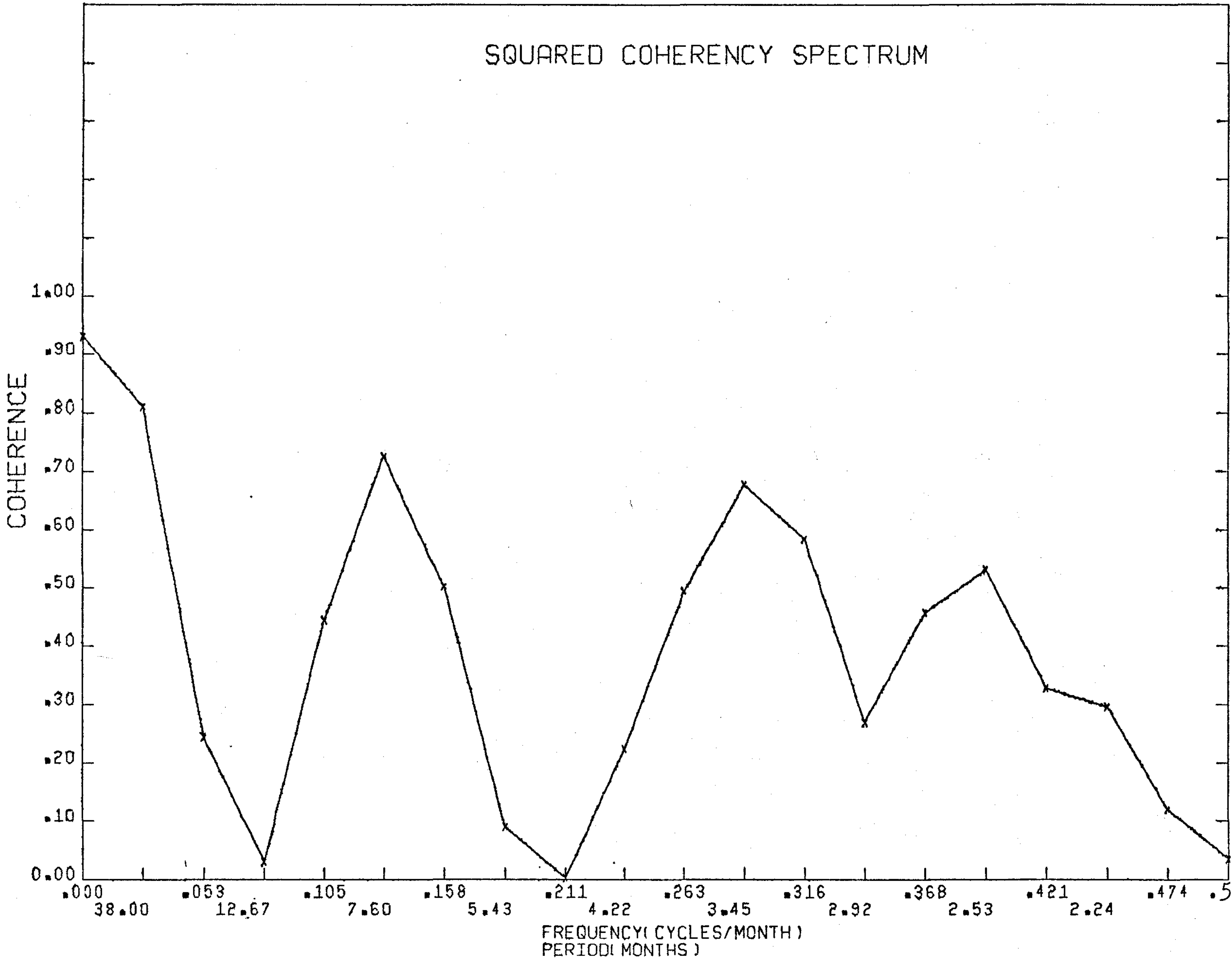


Figure 3.3.56: Coherency spectrum for Cu T & Cu F, Sudbury area (7204-7511)

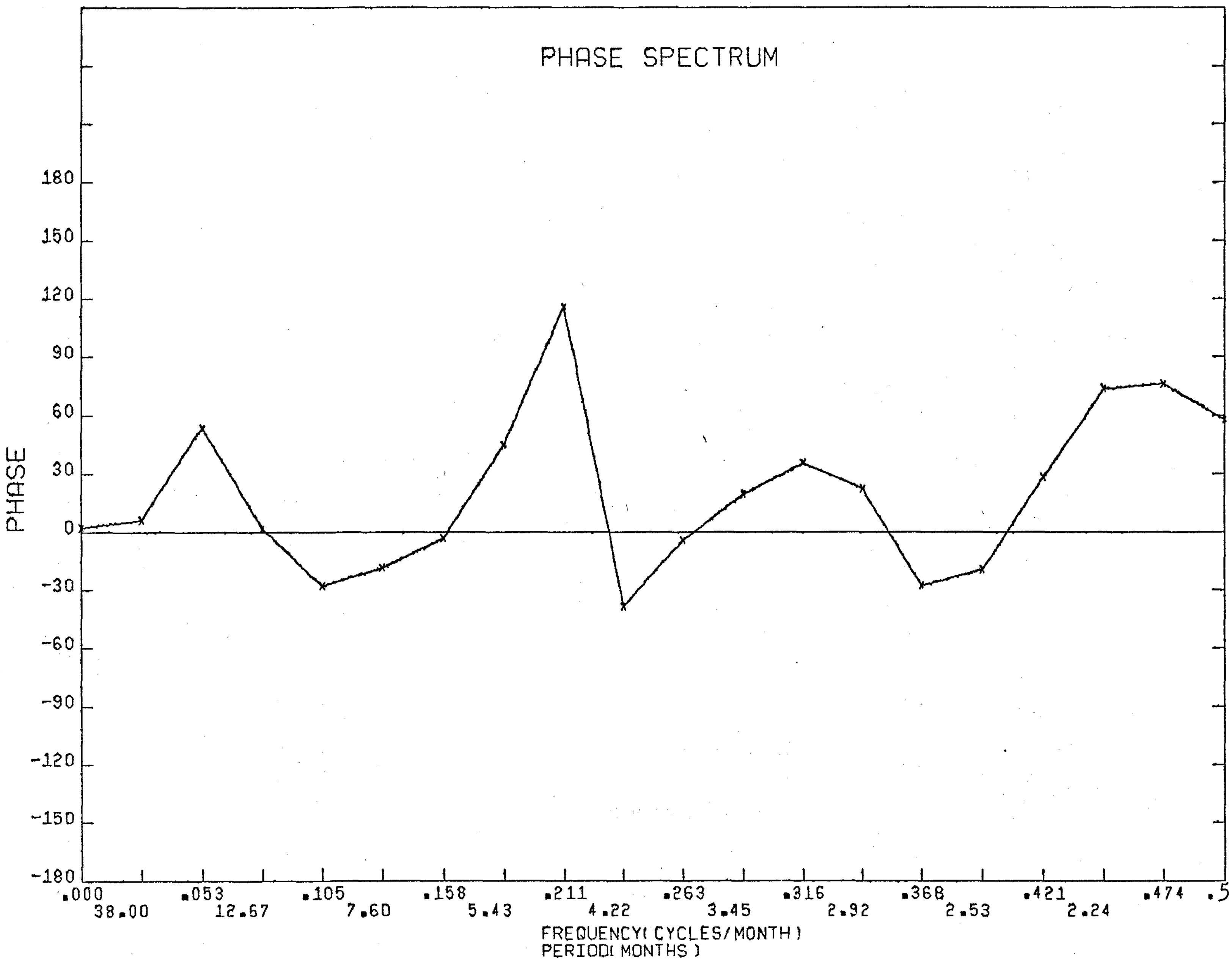


Figure 3.3.57: Phase spectrum for Cu T & Cu F, Sudbury area (7204-7511)

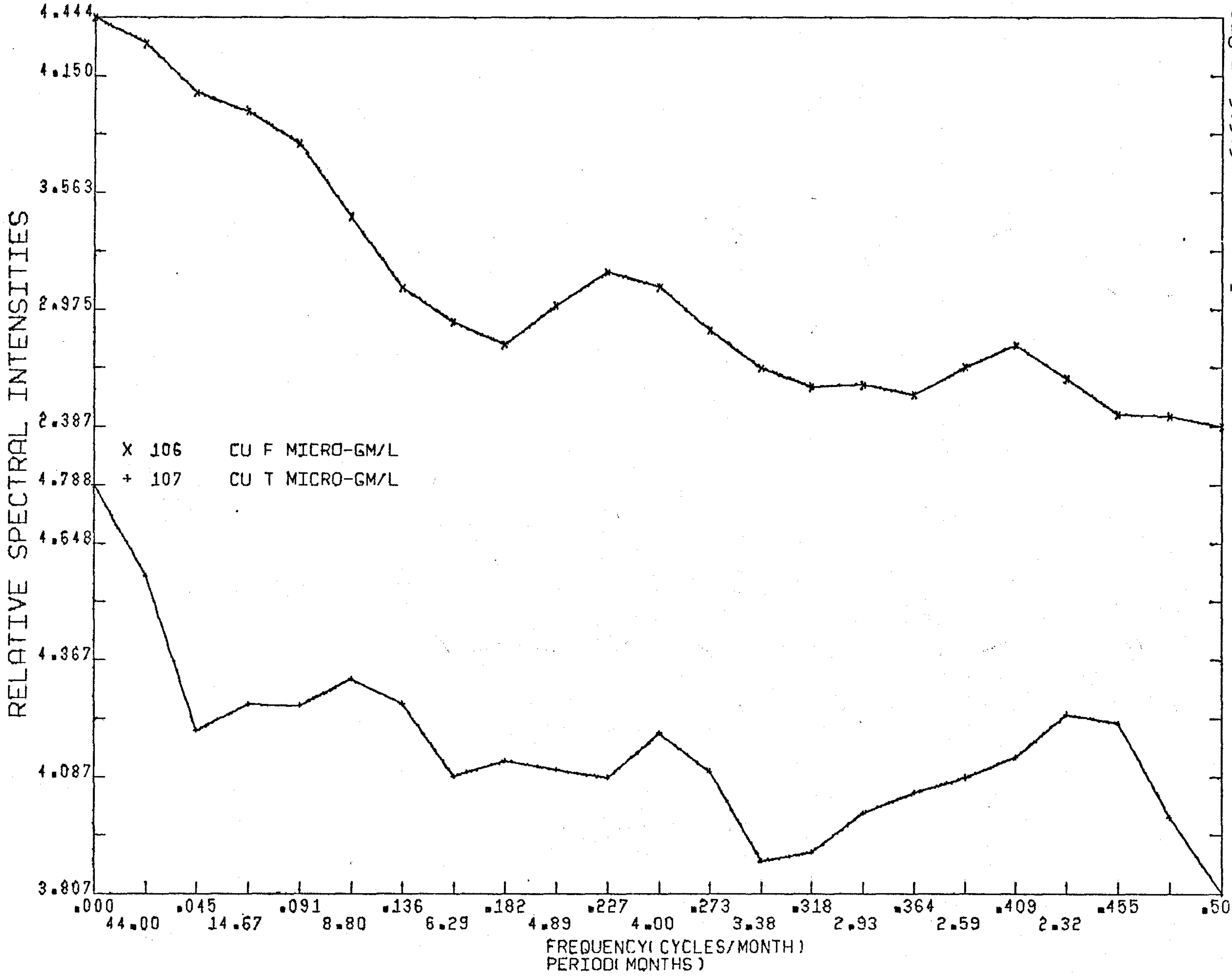


Figure 3.3.58: Power spectrum for Cu T & Cu F, U.G.I. area (7204-7511)

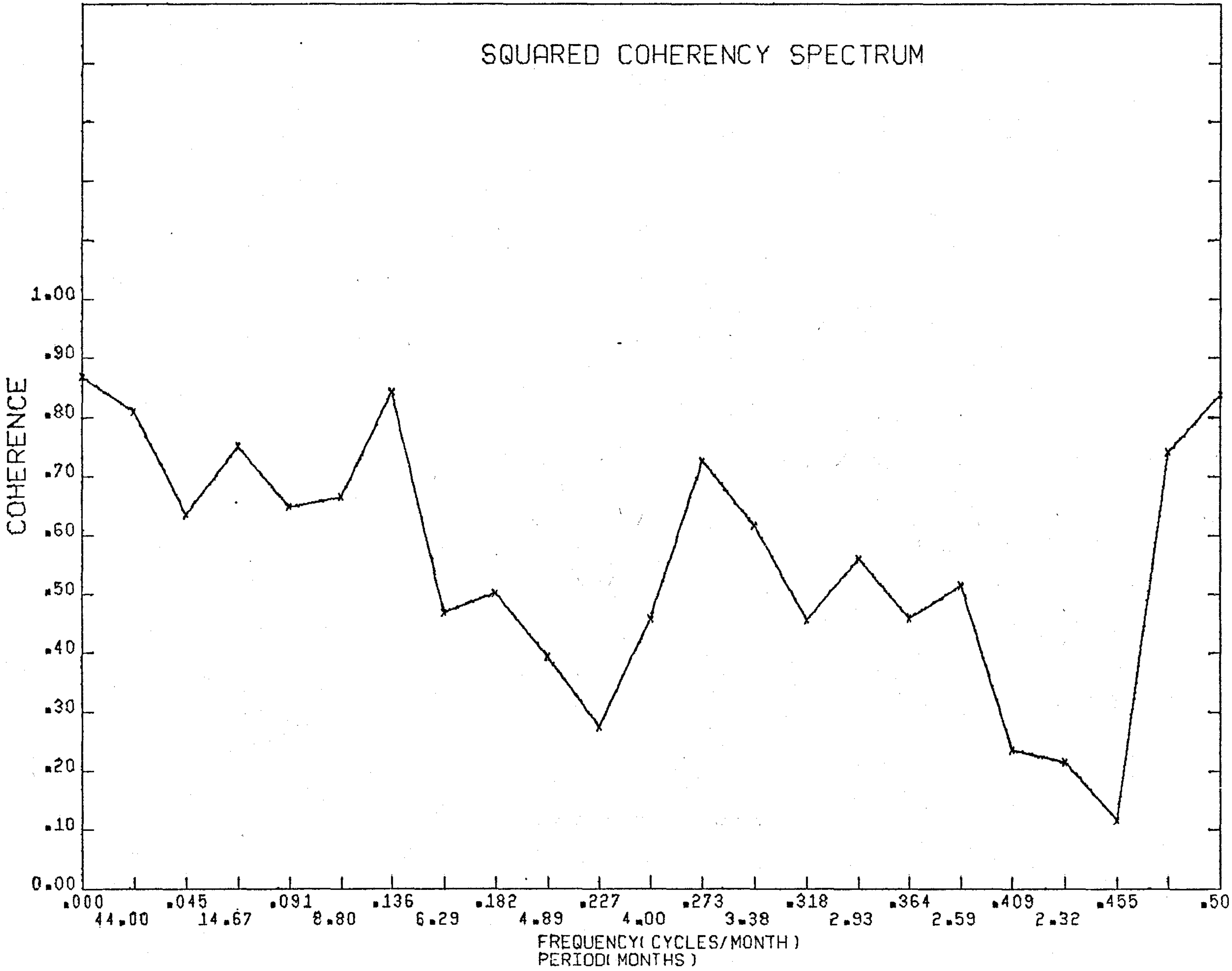


Figure 3.3.59: Coherency spectrum for Cu T & Cu F, U.G.L. area (7204-7511)

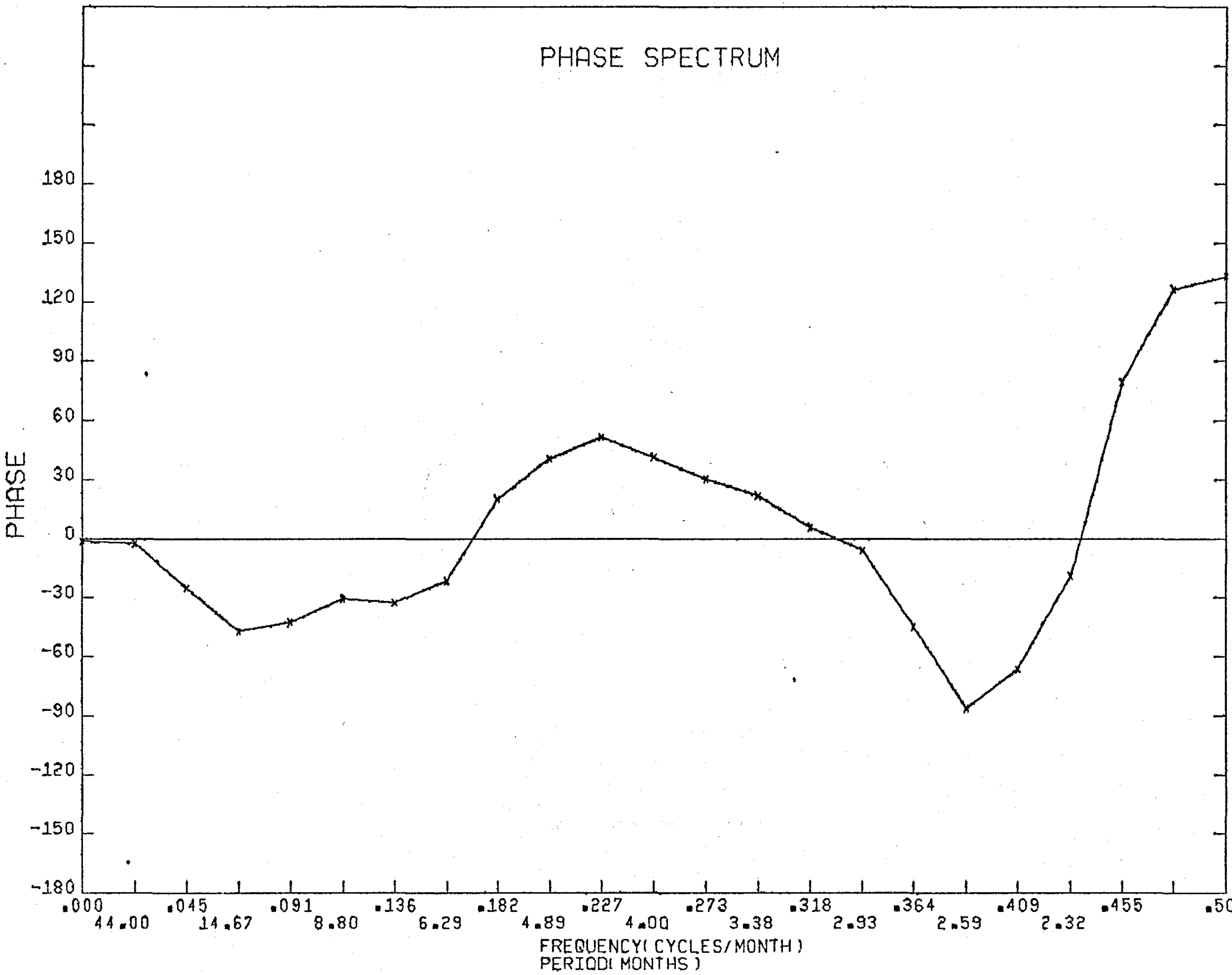


Figure 3.3.60: Phase spectrum for Cu T & Cu F, U.G.L. area (7204-7511)

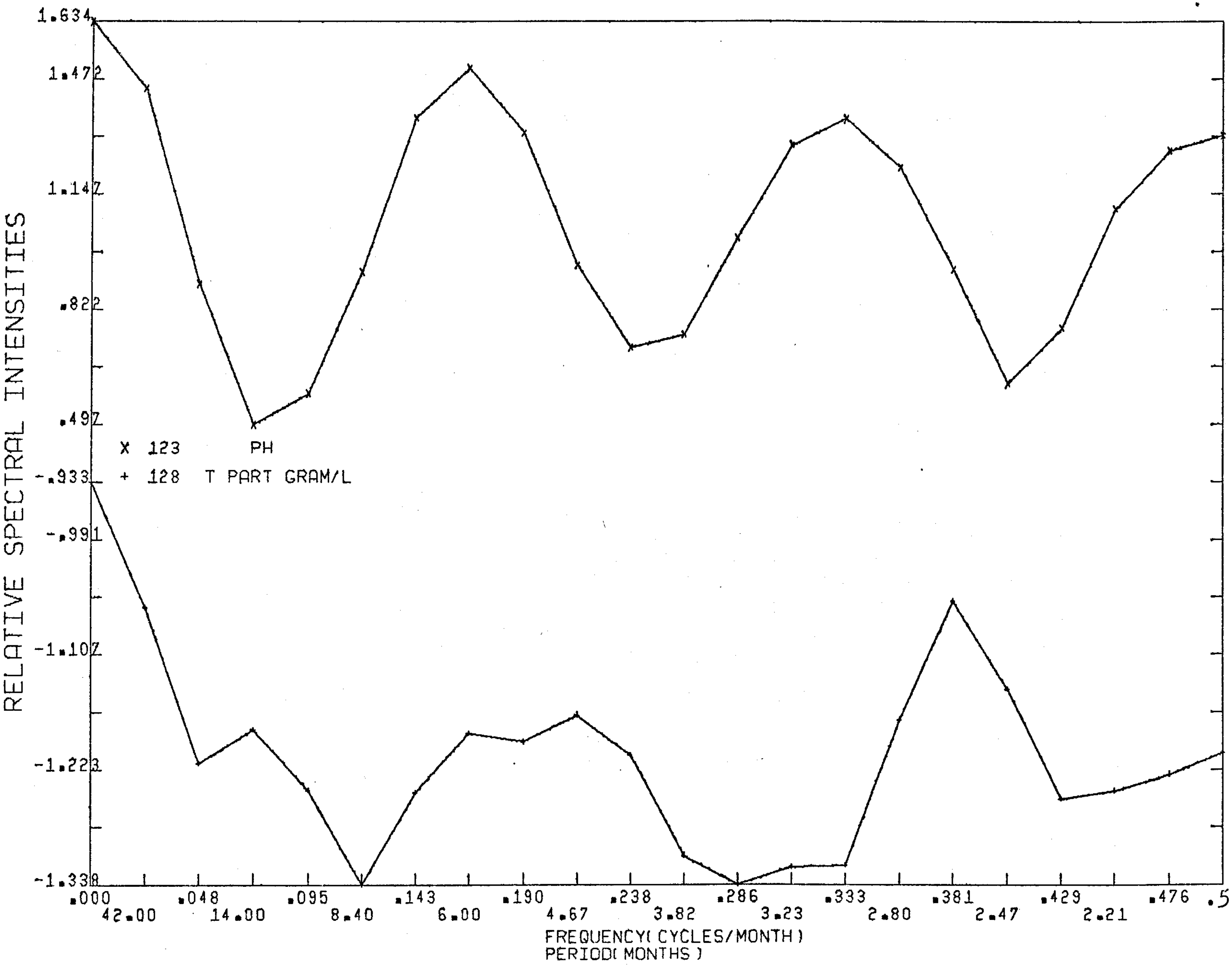


Figure 3.3.61: Power spectrum for pH & T PART, station 12 (7206-7511)

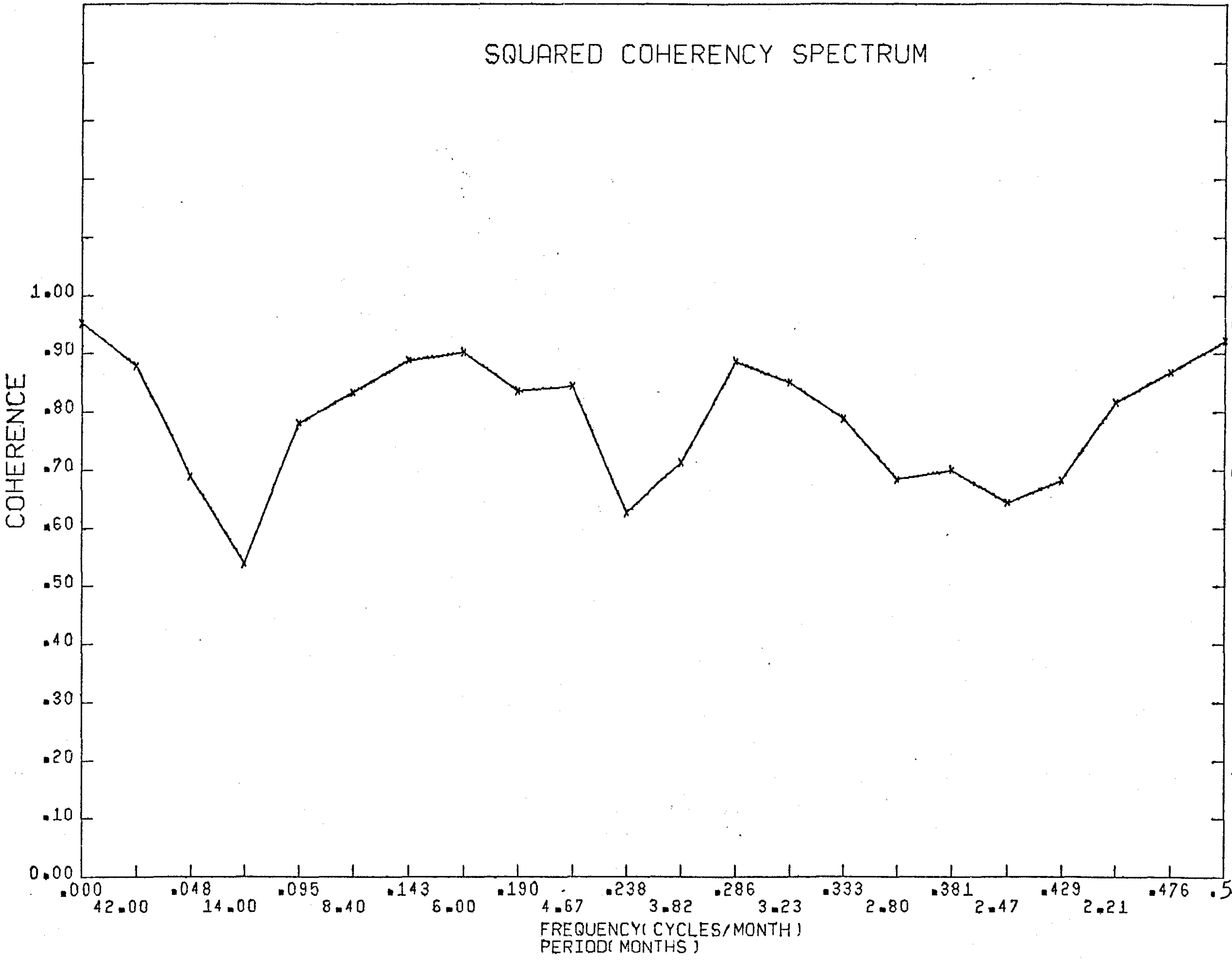


Figure 3.3.62: Coherency spectrum for pH & T PART, station 12 (7206-7511)

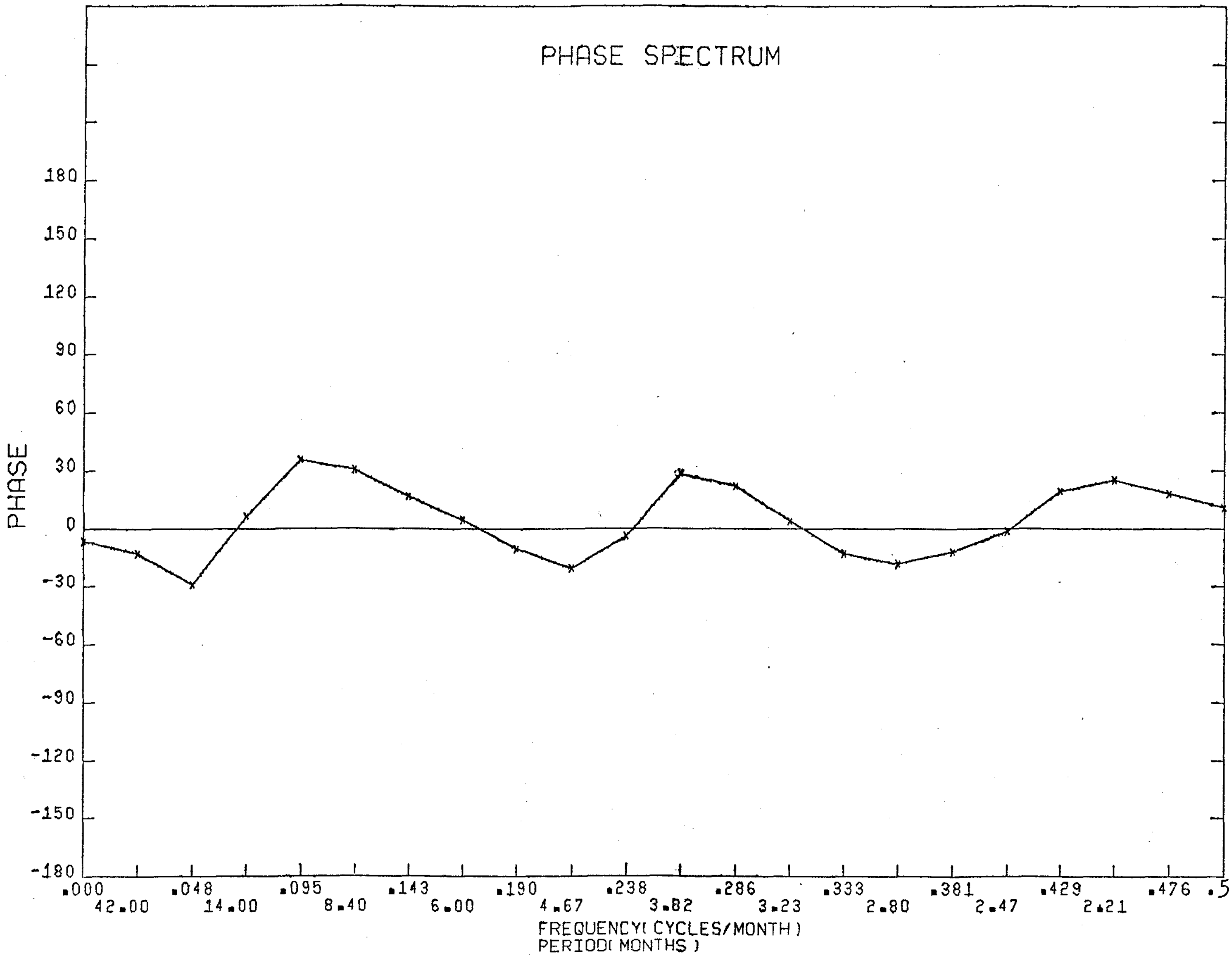


Figure 3.3.63: Phase spectrum for pH & T PART, station 12 (7206-7511)

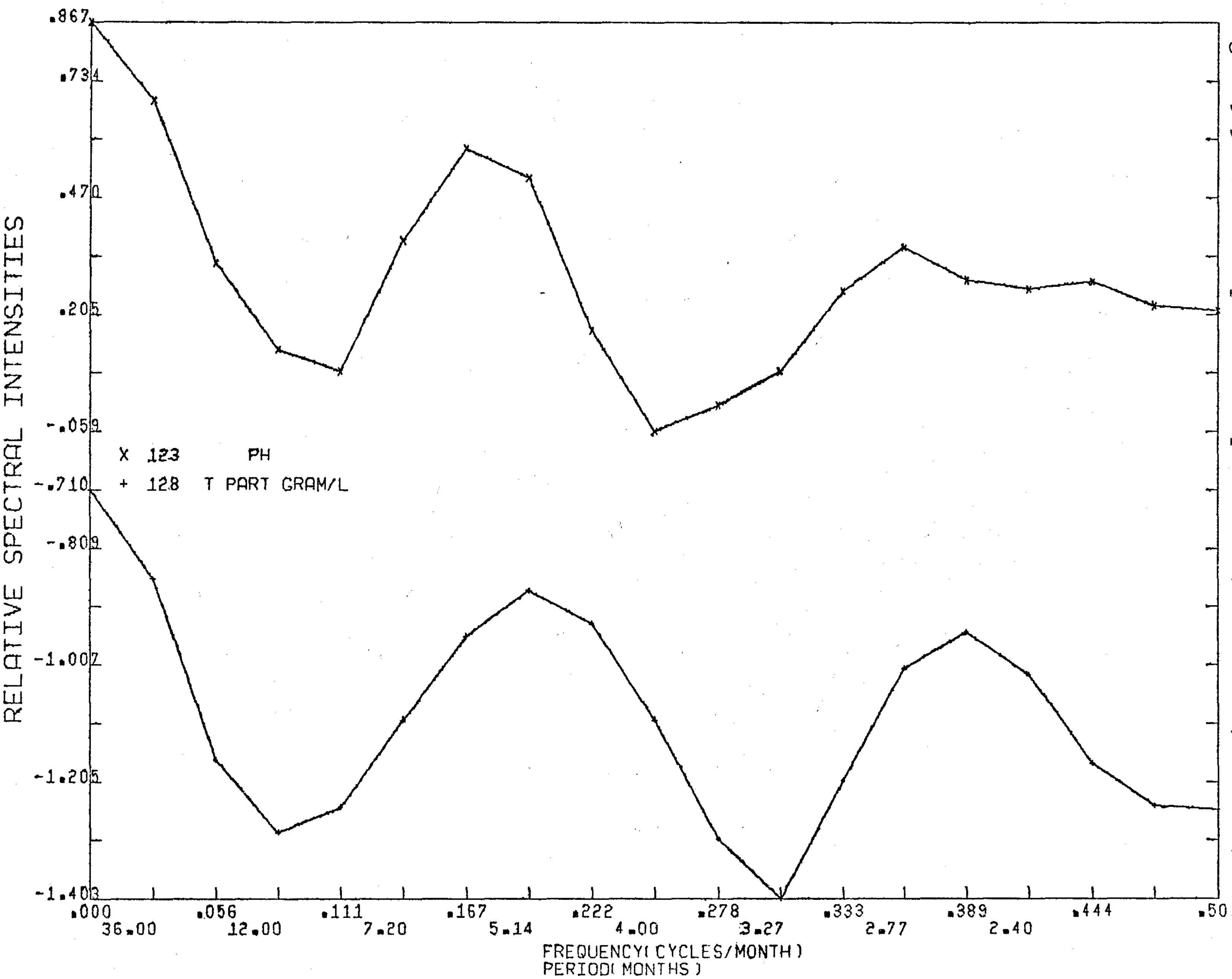


Figure 3.3.64: Power spectrum for pH & T PART, station 2 (7206-7511)

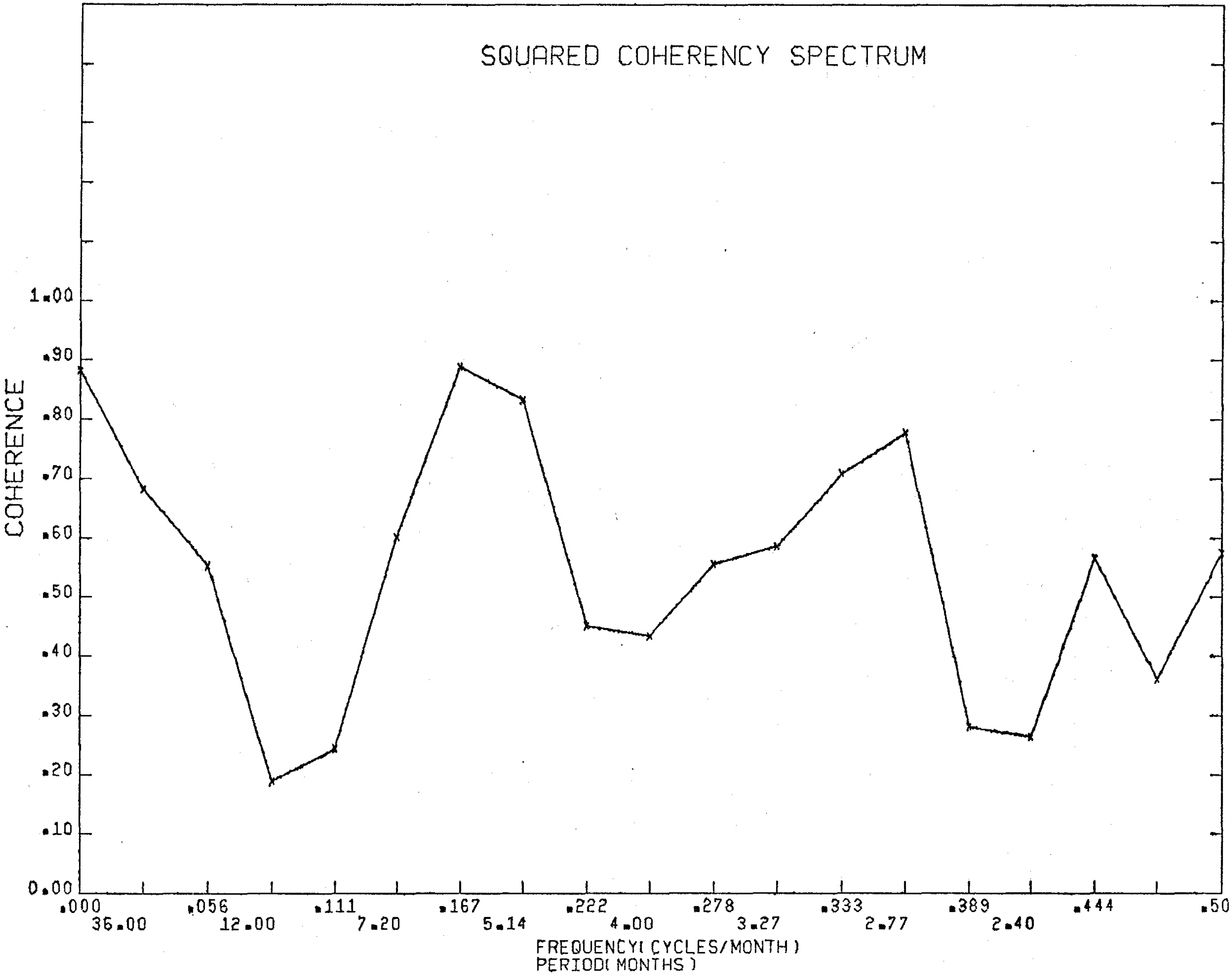


Figure 3.3.65: Coherency spectrum for pH & T PART, station 2 (7206-7511)

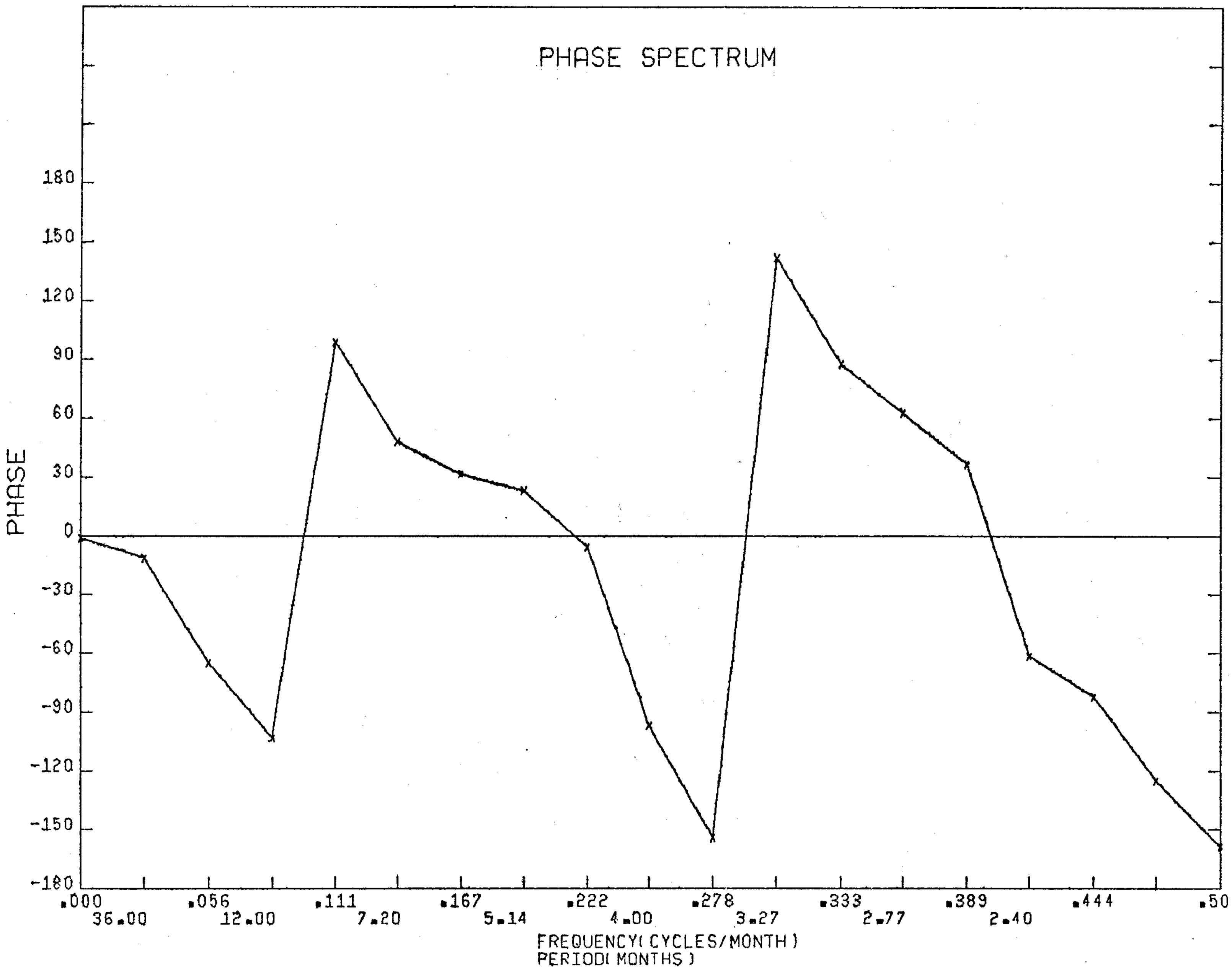


Figure 3.3.66: Phase spectrum for pH & T PART, station 2 (7206-7511)

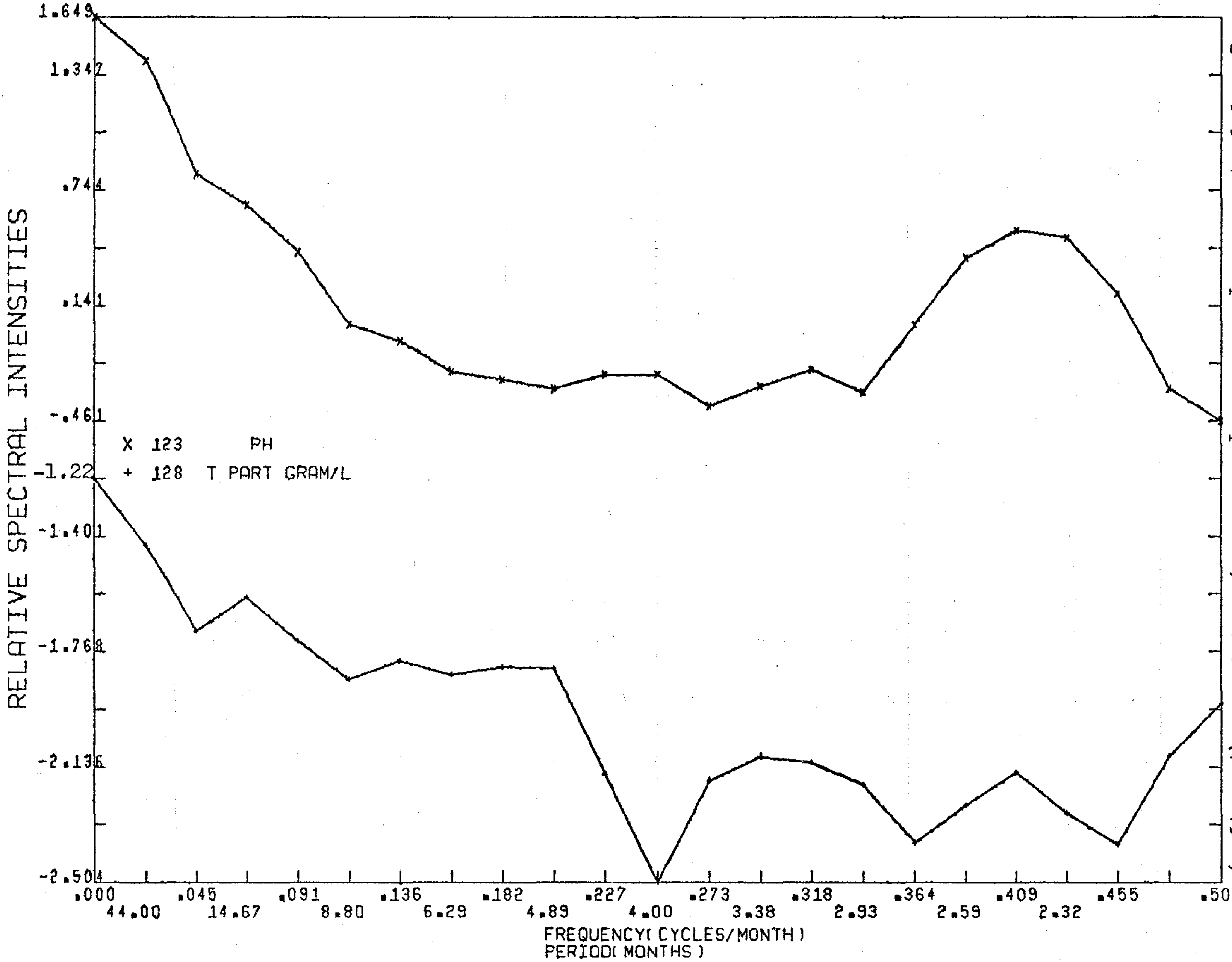


Figure 3.3.67: Power spectrum for pH & T PART, U.G.L. area (7204-7511)

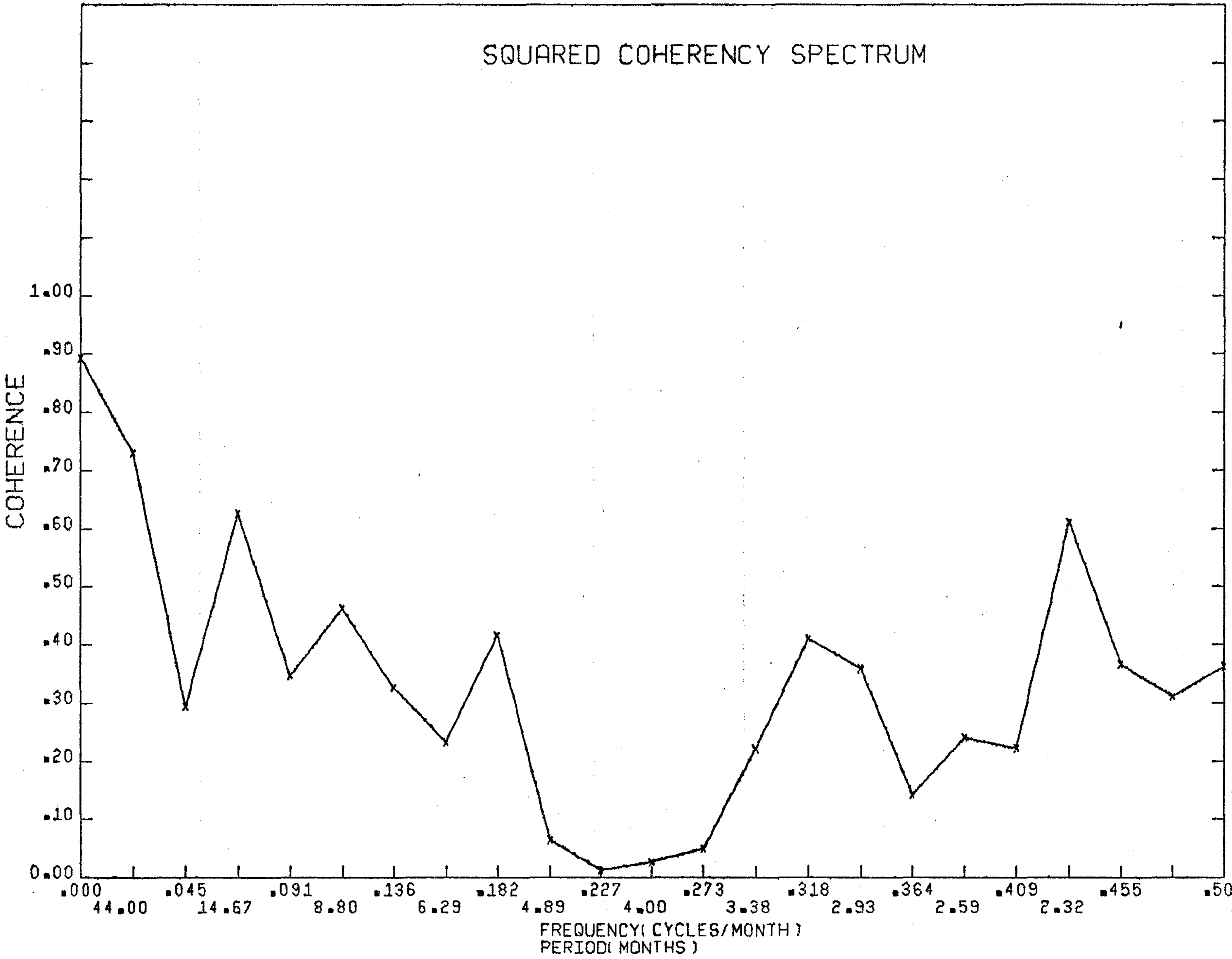


Figure 3.3.68: Coherency spectrum for pH & T PART, U.G.L. area (7204-7511)

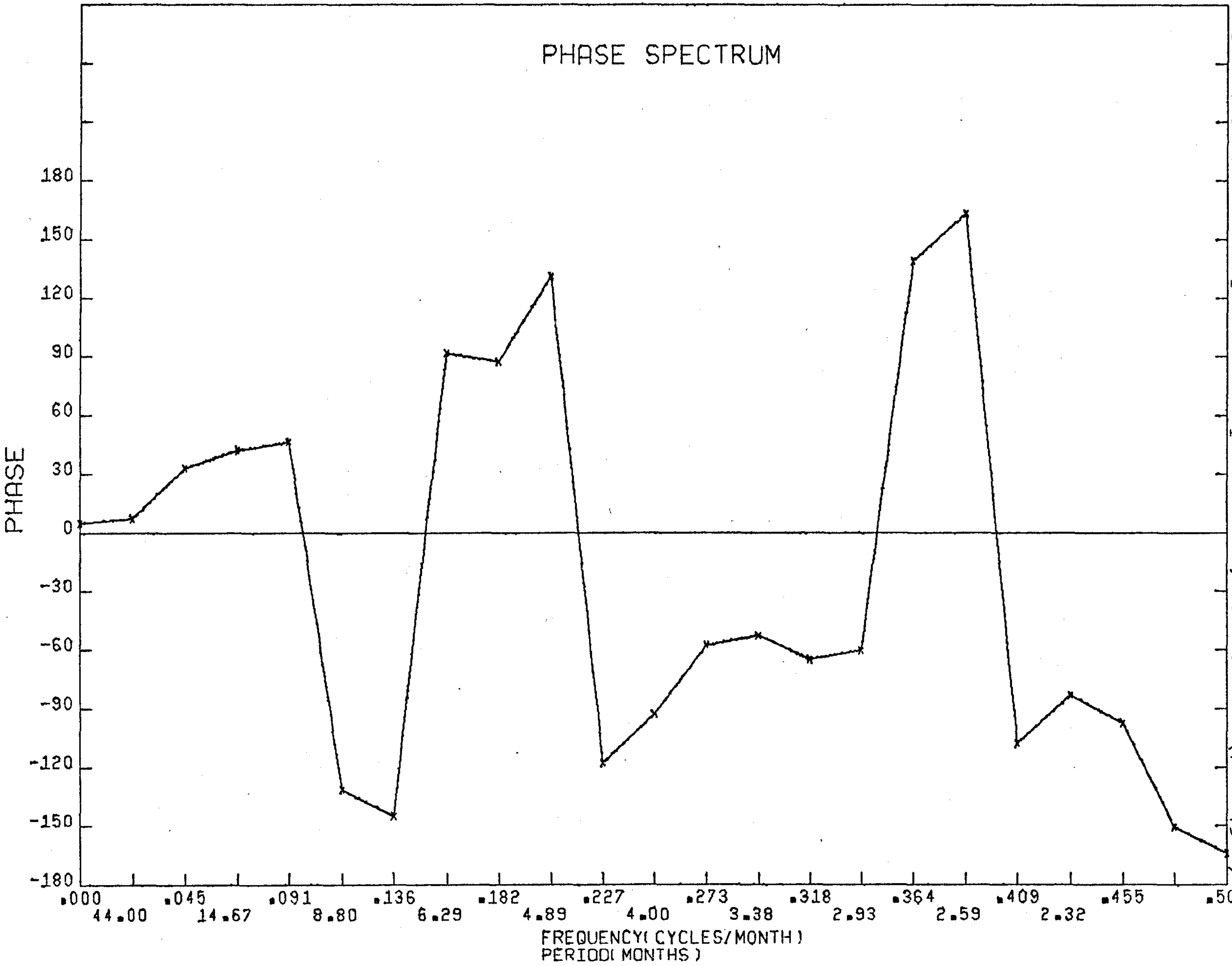


Figure 3.3.69: Phase spectrum for pH & T PART, U.G.L. area (7204-7511)

3.4 Errors for Precipitation Data

Section 3.3.2 discusses the errors involved in the spectral analysis with regards to the trigonometric functions. The same comments hold for the precipitation data. In addition, the precipitation data length is much shorter. On the average, about forty points are analyzed, which is less than half of that used for the trigonometric functions. This will introduce quite a bit of errors in the periodicities in particular. The writer has no control over this in that the data available are limited. However, as the record length increases the errors should decrease.

In an innocent example as the sine function, the trend could not be removed properly. Thus, for the rather fluctuating precipitation data, the trend removal is quite likely to be unreliable. It is extremely difficult to remove trends associated with the economy and at the moment it is the best one can do until the data are better understood. Only then methods other than the least squares fit can be used to remove the trends.

For a few data sets, gaps occur in the time series. If the duration is less than three months then the gaps are filled by substituting the result from the preceding month. Linear interpolation might have been better but since this is an initial investigation and greater errors occur for other reasons it was judged worthless of the effort.

It was assumed that chemical analyses are reliable to an extent. However, Kramer,(1975) observed variations among the different laboratories doing the analyses. This would certainly lead to some errors in the averaging procedure. It may be better to use data from one laboratory where the analysis is systematic to avoid random errors.

In view of the above remarks, a spectral analysis at this time is somewhat premature but should be very powerful as the record length of the time series increases and the data become better understood. Then confidence intervals can also be computed for the spectral estimates.

4. INTERPRETATION AND DISCUSSION

4.1 Choice of Pairs of Parameters

First and foremost, pairs of parameters are selected which are expected to have a high degree of correlation between them. The choice is guided by a knowledge of their chemistry and also from the similarity of the ratio of each parameter in rain to snow for 1973-74 period (Kramer, 1975). If these ratios for two parameters are close together, then presumably their chemical cycles are similar and a high degree of correlation may be expected. The choice is restricted somewhat by the length of the data which was recorded for each parameter.

The computer program is designed to compute spectral properties for two simultaneously recorded time series which by definition spans the same period of time. The length of the data is different for each parameter considered in this study. Therefore an appropriate truncation must be made to each pair of data set considered in order to obtain the data collected for both sets within the same period of time.

Furthermore, some parameters are studied only for their periodic components. For such parameters all the available data are used. However each of these parameters must be run against a dummy parameter which has data of equal or longer length than that of the parameter of interest. The pH data is the longest and is often used as a dummy parameter.

Emphasis in this work is placed on the heavy metals (Cd, Cu, Ni, Fe, Pb, Zn) due to their industrial association, particularly in view of the levels of atmospheric pollutants presently being emitted from the smoke stacks at Sudbury (Kramer, 1975) which act as a point source. Furthermore, the data for these metals are more or less complete.

The periodic patterns of parameters other than the heavy metals are also considered and compared to those of the heavy metals. Such parameters are pH, SO_4, SP CON, PART and P. These parameters are certainly related to industries, however, a substantial proportion of airborne phosphorus is thought to be associated with agriculture.

4.2 Periodicities

The periodic components of the precipitation data can be resolved from the power spectrum in that the "peaks" in the spectrum occur at persistent periodicities. The intensities of the peaks indicate the persistence of each periodic component. Therefore, in each power spectrum, the intensities at periodicities can be arbitrarily classified as strong (s), medium (m), or weak (w) to reflect the relative importance of each periodic component. It should be noted that the intensities are in log units, thus the values for the intensities appear to be similar, but in fact they are quite different. The weak periodicities in the power spectrum may be due to termination errors, but this is difficult to establish at the present time. Furthermore, since the data for the parameters are somewhat limited from the point of view of spectral analysis, only the pattern of periodicities associated with the various parameters at different locations, is important.

Figures 4.2.1(a) to 4.2.1(c) show the periodicities associated with the various parameters at different locations. The solid, dashed

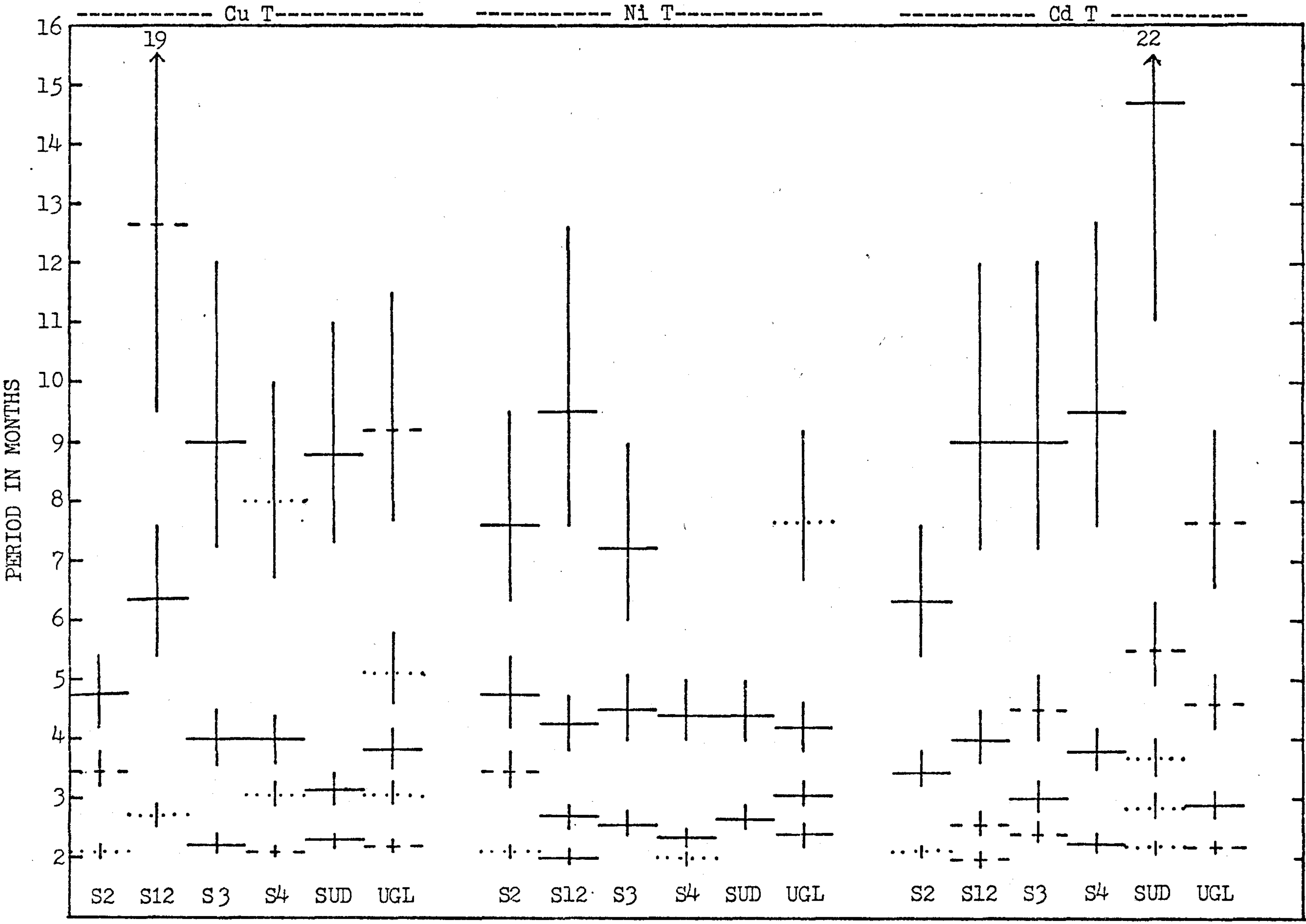


Figure 4.2.1(a): Periodicities of Parameters

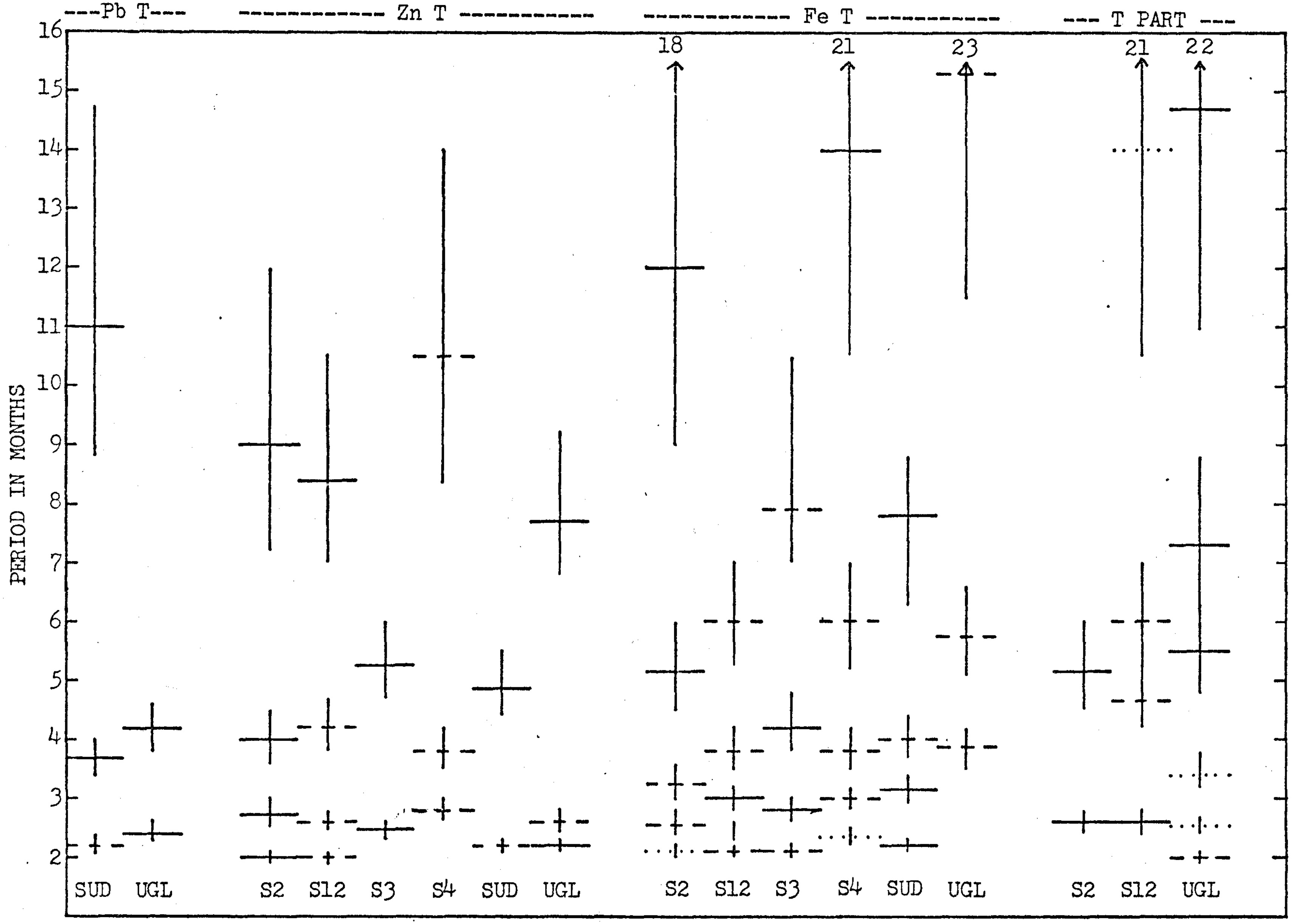


Figure 4.2.1(b): Periodicities of Parameters

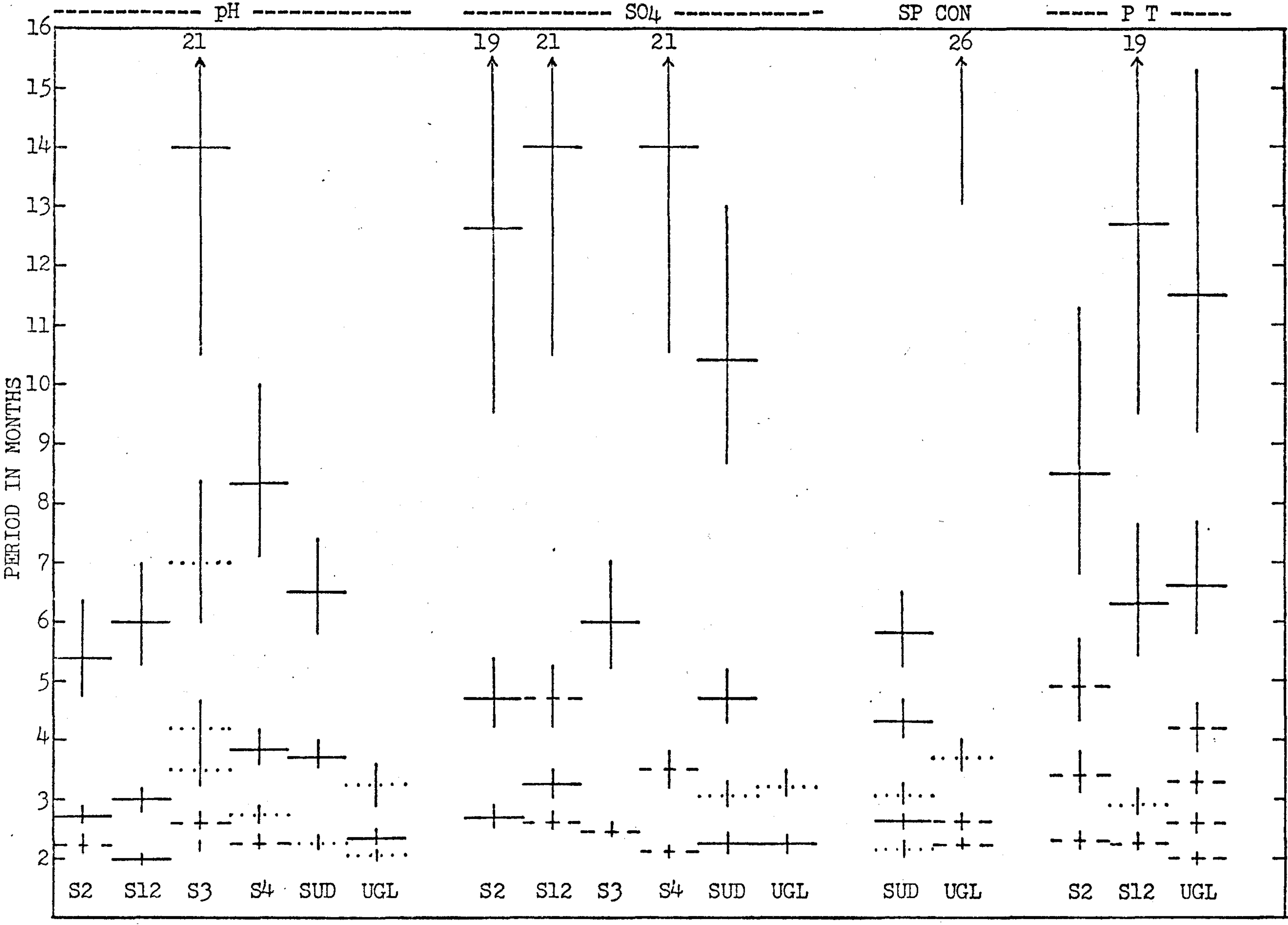


Figure 4.2.1(c): Periodicities of Parameters

and dotted lines represent strong, medium and weak periodicities respectively, with respect to the peak intensities in each power spectrum. The error bars shown in the figures result from the fact that if a smooth curve is drawn through three points then an error is associated with the positions of the maximum or the minimum. However, more than this error occurs in the present analysis and thus the error bars are taken as the two points on either side of the "maximum" point.

From the figures, definite patterns can be seen for the periodicities. The heavy metals, in particular Cu, Ni, Cd, Pb and Zn, generally show two frequency bands of periodicities. The first is a period of 9 cycles/month and the second is a period or periods in the range of 2 to 5 cycles/month.

Iron is the only heavy metal that, though it shows the lower periodicities, does not show the 9 monthly period as associated with the other heavy metals. However, it does show 12 monthly and 6 monthly periodicities. The particulates (T PART) have periodicities that are the same as for iron. This is due to the fact that most of the particulates consist of iron.

The other parameters also show periodicities ranging from 2 to 5 cycles/month. In addition, pH shows a 6 monthly period and SO_4 shows a 12 monthly period.

Phosphorus shows, in addition to the low periodicities, both 6 and 12 monthly periods. Since phosphorus is generally associated with agriculture, a 12 monthly period is expected. This represents high phosphorus concentration in summer and low in winter. The 6 monthly period may reflect the four seasons of the year.

The origin of the high periodicities for the other parameters is not well understood but the periodicities must be real due to the high intensities observed in the power spectra. For Fe and PART, the 6 monthly period may reflect the four seasons while some yearly cycle must be responsible for the 12 monthly period. Whatever this cycle is, it may also explain the 12 monthly period of SO_4. The yearly cycle may be seasonal, representing high concentration of the parameters in summer and low in winter. The 6 monthly period is less clear and the data for the parameters showing this period fluctuates throughout the year. It is quite likely therefore, that the four seasons may be responsible for this periodicity.

The 9 monthly period for the heavy metals is even more difficult to explain in view of the fact that not many natural cycles of this duration are known. However, since most of the metals are associated with industries, this periodicity is most likely reflecting some strong periodic components involved with industries.

Furthermore, all the parameters considered in this study show a high frequency component of periodicities ranging from 2 to 5 cycles/month. Thus there must be some universal phenomenon affecting all the parameters. A study of the spectrum of the vertical velocity field of atmospheric turbulence (Lumley and Panofsky, 1964) indicates that peak in the spectrum moves towards lower frequencies with increased solar radiation. This suggests that there are two different causes for the fluctuations in atmospheric turbulence, a high frequency component due to frictional forces and a low-frequency component due to heat convection caused by solar radiation. The precipitation data are certainly affected by atmospheric turbulence, thus the high frequency periodicities may very well be due to frictional forces associated with turbulence.

Periodicity is due to "natural" phenomena and in some way it may also reflect the origin and/or behaviour of the precipitation data. This study shows clearly that Fe is the only heavy metal that does not follow the pattern of the other heavy metals, and therefore cannot be as strongly linked to the smoke stacks at Copper Cliff as are the other heavy metals. This is further supported by correlation and phase studies discussed in the next section.

4.3 Correlation and Phase Relationships

The squared coherency and phase spectra give the correlation and phase relationships of any two parameters. A high coherence indicates that the two parameters are correlated at that frequency band and the phase spectrum gives the phase difference between the two series as a function of frequency.

4.3.1 Cu T and Ni T

The spectra for Cu and Ni are given in Figures 3.3.13 through 3.3.30. Table 4.3.1 summarizes the periodicities, peak intensities, correlation and phase relationships.

The correlation between Cu and Ni is in general very good for the major periodicities. At station 12, the correlation is somewhat low thus reflecting the questionable data for Ni (Fig. B.1.5). Data are judged to be unreliable if anomalous values occur for a few months. In such cases the least squares fit and trend removal are impractical. For the other five spectra where the data are better the correlation is quite good.

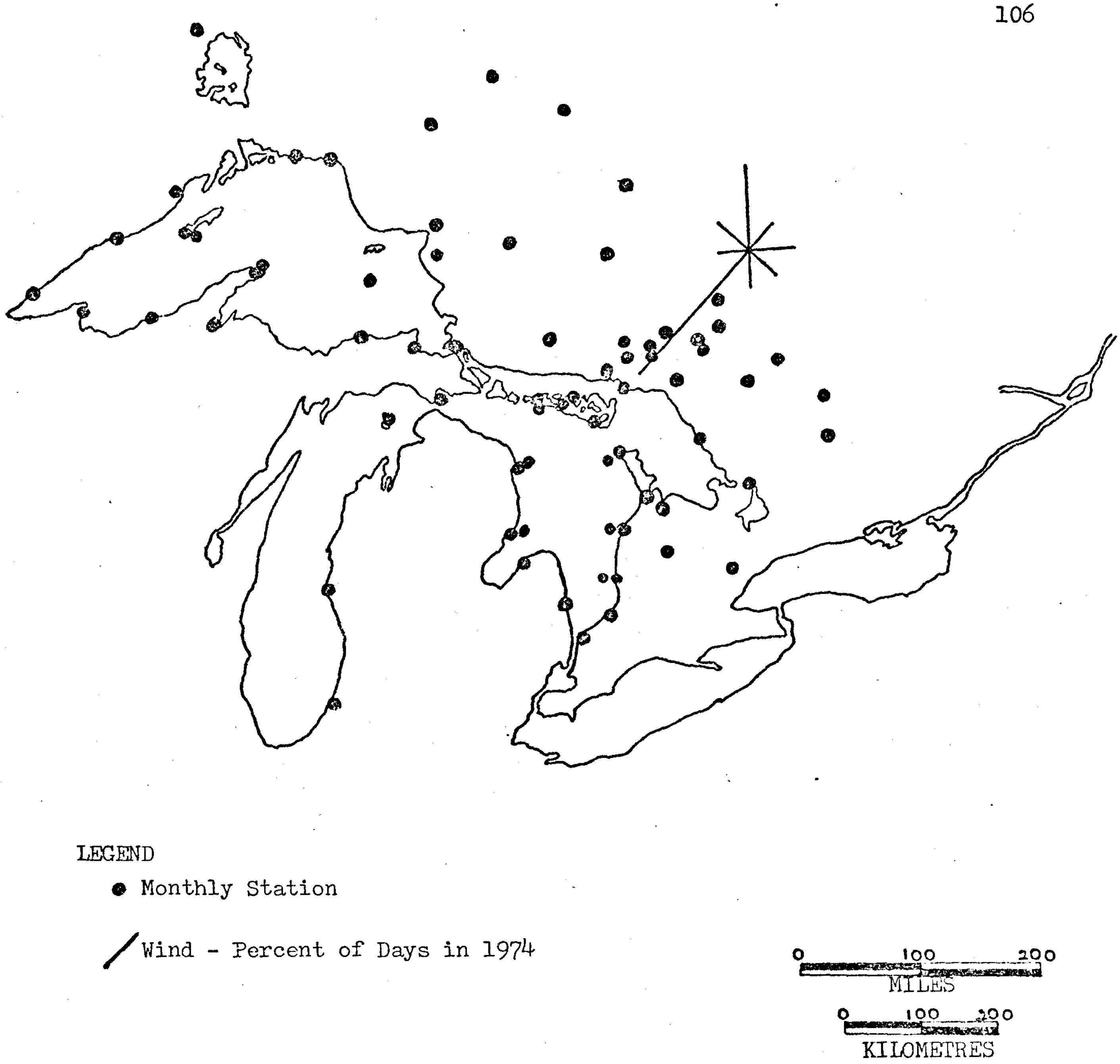


Figure 4.3.1: Precipitation chemistry stations and wind directions in the Upper Great Lakes region

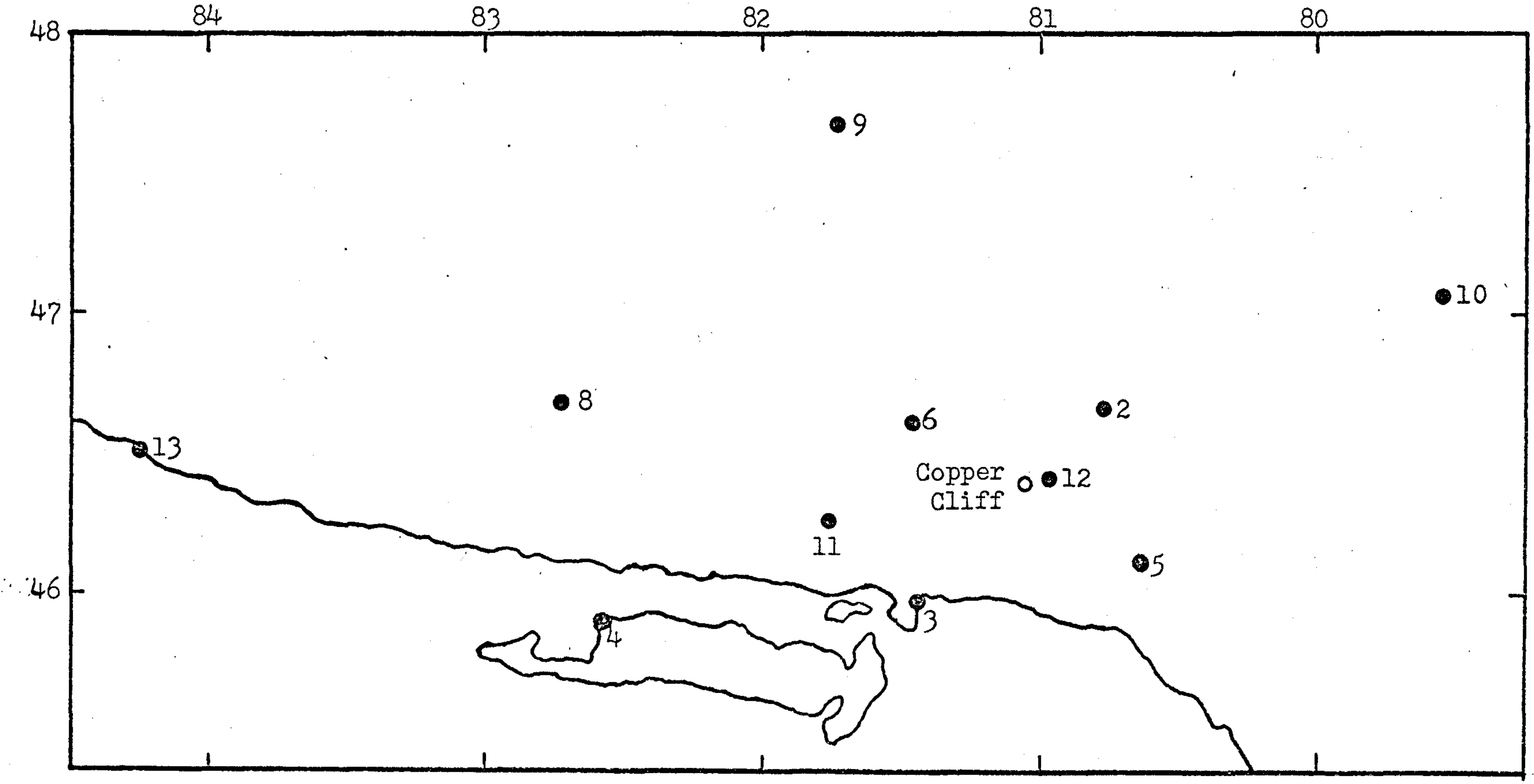


Figure 4.3.2: Precipitation chemistry stations in the Sudbury area. The spatial relationship of stations 12, 2, 3, and 4 to Copper Cliff should be noted. These four stations lie in the path of the predominant northeastward wind direction (Fig. 4.3.1)

The strong correlation between Cu and Ni implies that the two elements undergo a chemically similar cycle from their points of emission. These elements probably form similar complexes of nearly equal stability. Thus it may be possible to predict the behaviour of one element from the other.

In general, the phase difference between the two parameters are very small at the frequency bands of major periodicities. This inphase relationship is seen for the four individual stations as well as the Sudbury and U.G.L. areas. This would imply a common source of origin for Cu and Ni. Furthermore, this source must be a point source of loading that is little affected from other sources, as required for the inphase relationship.

A trend exists for the intensities of the peaks at the frequency bands of periodicities for the six areas. From the tables which give the intensities in log units, it can be seen that the intensities decrease in a general way from the individual stations to the U.G.L. region, that is 12 > 2 > 3 > 4 > Sudbury area > U.G.L. area, at least for Cu. The Ni data show a similar trend, but to a lesser degree.

In view of the location of the stations, this trend will point to INCO's 1250-foot smoke stacks at Copper Cliff as the source of emission of Cu and Ni. It should be noted that stations 12 and 2 are northwest of the smoke stacks with the former being closer, while stations 4 and 3 are south-east of the smoke stacks with the latter being closer. The variation of the intensities among these four individual stations reflects the predominant northeastward wind direction and further evidence for the smoke stack being the source of emission of Cu and Ni. Thus, even if

TABLE 4.3.1: Cross-correlation properties between Cu T and Ni T

STATION 12

	INTENSITIES (Log. Units)			
T	Cu T	Ni T*	r	Ø
12.7	5.67(m)	----	.34	15.2
9.5	----	5.24(s)	.41	2.8
6.3	5.80(s)	----	.07	-17.5
4.2	----	5.16(s)	.26	-38.0
2.7	5.26(w)	5.24(s)	.52	6.2
2.0	----	5.19(s)	.40	-31.2

STATION 2

	INTENSITIES (Log. Units)			
T	Cu T	Ni T	r	Ø
7.6	----	5.31(s)	.62	2.0
4.8	5.80(s)	5.38(s)	.99	-10.1
3.5	5.64(m)	5.21(m)	.94	8.1
2.1	5.33(w)	5.07(w)	.92	- 9.4

STATION 3

9.0	?4.50(s)	----	.15	52.7
7.2	----	?3.28(s)	.11	11.8
4.5	----	3.43(s)	.30	- 9.6
4.0	3.99(s)	----	.61	-14.2
2.6	----	3.46(s)	.64	- 0.3
2.3	4.04(s)	----	.86	-14.4

STATION 4

8.0	3.48(w)	----	.18	120.5
4.4	----	2.93(s)	.53	23.8
4.0	3.70(s)	----	.82	8.8
3.1	3.45(w)	----	.59	-14.4
2.4	----	3.02(s)	.28	17.3
2.1	3.51(m)	----	.56	11.8
2.0	----	2.67(w)	.71	11.5

SUDBURY

6.7	4.53(s)	----	.56	- 3.5
4.4	----	3.97(s)	.21	-44.4
2.9	4.39(s)	----	.91	3.2
2.7	----	4.22(s)	.75	- 8.0
2.1	4.38(s)	----	.60	22.8

U.G.L.

9.2	4.25(m)	----	.51	- 4.9
7.7	----	4.17(w)	.54	-22.8
5.1	4.14(w)	----	.64	10.9
4.2	----	4.32(s)	.60	25.8
3.8	4.45(s)	----	.67	13.2
3.1	4.08(w)	4.21(s)	.51	- 5.9
2.4	----	4.29(s)	.77	3.4
2.2	4.27(m)	----	.67	17.0

T = Period (months); r = Correlation; Ø = Phase (degrees)

* Ni T data unreliable to a certain extent.

there is loading of Cu and Ni to the atmosphere from sources other than Copper Cliff, the latter dominates the loading cycle for the U.G.L. region.

4.3.2 Ni(T) - Cd (T)

A sympathetic relationship exists between Cd and Ni at station 12 since they are highly correlated throughout the frequency range and also remarkably inphase (Figs. 3.3.31 to 3.3.33). The probable source of Cd and Ni is the effluent from Copper Cliff since station 12 is located in the immediate vicinity.

The above relationship is not seen in any of the other areas analyzed. The results are shown in Figures 3.3.34 to 3.3.48. This inconsistency may be explained by considering the molecular weight of Ni and Cd. The latter is about twice as heavy as the former and it would not be expected therefore, to be dispersed from the smoke stack as far as the Ni. Thus any similarity that exists near the smoke stack will not be the same at any great distance from it.

Total cadmium

The peak intensities at periodicities in the power spectrum for Cd T shows a general trend among the areas studied. Table 4.3.2 shows the intensities of the frequency bands of the periodicities.

The intensities for station 12 are greater than that for station 2, while those for station 3 are greater than that for station 4 in a general way. Furthermore, the intensities for these individual stations are greater than that for the Sudbury area. (The data for Cd T for the U.G.L. region is somewhat questionable, Fig. B.6.1). In view of the station locations, the intensities increase towards Copper Cliff and this points to the smoke stacks as the source of emission of Cd.

TABLE 4.3.2: Intensities for Cd T

STATION 12

PERIOD (months)	INTENSITIES (Log. Units) Cd T
9.0	1.68(s)
4.5	----
4.0	1.49(w)
2.8	----
2.6	1.51(m)
2.0	1.50(m)

STATION 2

PERIOD (months)	INTENSITIES (Log. Units) Cd T
7.6	----
6.3	1.56(s)
4.8	----
3.5	1.48(s)
2.1	1.10(w)

STATION 3

9.0	2.02(s)
4.5	1.99(m)
3.0	2.03(s)
2.6	----
2.4	1.98(m)

STATION 4

9.5	1.56(s)
4.2	----
3.8	1.53(s)
2.4	----
2.2	1.67(s)

SUDBURY

13.3	1.50(s)
5.7	1.43(m)
4.4	----
3.6	1.29(w)
2.9	1.15(w)
2.7	----
2.4	1.01(w)

*Cd T data for U.G.L. area unreliable.

4.3.3 Zn T - Zn F

Striking relationships exist between the total and filtered zinc for the Sudbury and U.G.L. areas. An exceedingly high correlation is seen for the total and filtered zinc and they are practically inphase at all frequencies. These relationships are demonstrated quite well in Figures 3.3.49 to 3.3.54 and imply that the chemistry for the total and filtered zinc are quite similar, and in fact the behaviour of one can be predicted for the other. Consequently, from the point of view of chemical analysis all one needs to do is to analyze one form of zinc and infer the trend for the other.

4.3.4 Cu T and Cu F

The Sudbury and U.G.L. areas are analyzed for the total and filtered Cu. The former shows moderately strong correlation and low phase difference at the frequency bands of the periodicities. A similar relationship is seen for the U.G.L. area but to a lesser degree. Figures 3.3.55 through 3.3.60 show the spectra for the total and filtered copper.

The relationship between total and filtered Cu is not as strong as that of the two forms of zinc. Thus the total and filtered copper have slightly different chemical cycles. However, Cu T and Zn T have a similar behaviour since they have similar periodicities.

4.3.5 pH and T PART

A local relationship exists between pH and T PART. Station 12 shows high correlation and inphase properties for pH and T PART throughout the frequency range of the spectra (Figs. 3.3.61 to 3.3.63). A similar trend is seen at station 2, but to a lesser degree (Figs. 3.3.64 to 3.3.66). However, no such relationship exists for the U.G.L. region as can be seen from Figures 3.3.67 to 3.3.69.

The spectral intensities at periodicities for the parameters decrease in a general way from station 12 to station 2 and are lowest for the U.G.L. region. Thus the pH and T PART show some association with the smoke stack.

The restriction of these parameters to the Sudbury area is expected since particulates are quite heavy and "rains" near its source of origin. Furthermore, on a regional scale as the U.G.L., the pH is buffered by carbonates, etc. and no association between pH and T PART exists on such a scale.

The association between pH and T PART locally is probably due to the surface chemistry of the particulates which have considerable surface area. Presumably, the hydrogen ions attach themselves to the surface of the particulates by electrostatic forces and they are dispersed together as a unit.

4.3.6 pH, SO_4, SP CON

It is a common thought that pH should be related to SO_4 in someway since the latter may be responsible for the low pH in the U.G.L. area. Therefore an intensive analysis is made to investigate the relationship, if any. Four individual stations (12, 2, 3, 4) and also the Sudbury and U.G.L. areas are analyzed and the results are given in Appendix B.

Generally, no relationship is found to exist between the two parameters. This would indicate that the chemistry between the two ions

TABLE 4.3.3: Cross-correlation properties between pH and SO$_4$

STATION 12

	INTENSITIES (Log. Units)			
T	pH	SO$_4$	r	ϕ
14.0	----	2.67(s)	.26	110.8
6.0	1.50(s)	----	.53	-62.6
4.7	----	2.29(m)	.39	10.2
3.2	----	2.47(s)	.06	-110.1
3.0	1.36(s)	----	.16	-131.8
2.6	----	2.28(m)	.37	16.9
2.0	1.31(s)	----	.73	-35.3

STATION 2

	INTENSITIES (Log. Units)			
T	pH	SO$_4$	r	ϕ
12.7	----	2.53(s)	.02	35.0
5.4	0.50(s)	----	.22	80.9
4.8	----	2.31(s)	.26	82.1
2.7	0.45(s)	2.37(s)	.64	92.5
2.2	0.32(m)	----	.32	-162.2

STATION 3

14.0	1.43(s)	----	.44	39.4
7.0	1.26(w)	----	.45	135.4
6.0	----	1.77(s)	.23	114.8
4.2	1.23(w)	----	.41	-32.5
3.5	1.23(w)	----	.89	-15.8
2.6	1.32(m)	----	.53	16.1
2.5	----	1.58(m)	.19	7.1
2.2	1.30(m)	----	.05	-159.6

STATION 4

14.0	----	2.58(s)	.73	.1
8.4	1.51(s)	----	.12	-120.3
3.8	1.37(s)	----	.00	54.9
3.5	----	2.04(m)	.14	- 5.3
2.8	1.20(w)	----	.62	39.0
2.3	1.31(m)	----	.54	-84.2
2.1	----	2.00(m)	.08	58.3

SUDBURY

10.4	----	2.33(s)	.25	94.2
6.5	-.06(s)	----	.14	-152.5
4.7	----	2.26(s)	.40	-96.5
3.7	-.03(s)	----	.54	-121.7
3.1	----	1.39(w)	.60	56.9
2.3	-.11(w)	1.88(s)	.32	102.0

U.G.L.

3.3	.11(w)	1.52(w)	.56	- 8.8
2.4	.41(s)	----	.09	-141.0
2.3	----	2.01(s)	.12	-98.5
2.1	-.09(w)	----	.18	-11.4

T = Period (months); r = Correlation; ϕ = Phase (degrees) is not very simple or that other factors are involved, for example, many sources of the ions may be interfering.

The spectral intensities for the individual stations decrease in a general way from the smoke stack at Copper Cliff. The lowest intensities are associated with the U.G.L. area. This trend can be seen from Table 4.3.3.

Generally, hydrogen ions are considerably more mobile than most of the other ions in solution. Thus it may be related to the super conductivity of the solution. Consequently, pH and SP CON are analyzed for the Sudbury and the U.G.L. areas and the results are given in Appendix B. In general, the correlation is low and they are out of phase. Perhaps, the super conductivity is due to ions other than the hydrogen ions. In fact this is brought out in the light that pH is associated with particulates.

4.3.7 Fe T and Zn T

These two elements are analyzed for four individual stations (12, 2, 3, 4). The spectra are given in Figures B.7.25 to B.7.36. Table 4.3.4 summarizes the frequency bands of periodicities, correlation and phase relationships.

Stations 12 and 2 show little correlation and out of phase relationships between Fe and Zn. However, stations 3 and 4 are quite the opposite as the correlation is high and they are generally inphase. This relationship is strongest for station 4. Furthermore, the spectral intensities do not show any trend among the four stations and the correlation is high for the leeward stations (3,4) with respect to the smoke stack. This suggests that the loading of these elements are not from the smoke stack but from some other source which is probably close to station 4.

TABLE 4.3.4: Cross-correlation properties between Fe T and Zn T

STATION 12

T	INTENSITIES (Log. Units)		r	ϕ
	Fe T	Zn T		
8.4	----	6.23(s)	.43	128.6
6.0	7.30(m)	----	.55	-72.6
4.2	----	6.08(m)	.31	57.9
3.8	7.22(m)	----	.30	52.5
3.0	7.39(s)	----	.39	-143.2
2.6	----	6.02(m)	.33	40.4
2.5	7.17(w)	----	.50	41.5
2.1	7.31(m)	----	.51	-160.6
2.0	----	6.07(m)	.61	-167.0

STATION 2

T	INTENSITIES (Log. Units)		r	ϕ
	Fe T	Zn T		
12.0	6.91(s)	----	.02	-132.1
9.0	----	6.24(s)	.18	-128.6
5.1	6.92(s)	----	.08	150.8
4.0	----	6.25(s)	.10	73.2
3.3	6.80(m)	----	.44	-169.3
2.8	----	6.26(s)	.56	-11.5
2.6	6.77(m)	----	.39	19.6
2.1	6.57(w)	----	.27	-37.5
2.0	----	6.32(s)	.54	-13.7

STATION 3

T	Fe T	Zn T	r	ϕ
8.4	5.43(m)	----	.26	88.7
5.3	----	6.38(s)	.14	- 3.8
4.2	5.68(s)	----	.60	-15.5
2.8	5.70(s)	----	.57	35.4
2.5	----	6.33(s)	.58	-40.4
2.1	5.41(m)	----	.43	-56.1

STATION 4

T	Fe T	Zn T	r	ϕ
14.0	6.60(s)	----	.64	17.4
10.5	----	6.36(m)	.53	-15.6
6.0	6.32(m)	----	.60	19.2
3.8	6.23(m)	6.30(m)	.82	13.9
3.0	6.27(m)	----	.82	15.6
2.8	----	6.21(m)	.75	3.7
2.3	5.88(w)	----	.78	- 6.4

4.4 Summary

1. All the parameters considered in this study have periodicities in the range 2 to 5 cycles/month. In addition to these periodicities, they also have higher periodicities which are as follows:

(a) 9 cycles/month for Cu, Ni, Cd, Pb, and Zn;
(b) 12 cycles/month for Fe, PART, P, and SO_4;
(c) 6 cycles/month for pH, Fe, PART, and P.

2. The yearly cycle corresponds to high and low atmospheric loadings in the summer and winter respectively; the 6 monthly period reflects the four seasons of the year; the 9 monthly period is possibly associated with the smelting and mining industry at Sudbury and the 2 to 5 cycles/month periodicities are due to atmospheric turbulence.

3. Good correlation and low phase difference are found between the following parameters:

(a) Cu T and Ni T at stations 12, 2, 3, 4, Sudbury and U.G.L. areas;
(b) Cd T and Ni T at station 12;
(c) Zn T and Zn F for Sudbury area and U.G.L. areas;
(d) Cu T and Cu F for Sudbury area; and
(e) pH and PART at stations 12 and 2.

4. The mining and smelting industry at Sudbury has been established by this work as a major point source of atmospheric emissions of acid, sulfate and the heavy metals, with the exception of iron. This phenomena is well-substantiated by (i) the pattern of the heavy metal periodicities; (ii) the general correlation and phase relationships given in 3. above and (iii) the general decrease in spectral intensities at major periodicities, away from the mining-smelting centre.

5. Semkin and Kramer (1976) found that the Sudbury-area lakes effectively act as a sink for heavy metals that have been introduced via atmospheric precipitation. Their evidence for this phenomena has been well-substantiated by observing heavy metal trends in both lake sediment and atmoshperic fall-out. This conclusion has been arrived at since their results indicate (i) similar metal ratios in both media; (ii) increasing concentration gradients for the trace elements in sediments and atmospheric precipitation toward the mining-smelting centre; and (iii) significant correlation of the sediment parameters, suggesting a point source emission of heavy metals.

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APPENDIX A

SPECTRAL ANALYSIS PROGRAM

COMPUTER PROGRAM

```
      PROGRAM OFF(INPUT,OUTPUT,TAPE1,TAPE2,TAPE3,PUNCH,
     $TAPE5=INPUT,TAPE6=OUTPUT)
      DIMENSION ICOD(200),FLA(200),VAL(200)
      DIMENSION DAT1(2,485),DAT2(2,485)
      DIMENSION IXX(100)
      COMMON/NAT/NAM(200),INO(200),ZNAM(200),ND
      COMMON/PLT/XNIS(485),FACTOR,AINTER
      NNO=0
      NVAL1=NVAL2=0
      NTX=0
      VALX=-1.0
      VALY=-1.0
      NTEST=7111
      NO=0
      I10=10**10
      DO 40 I=1,ND
 40   IXX(I)=1H.
      READ 100,ICD1,ICD2
100   FORMAT(2I5)
999   READ(1)NTAG,NSET,ICOM
      IF(NTAG.EQ.99)GO TO 26
      NTAG=NTAG/I10
      IF(NTAG .LT. NTEST)GO TO 20
      IF(NTX.EQ.0)GO TO 10
 98   IF(NTAG-NTX)1,20,1
  1   I2=I2+1
      IF(I2-13)26,2,99
  2   I2=1
      I1=I1+1
      GO TO 26
 10   I1=NTAG/100
      I2=NTAG-I1*100
      NTX=I1*100+I2
 11   IF(NTAG-NTX)31,12,99
 12   VAL1=VAL2=0.0
      IC1=IC2=0
 20   DO 21 I=1,NSET
        READ(1)INST,NV,(ICOD(J),FLA(J),VAL(J),J=1,NV)
 21   CONTINUE
      IF(NTAG .LT. NTEST)GO TO 254
      DO 25 J=1,NV
        IF(FLA(J) .NE. 1H )GO TO 25
      DO 42 I=1,ND
      IF(NAM(I)-ICOD(J))42,43,44
 43   IXX(I)=1HX
      GO TO 44
 42   CONTINUE
 44   CONTINUE
      IF(ICOD(J)-ICD1)23,22,23
 22     VAL1=VAL1+VAL(J)
        IC1=IC1+1
      GO TO 25
 23   IF(ICOD(J)-ICD2)25,24,25
24    VAL2=VAL(J)+VAL2
        IC2=IC2+1
 25   CONTINUE
254   IF(ICOM)999,999,255
255   DO 256 I=1,ICOM
        READ(1)
256   CONTINUE
```

```
      GO TO 999
 25   PRINT 102,NTAG,IXX
 102  FORMAT(1X,I4,1X,103A1)
      DO 41 I=1,ND
 41   IXX(I)=1H.
 28   IF(IC1)51,51,50
 50   NVAL1=0
      VAL1=VAL1/FLOAT(IC1)
      GO TO 52
 51   NVAL1=NVAL1+1
      VAL1=VALX
 52   VALX=VAL1
      IF(IC2)54,54,53
 53   NVAL2=0
      VAL2=VAL2/FLOAT(IC2)
      GO TO 55
 54   NVAL2=NVAL2+1
      VAL2=VALY
 55    VALY=VAL2
      IF(NVAL1-2)56,56,58
 56   IF(NVAL2-2)57,57,59
 58   NO=NO-NVAL1+1
      GO TO 57
 59   NO=NO-NVAL2+1
 57   NTX=I1*100+I2
      NNO=NNO+1
      NO=NO+1
      XNIS(NO)=NTX
      DAT1(1,NO)=VAL1
      DAT2(1,NO)=VAL2
      IF(NTAG .EQ. 99)GO TO 99
      GO TO 11
 31   IC1=IC2=0
      GO TO 1
   99 PRINT 101,NO
101   FORMAT(1X,I10, 2(I5,F10.2))
C     USE CM. AS SCALE.
      CALL PLOT(5.,0.,-20)
      CALL LETTER(10,.5,90.,1.,1.,10HHASSAN,I.  )
      CALL PLOT(2.,0.,-3)
      CALL FOUR(NO,DAT1.DAT2,ICD1,ICD2)
      CALL PLOT(0.,0.,999)
      STOP
      END


      SUBROUTINE FOUR (N,DAT1,DAT2,ICD1,ICD2)
      DIMENSION DAT(2,1),DAT2(2,1)
      DELTAT=1.0
      NUMSER=2
      LAGS=N/2
 2    FORMAT(F5.2,I5,I5,A10,2F5.0,3I5)
      IF(LAGS.GT.200) GO TO 9
      CALL INPUT(DAT1,N,DELTAT,NWORK,ICD1)
      IF( N.EQ.0) GO TO 9
      BW=FLOAT(NWORK)/(2.0*FLOAT(LAGS))
      WRITE(6,3)N,LAGS,DELTAT
      IF( NUMSER-2)4,5,11
 4    CALL SPEC1 (DAT1,NWORK/2,DELTAT,LAGS,BW )
      GO TO 99
 5    CALL SPEC2(DAT1,DAT2,N,DELTAT,LAGS,BW,ICD2,ICD1)
      GO TO 99
 9    WRITE(6,10)
      GO TO 99
 11   WRITE(6,12)
```

```
 3    FORMAT(1H1,10X,             * N = *,I5,5X,* LAGS = *,
     $I5,5X,* TIME INTERVAL = ERVAL = *,F5.2)
 12   FORMAT(*    JOB TERMINATED ..NUMSER GREATER THAN 2 *)
 10   FORMAT(*  JOB TERMINATED ..LAG TOO LARGE *)
   99 RETURN
      END



      SUBROUTINE INPUT(DATA,N,DELTAT,NWORK,ICD)
      DIMENSION DATA(2,1),NN(1),WORK(200)
      DIMENSION A(3),B(2),X(100),Y(100)
      COMMON/PLT/XNIS(485),FACTOR,AINTER
C.....FOR PH ONLY.
      IF(ICD .NE.123)GO TO 13
      DO 14 I=1,N
      DATA(1,I)=-ALOG(DATA(1,I))
 14   CONTINUE
 13   DO 1 I=1,N
      Y(I)=DATA(1,I)
      X(I)=FLOAT(I-1)
 1    CONTINUE
      CALL LESQ(A,B,X,Y,1,N)
      AINTER=B(1)
      SLOPE=B(2)
      FACTOR = SLOPE*DELTAT
      DO 10 I=1,N
      WRITE(6,11)DATA(1,I)
 10     CONTINUE
  11  FORMAT(F10.2)
      PRINT 12,ICD,N
 12     FORMAT(2I10)
      CALL PLOTT(DATA,N,ICD)
      DO 3 I=1,N
3     DATA(1,I) = DATA(1,I)-AINTER-FACTOR*FLOAT(I)
      PRINT 100 ,AINTER,FACTOR
100   FORMAT(1X,*INTERCEPT=*,F10.5,*  FACTOR=*,F10.5)
      DO 2 I=1,N
      IF(DATA(1,I) .LT. 0.0) DATA(1,I)=0.0
      PRINT 101,DATA(1,I)
 2    CONTINUE
 101  FORMAT(1X,F10.2)
      CALL PLOTT(DATA,N,ICD)
      NWORK = 2
4     IF(NWORK-N) 9,7,5
9     NWORK = 2*NWORK
      GO TO 4
5     N1 = N+1
      DO 6 I=N1,NWORK
6     DATA(1,I) = 0.0
7     DO 8 I=1,NWORK
8     DATA(2,I) = 0.0
      NN(1) = NWORK
      CALL FOURT(DATA,NN,1,-1,0,WORK)
      RETURN
      END



      SUBROUTINE HANN(A,LAGS)
      DIMENSION A(1)
      T1=A(1)
      A(1) = (0.5*(T1+A(2)))
      K=LAGS-1
      DO 1 I=2,K
```

```
      T2 = A(I)
      A(I) = 0.5*T2 + 0.25*(T1+A(I+1))
 1    T1 = T2
      A(LAGS) = (0.5*(T1+A(LAGS)))
      RETURN
      END



      SUBROUTINE SPEC1(DATA,N,DELTAT,NDIM,BW)
      DIMENSION DATA(2, 485),WORK(200)
      FACTOR = 0.5/(FLOAT(NDIM)*DELTAT)
      LAGS=NDIM+1
      WRITE(6,1)
 1    FORMAT(1H0,45X,*LAG*,7X,*CR-CO*,7X,*PERIOD*,2X,
     $*LOG10(SPECTRUM)*/)
      K=0
      P = 0.0
      N=N+1
      DO 6 I=1,LAGS
      WORK(I) = 0.0
      J=K+1
      IF(J-1) 13,13,2
 13   P = BW/2.0+1
      K= IFIX(P)
      GO TO 4
 2    P = P+BW
      IF(P-N) 14,14,3
 14   K = IFIX(P+0.5)
      GO TO 4
 3    K=N
 4    DO 5 L=J,K
 5    WORK(I) = WORK(I)+DATA(1,L)*DATA(1,L)+DATA(2,L)*DATA(2,L)
 6    WORK(I) = WORK(I)/FLOAT(K-J+1)
      CALL HANN(WORK,LAGS)
      DO 7 I=1,LAGS
 7    WORK(I) = ALOG10(WORK(I))
      DO 10 I=1,LAGS
      FREQ = FLOAT(I-1)*FACTOR
      IF(I-1) 15,15,8
 15   PERIOD=999.9999
      GO TO 9
 8    PERIOD = 1.0/FREQ
 9    J=I-1
 10   WRITE(6,11) J,FREQ,PERIOD,WORK(I)
 11   FORMAT(43X,I5,3(4X,F8.4))
      WRITE(3,12)  (WORK(I),I=1,LAGS)
 12   FORMAT(10F8.4)
      RETURN
      END


      SUBROUTINE SPEC2(DATA1,DATA2,N,DELTAT,NDIM,BW,ICD2,ICD1)
      DIMENSION DATA1(2,1),DATA2(2,1)
      DIMENSION COSP(200),QUASP(200),SPECT1(200),SPECT2(200)
      DIMENSION SPECTD(2,485)
      DIMENSION SPD2(2,485)
      COMMON/POT/YPHASE(50) ,YCOHER(50)
      CALL INPUT(DATA2,N,DELTAT,NWORK,ICD2)
      FACTOR = 0.5/(FLOAT(NDIM)*DELTAT)
      N=NWORK/2+1
      LAGS=NDIM+1
      K=0
      P = 0.0
      DO 5 I=1,LAGS
      COSP(I) = 0.0
```

```
      QUASP(I) = 0.0
      SPECT1(I) = 0.0
      SPECT2(I) = 0.0
      J=K+1
      IF(J-1) 1,14,1
14    P=BW/2.0+1
      K = IFIX(P)
      GO TO 3
1     P = P+BW
      IF(P-N) 13,13,2
13    K = IFIX(P+0.5)
      GO TO 3
2     K = N
3     DO 4 L=J,K
      COSP(I) = COSP(I)+DATA1(1,L)*DATA2(1,L)
     $+DATA1(2,L)*DATA2(2,L)
      QUASP(I)=QUASP(I)+DATA1(1,L)*DATA2(2,L)
     $-DATA1(2,L)*DATA2(1,L)
      SPECT1(I) = SPECT1(I)+DATA1(1,L)*DATA1(1,L)
     $+DATA1(2,L)*DATA1(2,L)
 4    SPECT2(I) = SPECT2(I)+DATA2(1,L)*DATA2(1,L)
     $+DATA2(2,L)*DATA2(2,L)
      IF(K-J+1)35,35,36
 35   FACT=0.
      GO TO 37
 36   FACT=1.0/FLOAT(K-J+1)
 37   CONTINUE
      COSP(I)=COSP(I)*FACT
      QUASP(I) = QUASP(I)*FACT
      SPECT1(I) = SPECT1(I)*FACT
      SPECT2(I) = SPECT2(I)*FACT
 5    CONTINUE
      CALL HANN(SPECT1,LAGS)
      CALL HANN(SPECT2,LAGS)
      CALL HANN(QUASP,LAGS)
      CALL HANN(COSP,LAGS)
      WRITE(6,20)
 20   FORMAT(1H1)
      WRITE(6,6)
 6    FORMAT(*0    LAG*,3X,*FREQ*,4X,*PERIOD*,5X,*LOG(SPEC1)*,
     $2X,*LOG(SPEC2)*,3X,8HCOHER**2,6X,*PHASE*,
     $10X,*CO-SPEC*,9X,*QUA-SPEC*//)
      DO 10 I=1,LAGS
      COHER = (COSP(I)*COSP(I)+QUASP(I)*QUASP(I))/(SPECT1(I)
     $*SPECT2(I))
      YCOHER(I)=COHER
      PHASE = ATAN2(QUASP(I),COSP(I))*57.29578
      YPHASE(I)=PHASE
      SPECT1(I) = ALOG10(SPECT1(I))
      SPECT2(I) = ALOG10(SPECT2(I))
      FREQ = FLOAT(I-1)*FACTOR
      J=I-1
      IF(I-1) 12,12,7
12    PERIOD = 999.9999
      GO TO 8
7     PERIOD = 1.0/FREQ
 8    WRITE(6,9)  J,FREQ,PERIOD,SPECT1(I),SPECT2(I),COHER,
     $PHASE,COSP(I),QUASP(I)
 9    FORMAT(1H0,I5,2(F8.4,2X),3(3X,F9.5),3X,F10.5,2(3X,E15.9))
 10   WRITE(3,11)FREQ,SPECT1(I),SPECT2(I),COHER,PHASE,
     $COSP(I),QUASP(I)
 11   FORMAT(5E12.5,2E10.3)
      DO 100 I=1,LAGS
      SPECTD(1,I)=SPECT1(I)
100   CONTINUE
      CALL PLOTT(SPECTD,LAGS,ICO1)
      DO 101 I=1,LAGS
      SPC2(1,I)=SPECT2(I)
```

```
 101   CONTINUE
       CALL PLOTT(SPC2,LAGS,ICD2)
       RETURN
       END


       SUBROUTINE FOURT(DATA,NN,NDIM,ISIGN,IFORM,WORK)
       DIMENSION DATA(1),NN(1),IFACT(32),WORK(1)
       RTHLF = 0.7071067812
       TWOPI=6.283185307
       IF(NDIM-1)920,1,1
 1     NTOT=2
       DO 2 IDIM=1,NDIM
       IF(NN(IDIM))920,920,2
 2     NTOT=NTOT*NN(IDIM)
       ISAVE = NTOT
       NP1=2
       DO 910 IDIM=1,NDIM
       N=NN(IDIM)
       NP2=NP1*N
       IF(N-1)920,900,5
 5     M=N
       NTWO=NP1
       IF=1
       IDIV=2
 10    IQUOT=M/IDIV
       IREM=M-IDIV*IQUOT
       IF(IQUOT-IDIV)50,11,11
 11    IF(IREM) 20,12, 20
 12    NTWO=NTWO+NTWO
       IFACT(IF) = IDIV
       IF = IF+1
       M=IQUOT
       GO TO 10
 20    IDIV=3
       INON2 = IF
 30    IQUOT=M/IDIV
       IREM=M-IDIV*IQUOT
       IF(IQUOT-IDIV)60,31,31
 31    IF(IREM)40,32,40
 32    IFACT(IF)=IDIV
       IF=IF+1
       M=IQUOT
       GO TO 30
 40    IDIV=IDIV+2
       GO TO 30
 50    INON2 = IF
       IF(IREM) 60,51,60
  51   NTWO=NTWO+NTWO
       GO TO 70
 60    IFACT(IF)=M
 70    ICASE = 1
       IFMIN = 1
       I1RNG = NP1
       IF(IDIM-1) 71,100,100
 71    IF(IFORM)72,72,100
 72    IF(IDIM-1) 73,74,73
 73    ICASE = 2
       I1RNG = NP0*(1+NPREV/2)
       GO TO 100
 74    IF(NTWO-NP1) 75,75,76
 75    ICASE = 3
       GO TO 100
 76    ICASE = 4
       IFMIN=2
       NTWO=NTWO/2
```

```
      N=N/2
      NP2=NP2/2
      NTOT=NTOT/2
      I = 3
      DO 80 J=2,NTOT
      DATA(J)=DATA(I)
80    I=I+2
100   IF(NTWO-NP2)200,110,110
110   NP2HF=NP2/2
      J=1
      DO 150 I2=1,NP2,NP1
      IF(J-I2)120,130,130
120   I1MAX=I2+NP1-2
      DO 125 I1=I2,I1MAX,2
      DO 125 I3=I1,NTOT,NP2
      J3=J+I3-I2
      TEMPR=DATA(I3)
      TEMPI=DATA(I3+1)
      DATA(I3)=DATA(J3)
      DATA(I3+1)=DATA(J3+1)
      DATA(J3)=TEMPR
125   DATA(J3+1)=TEMPI
130   M=NP2HF
140   IF(J-M)150,150,145
145   J=J-M
      M=M/2
      IF(M-NP1)150,140,140
150   J=J+M
      GO TO 300
200   NWORK = 2*N
      DO 270 I1=1,NP1,2
      DO 270 I3=I1,NTOT,NP2
      J = I3
      DO 260 I=1,NWORK,2
      IF(ICASE-3)210,220,210
210   WORK(I) = DATA(J)
      WORK(I+1)=DATA(J+1)
      GO TO 230
220   WORK(I) = DATA(J)
      WORK(I+1) = 0.0
 230  IFP2 = NP2
      IF = IFMIN
240   IFP1 = IFP2/IFACT(IF)
      J = J+IFP1
      IF(J-I3-IFP2) 260,250,250
250   J = J-IFP2
      IFP2 = IFP1
      IF = IF+1
      IF(IFP2-NP1) 260,260,240
260   CONTINUE
      I2MAX = I3+NP2-NP1
      I = 1
      DO 270 I2=I3,I2MAX,NP1
      DATA(I2) = WORK(I)
      DATA(I2+1) = WORK(I+1)
270   I = I+2
300   IF(NTWO-NP1) 600,600,305
305   NP1TW = NP1+NP1
      IPAR=NTWO/NP1
310   IF(IPAR-2)350,330,320
320   IPAR=IPAR/4
      GO TO 310
330   DO 340 I1=1,I1RNG,2
      DO 340 K1=I1,NTOT,NP1TW
      K2 = K1+NP1
      TEMPR=DATA(K2)
      TEMPI=DATA(K2+1)
      DATA(K2)=DATA(K1)-TEMPR
```

```
      DATA(K2+1)=DATA(K1+1)-TEMPI
      DATA(K1)=DATA(K1)+TEMPR
340   DATA(K1+1)=DATA(K1+1)+TEMPI
350   MMAX=NP1
360   IF(MMAX-NTWO/2)370,600,600
370   IF(NP1TW-MMAX/2) 372,375,375
372   LMAX = MMAX/2
      GO TO 377
375   LMAX = NP1TW
377   DO 570 L=NP1,LMAX,NP1TW
      M = L
      IF(MMAX-NP1) 420,420,380
380   THETA=-TWOPI*FLOAT(L)/FLOAT(4*MMAX)
      IF(ISIGN)400,390,390
390   THETA=-THETA
400   WR=COS(THETA)
      WI=SIN(THETA)
410   W2R=WR*WR-WI*WI
      W2I=2.*WR*WI
      W3R=W2R*WR-W2I*WI
      W3I=W2R*WI+W2I*WR
420   DO 530 I1=1,I1RNG,2
      KMIN = I1+IPAR*M
      IF(MMAX-NP1)430,430,440
430   KMIN=I1
440   KDIF=IPAR*MMAX
450   KSTEP=4*KDIF
      IF(KSTEP-NTWO) 460,460,530
460   DO 520 K1=KMIN,NTOT,KSTEP
      K2=K1+KDIF
      K3=K2+KDIF
      K4=K3+KDIF
      IF(MMAX-NP1) 470,470,480
470   U1R=DATA(K1)+DATA(K2)
      U1I=DATA(K1+1)+DATA(K2+1)
      U2R=DATA(K3)+DATA(K4)
      U2I=DATA(K3+1)+DATA(K4+1)
      U3R=DATA(K1)-DATA(K2)
      U3I=DATA(K1+1)-DATA(K2+1)
      IF(ISIGN)471,472,472
471   U4R=DATA(K3+1)-DATA(K4+1)
      U4I=DATA(K4)-DATA(K3)
      GO TO 510
472   U4R=DATA(K4+1)-DATA(K3+1)
      U4I=DATA(K3)-DATA(K4)
      GO TO 510
480   T2R=W2R*DATA(K2)-W2I*DATA(K2+1)
      T2I=W2R*DATA(K2+1)+W2I*DATA(K2)
      T3R=WR*DATA(K3)-WI*DATA(K3+1)
      T3I=WR*DATA(K3+1)+WI*DATA(K3)
      T4R=W3R*DATA(K4)-W3I*DATA(K4+1)
      T4I=W3R*DATA(K4+1)+W3I*DATA(K4)
      U1R=DATA(K1)+T2R
      U1I=DATA(K1+1)+T2I
      U2R=T3R+T4R
      U2I=T3I+T4I
      U3R=DATA(K1)-T2R
      U3I=DATA(K1+1)-T2I
      IF(ISIGN)490,500,500
490   U4R=T3I-T4I
      U4I=T4R-T3R
      GO TO 510
500   U4R=T4I-T3I
      U4I=T3R-T4R
510   DATA(K1)=U1R+U2R
      DATA(K1+1)=U1I+U2I
      DATA(K2)=U3R+U4R
      DATA(K2+1)=U3I+U4I
```

```
      DATA(K3)=U1R-U2R
      DATA(K3+1)=U1I-U2I
      DATA(K4)=U3R-U4R
  520 DATA(K4+1)=U3I-U4I
      KMIN=4*(KMIN-I1)+I1
      KDIF=KSTEP
      IF(KDIF-NP2HF) 450,530,530
  530 CONTINUE
      M = M+LMAX
      IF(M-MMAX) 540,540,570
  540 IF(ISIGN) 550,560,560
  550 TEMPR=WR
      WR = (WR+WI)*RTHLF
      WI = (WI-TEMPR)*RTHLF
      GO TO 410
   560 TEMPR=WR
      WR = (WR-WI)*RTHLF
      WI = (TEMPR+WI)*RTHLF
      GO TO 410
  570 CONTINUE
      IPAR=3-IPAR
      MMAX=MMAX+MMAX
      GO TO 360
  600 IF(NTWO-NP2)605,700,700
  605 IFP1 = NTWO
      IF = INON2
      NP1HF=NP1/2
   610 IFP2 = IFACT(IF)*IFP1
      J1MIN = NP1+1
      IF(J1MIN-IFP1) 615,615,640
  615 DO 635 J1=J1MIN,IFP1,NP1
      THETA=-TWOPI*FLOAT(J1-1)/FLOAT(IFP2)
      IF(ISIGN)625,620,620
  620 THETA=-THETA
  625 WSTPR = COS(THETA)
      WSTPI=SIN(THETA)
      WR=WSTPR
      WI=WSTPI
      J2MIN = J1+IFP1
      J2MAX = J1+IFP2-IFP1
      DO 635 J2=J2MIN,J2MAX,IFP1
      I1MAX=J2+I1RNG-2
      DO 630 I1=J2,I1MAX,2
      DO 630 J3=I1,NTOT,IFP2
      TEMPR=DATA(J3)
      DATA(J3)=DATA(J3)*WR-DATA(J3+1)*WI
  630 DATA(J3+1)=TEMPR*WI+DATA(J3+1)*WR
      TEMPR=WR
      WR=WR*WSTPR-WI*WSTPI
  635 WI=TEMPR*WSTPI+WI*WSTPR
   640 THETA=-TWOPI/FLOAT(IFACT(IF))
      IF(ISIGN)650,645,645
  645 THETA=-THETA
  650 WSTPR = COS(THETA)
      WSTPI=SIN(THETA)
      J2RNG = IFP1*(1+IFACT(IF)/2)
      DO 695 I1=1,I1RNG,2
      DO 695 I3=1,NTOT,NP2
      J2MAX = I3+J2RNG-IFP1
      DO 690 J2=I3,J2MAX,IFP1
      J1MAX = J2+IFP1-NP1
      DO 660 J1=J2,J1MAX,NP1
      J3MAX=J1+NP2-IFP2
      DO 680 J3=J1,J3MAX,IFP2
      JMIN = J3-J2+I3
      JMAX = JMIN+IFP2-IFP1
      I = 1+(J3-I3)/NP1HF
      IF(J2-I3) 655,655,565
```

```
655     SUMR=0.
        SUMI=0.
        DO 660  J=JMIN,JMAX,IFP1
        SUMR=SUMR+DATA(J)
660     SUMI=SUMI+DATA(J+1)
        WORK(I)=SUMR
        WORK(I+1)=SUMI
        GO TO 680
665     ICONJ = 1+(IFP2-2*J2+I3+J3)/NP1HF
        J=JMAX
        SUMR=DATA(J)
        SUMI=DATA(J+1)
        OLDSR=0.
        OLDSI=0.
        J=J-IFP1
670     TEMPR=SUMR
        TEMPI=SUMI
        SUMR=TWOWR*SUMR-OLDSR+DATA(J)
        SUMI=TWOWR*SUMI-OLDSI+DATA(J+1)
        OLDSR=TEMPR
        OLDSI=TEMPI
        J = J-IFP1
        IF(J-JMIN) 675,675,670
675     TEMPR=WR*SUMR-OLDSR+DATA(J)
        TEMPI=WI*SUMI
        WORK(I)=TEMPR-TEMPI
        WORK(ICONJ)=TEMPR+TEMPI
        TEMPR=WR*SUMI-OLDSI+DATA(J+1)
        TEMPI=WI*SUMR
        WORK(I+1)=TEMPR+TEMPI
        WORK(ICONJ+1)=TEMPR-TEMPI
680     CONTINUE
        IF(J2-I3) 685,685,686
685     WR=WSTPR
        WI=WSTPI
        GO TO 690
686     TEMPR=WR
        WR=WR*WSTPR-WI*WSTPI
        WI=TEMPR*WSTPI+WI*WSTPR
690     TWOWR=WR+WR
        I = 1
        I2MAX = I3+NP2-NP1
        DO 695 I2=I3,I2MAX,NP1
        DATA(I2)=WORK(I)
        DATA(I2+1)=WORK(I+1)
695     I = I+2
        IF = IF+1
        IFP1 = IFP2
        IF(IFP1-NP2) 610,700,700
700     GO TO (900,800,900,701),ICASE
701     NHALF=N
        N=N+N
        THETA=-TWOPI/FLOAT(N)
        IF(ISIGN)703,702,702
702     THETA=-THETA
703     WSTPR = COS(THETA)
        WSTPI=SIN(THETA)
        WR=WSTPR
        WI=WSTPI
        IMIN=3
        JMIN=2*NHALF-1
        GO TO 725
710     J=JMIN
        DO 720  I=IMIN,NTOT,NP2
        SUMR=(DATA(I)+DATA(J))/2.
        SUMI=(DATA(I+1)+DATA(J+1))/2.
        DIFR=(DATA(I)-DATA(J))/2.
        DIFI=(DATA(I+1)-DATA(J+1))/2.
```

```
      TEMPR=WR*SUMI+WI*DIFR
      TEMPI=WI*SUMI-WR*DIFR
      DATA(I)=SUMR+TEMPR
      DATA(I+1)=DIFI+TEMPI
      DATA(J)=SUMR-TEMPR
      DATA(J+1)=-DIFI+TEMPI
720   J=J+NP2
      IMIN=IMIN+2
      JMIN=JMIN-2
      TEMPR=WR
      WR=WR*WSTPR-WI*WSTPI
      WI=TEMPR*WSTPI+WI*WSTPR
 725  IF(IMIN-JMIN)710,730,740
 730  IF(ISIGN)731,740,740
731   DO 735 I=IMIN,NTOT,NP2
735   DATA(I+1)=-DATA(I+1)
740   NP2=NP2+NP2
      NTOT=NTOT+NTOT
      J=NTOT+1
      IMAX=NTOT/2+1
745   IMIN=IMAX-2*NHALF
      I=IMIN
      GO TO 755
750   DATA(J)=DATA(I)
      DATA(J+1)=-DATA(I+1)
755   I=I+2
      J=J-2
      IF(I-IMAX)750,760,760
760   DATA(J)=DATA(IMIN)-DATA(IMIN+1)
      DATA(J+1)=0.
      IF(I-J)770,780,780
765   DATA(J)=DATA(I)
      DATA(J+1)=DATA(I+1)
770   I=I-2
      J=J-2
      IF(I-IMIN)775,775,765
775   DATA(J)=DATA(IMIN)+DATA(IMIN+1)
      DATA(J+1)=0.
      IMAX=IMIN
      GO TO 745
780   DATA(1)=DATA(1)+DATA(2)
      DATA(2)=0.
      GO TO 900
800   IF(I1RNG-NP1)805,900,900
805   DO 860 I3=1,NTOT,NP2
      I2MAX=I3+NP2-NP1
      DO 860 I2=I3,I2MAX,NP1
      IMIN=I2+I1RNG
      IMAX=I2+NP1-2
      JMAX=2*I3+NP1-IMIN
      IF(I2-I3)820,820,810
810   JMAX=JMAX+NP2
820   IF(IDIM-2)850,850,830
830   J=JMAX+NP0
      DO 840 I=IMIN,IMAX,2
      DATA(I)=DATA(J)
      DATA(I+1)=-DATA(J+1)
840   J=J-2
850   J=JMAX
      DO 860 I=IMIN,IMAX,NP0
      DATA(I)=DATA(J)
      DATA(I+1)=-DATA(J+1)
860   J=J-NP0
900   NP0=NP1
      NP1=NP2
910   NPREV=N
920   IF(ISIGN) 950,950,930
930   FACT= 2.0/FLOAT(ISAVE)
      DO 940 I=1,ISAVE
940   DATA(I) = DATA(I)*FACT
950   RETURN
      END
```

```
      SUBROUTINE PLOTT(D,M,ICD)
      DIMENSION D(2,485),Y(485),X(485)
      DIMENSION IY(20),XLAGS(30)
      DIMENSION LUM(10)
      DIMENSION XPIOD(50)
      DIMENSION CHECK(75),YSP(10)
      COMMON/NAT/NAM(200),INO(200),ZNAM(200),ND
      COMMON/POT/YPHASE(50) ,YCOHER(50)
      COMMON/PLT/XNIS(485),FACTM ,AINTER
      INTEGER Z,ZZ,ZNAM
      DATA ICOUNT/0/
      ICOUNT=ICOUNT+1
      LUM(1)=10HMICRO-GM/L
      LUM(2)=10HX 10(-9) G
      LUM(3)=10HMILLI-GM/L
      LUM(4)=10HX 10(-6)G
      LUM(5)=10HGRAM/L
      LUM(6)=10HX 10(-3)
      LUM(7)=10H
      LUM(8)=10H(EXP) MEQ
      LUM(9)=10HUHMO/CM-25
      LUM(10)=10H
      LOAD=0
      DO 91 I=1,ND
      IF(ICD .NE. NAM(I))GO TO 91
      Z=ZNAM(I)
      JJ=INO(I)-(INO(I)/10)*10+LOAD
      ZZ=LUM(JJ)
      GO TO 92
 91   CONTINUE
 92   CONTINUE
      XMAX=-99.0
      XMIN=10.0**20
      DO 1 I=1,M
      Y(I)=D(1,I)
      XMAX=AMAX1(XMAX,Y(I))
      XMIN=AMIN1(XMIN,Y(I))
 1    CONTINUE
      IF((ICOUNT .EQ. 5) .OR.(ICOUNT .EQ. 6))GO TO 27
      CHECK(1)=0.0
      DO 25 I=2,75
      CHECK(I)=CHECK(I-1)+20.0
      IF(XMAX .LT.  CHECK(I))GO TO 26
 25   CONTINUE
 26   Y(M+2)=CHECK(I) +5.0
      IADD=(Y(M+2)/15.0)*3.0
      Y(M+2)=FLOAT(IADD)*5.0
      X(M+1)=0.0
      X(M+2)=M-1
      Y(M+1)=0.0
      DO 20 L=1,M
      X(L)=L-1
20    CONTINUE
      GO TO 21
 27   Y(M+2)=XMAX
      Y(M+1)=XMIN
      YADD=(XMAX-XMIN)/7.0
      X(M+1)=0.0
      X(M+2)=0.5
      XCM=24.0
      XMARG=4.0
      IF(ICOUNT .EQ. 6)GO TO 28
      YMARG=12.0
      YCM=19.0
      GO TO 9
 28   YCM=11.0
      YMARG=4.0
      GO TO 9
```

```
9       X(1)=0.0
        XDEL=0.5/FLOAT(M-1)
        DO 8 L=2,M
        X(L)=X(L-1)+XDEL
8       CONTINUE
        IF(ICOUNT .EQ. 7)GO TO 10
        IF((ICOUNT .EQ. 5).OR.(ICOUNT .EQ. 6))M=M+2
        IF(ICOUNT .EQ. 8)GO TO 46
        GO TO 45
46      Y(M+1)=0.0
        Y(M+2)=1.5
        DO 47 I=1,M
        Y(I)=YCOHER(I)
47      CONTINUE
        GO TO 21
10      Y(M+1)=270.0
        Y(M+2)=-180.0
        X(M+1)=0
        X(M+2)=.5
        DO 16 I=1,M
        Y(I)=YPHASE(I)
15      CONTINUE
        GO TO 21
21       M=M+2
        XCM=24.0
        YCM=19.0
        XMARG=4.0
        YMARG=4.0
45      CALL FACTOR(M,X,Y,XCM,YCM,XMARG,YMARG)
        IF(ICOUNT .EQ. 6)GO TO 999
        CALL PLOT(4.0,4.0,3)
        CALL PLOT(4.0,4.0,2)
        CALL PLOT(24.0,4.0,2)
        CALL PLOT(24.0,19.0,2)
        CALL PLOT(4.0,19.0,2)
        CALL PLOT(4.0,4.0,2)
        M=M-2
        CALL PLTMSP(X,Y,.12,1HX,0.,M)
        CALL PLTMPL(X,Y,M)
        X2=Y2=4.0
        IPTS=16
        YINCR=1.0
        CALL YCALIB(IPTS,X2,Y2,YINCR)
        IPTS=M
        XM=M-1.0
        XINCR=20.0/XM
        X2=Y2=4.0
        CALL XCALIB(IPTS,X2,Y2,XINCR)
        X2=23.8
        IPTS=16
        YINCR=1.0
        CALL YCALIB(IPTS,X2,Y2,YINCR)
        IF(ICOUNT .EQ. 1    )GO   TO   40
        IF(ICOUNT .EQ. 2    )GO   TO 41
        IF(ICOUNT .EQ. 3)GO TO 40
        IF(ICOUNT .EQ. 4)GO TO 41
        IF(ICOUNT .EQ. 5    )GO   TO 42
        IF(ICOUNT .EQ. 7)GO TO 31
        IF(ICOUNT .EQ. 8)GO TO 31
41      CALL LETTER(17,.3,0.,12.0,18.5,17HDATA-TREND REMOVE)
        CALL LETTER(12,.3,0.,12.0,2.3,12HTIME(MONTHS))
        CALL LETTER(13,.3,90.,3.0,8.0,13HCONCENTRATION)
        PRINT 101,ICO,Z,ZZ
15      IY(1)=0
          IPTS=6
        DO 3 I=2,IPTS
```

```
      IY(I)=IY(I-1)+IADD
3     CONTINUE
23    XX=3.1
      YY=4.0
      THETA=0.0
       HEIGHT=.20
      DO 4 I=1,IPTS
      INUM=IY(I)
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,4H(I4),4)
      YY=YY+3.0
4     CONTINUE
      HEIGHT=.20
      XX=4.0
      YY=3.2
      THETA=90.0
      XNS=XNIS(1)-1.0
      INUM=XNS
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,4H(I4),4)
      XX=XX+6.0*XINCR
      IPTS=M-1
      DO 5 I=1,IPTS,6
      INUM=XNIS(I+5)
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,4H(I4),4)
      IF((IPTS*XINCR+4.0-XX) .LT. 6.0*XINCR)GO TO 22
      XX=XX+6.0*XINCR
5     CONTINUE
22    XX=IPTS*XINCR+4.0
      INUM=XNIS(IPTS)
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,4H(I4),4)
      YY=18.5
      GO TO 7
42    CALL LETTER(29,.3,90.,2.8,7.0,
     $29HRELATIVE SPECTRAL INTENSITIES)
83    IPTS=4
      YSP(1)=XMIN
      DO 11 I=2,IPTS
      YSP(I)=YSP(I-1)+2*YADD
 11    CONTINUE
      XX=3.0
      YY=12.0
      IF(ICOUNT .EQ. 6)YY=4.0
      THETA=0.0
      HEIGHT=.20
      DO 12 I=1,IPTS
      XNUM=YSP(I)
      CALL NUMBER(XX,YY,HEIGHT,XNUM,THETA,6H(F5.3),6)
      YY=YY+2
12    CONTINUE
      XNUM=XMAX
      YY=19.0
      IF(ICOUNT .EQ. 6)YY=11.0
      CALL NUMBER(XX,YY,HEIGHT,XNUM,THETA,6H(F5.3),6)
      IF(ICOUNT .EQ. 6)GO TO 7
31    CALL LETTER(23,.2,0.,12.0,3.0,23HFREQUENCY(CYCLES/MONTH))
      CALL LETTER(14,.2,0.,12.0,2.7,14HPERIOD(MONTHS))
      XLAGS(1)=0.0
      IPTS=M/2+1
      XDEL=0.5/FLOAT(M-1)
      DO 13 I=2,IPTS
      XLAGS(I)=XLAGS(I-1)+2*XDEL
      XPIOD(I)=1.0/(XLAGS(I-1)+XDEL)
13    CONTINUE
      XX=3.6
      YY=3.7
      THETA=0.0
      HEIGHT=.20
      DO 14 I=1,IPTS
      XNUM=XLAGS(I)
```

```
      CALL NUMBER(XX,YY,HEIGHT,XNUM,THETA,6H(F4.3),4 )
      F=FLOAT(IPTS-1)*2.0
      IF((F*XINCR+4.0-XX).EQ.(3.0*XINCR))GOTO 71
      XX=XX+2*XINCR
14    CONTINUE
      GO TO 72
71    XNUM=0.5
      XX=F*XINCR+4.0
      CALL NUMBER(XX,YY,HEIGHT,XNUM,THETA,6H(F4.3),5)
72    XX=4.2
      YY=3.4
      IPTS=IPTS-1
      DO 24 I=2,IPTS
      XNUM=XPIOD(I)
      CALL NUMBER(XX,YY,HEIGHT,XNUM,THETA,6H(F5.2),6)
      XX=XX+2*XINCR
24    CONTINUE
      IF(ICOUNT .EQ. 7)GO TO 32
      IF(ICOUNT .EQ. 8)GO TO 48
      IF(ICOUNT .EQ. 5)YY=11.5
      GO TO 7
40    CALL LETTER(16,.3,0.,12.0,18.5,18HRAW-DATA AND TREND)
      CALL LETTER(12,.3,0.,12.0,2.8,12HTIME(MONTHS))
      CALL LETTER(13,.3,90.,3.0,6.0,13HCONCENTRATION)
      PRINT 101,ICD,Z,ZZ
      X1=0.0
      Y1=AINTER
      X2=M-1
      Y2=FACTM*X2+AINTER
      CALL UNITTO(X1,Y1,XT1,YT1)
      CALL UNITTO(X2,Y2,XT2,YT2)
      CALL PLOT(XT1,YT1,3)
      CALL PLOT(XT2,YT2,2)
      GO TO 15
101   FORMAT(1X,I5,2(2X,A10))
7     INUM=ICD
      XX=5.0
      HEIGHT=.2
      THETA=0.0
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,4H(I3),3)
      IF(ICOUNT .EQ. 6)GO TO 17
      CALL LETTER(2,.2,0.,4.5,YY,2HX )
17    XX=5.8
      INUM=Z
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,4H(A7),7)
      XX=7.4
      INUM=ZZ
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,5H(A10),10)
      IF(ICOUNT .EQ. 5)RETURN
      CALL PLOT(28.,0.,-3)
      IF(ICOUNT .EQ. 6)ICOUNT=7
      IF(ICOUNT .EQ. 7)GO TO 9
      RETURN
999   M=M-2
      CALL PLTMSP(X,Y,.12,1H+,0.,M)
      CALL PLTMPL(X,Y,M)
      CALL LETTER(2,.2,0.,4.5,11.0,2H+ )
      GO TO 83
32    CALL LETTER(5,.3,90.,3.0,10.0,5HPHASE)
      CALL LETTER(14,.3,0.,12.,18.,14HPHASE SPECTRUM)
      IPTS=13
      IY(1)=-180
      DO 33 I=2,IPTS
      IY(I)=IY(I-1)+30
33    CONTINUE
      XX=3.1
      YY=4.0
      HEIGHT=.20
```

```
      THETA=0.0
      DO 34 I=1,IPTS
      INUM=IY(I)
      CALL NUMBER(XX,YY,HEIGHT,INUM,THETA,4H(I4),4)
      YY=YY+1.0
34    CONTINUE
      CALL PLOT(4.0,10.0,3)
      CALL PLOT (24.0,10.0,2)
      CALL PLOT(28.,0.,-3)
      IF(ICOUNT .EQ. 7)ICOUNT=8
      IF(ICOUNT .EQ. 8)GO TO 9
48    CALL LETTER(9,.3,90.,3.0,10.0,9HCOHERENCE)
      IPTS=11
      CALL LETTER(26,.3,0.,11.,18.,
     $26HSQUARED COHERENCY SPECTRUM)

      YSP(1)=0.0
      DO 81 I=2,IPTS
      YSP(I)=YSP(I-1)+0.1
81    CONTINUE
      YY=4.0
      DO 82 I=1,IPTS
      XNUM=YSP(I)
      CALL NUMBER(3.1,YY,.2,XNUM,0.0,6H(F4.2),4)
      YY=YY+1.0
82     CONTINUE
      CALL PLOT(28.,0.,-3)
99    RETURN
      END



      SUBROUTINE XCALIB(IPTS,X2,Y2,XINCR)
      DO 1 I=1,IPTS
      IFLAG=3
      CALL PLOT(X2,Y2,IFLAG)
      Y2=Y2+0.2
      IFLAG=2
      CALL PLOT(X2,Y2,IFLAG)
      X2=X2+XINCR
      Y2=Y2-0.2
1     CONTINUE
      RETURN
      END



      SUBROUTINE YCALIB(IPTS,X2,Y2,YINCR)
      DO 1 I=1,IPTS
      IFLAG=3
      CALL PLOT(X2,Y2,IFLAG)
      X2=X2+0.2
      IFLAG=2
      CALL PLOT(X2,Y2,IFLAG)
      Y2=Y2+YINCR
      X2=X2-0.2
1     CONTINUE
      RETURN
      END
```

```
      SUBROUTINE NUMBER(X,Y,HEIGHT,INUM,THETA,FMT,IPTS)
      DIMENSION BCD(1)
      ENCODE(IPTS,FMT,BCD)INUM
      CALL SYMBOL(X,Y,HEIGHT,BCD,THETA,IPTS)
      RETURN
      END


      BLOCK DATA
C     10 NO. REFER TO NUMBERS BEYOND DECIMAL.
C     1 NO. COLUMN DETERMINE UNIT.
      COMMON/NAT/NAM(200),INO(200),ZNAM(200),ND
      DATA ZNAM(  1),NAM(  1),INO(  1)/7H   CD F,100,11/
      DATA ZNAM(  2),NAM(  2),INO(  2)/7H   CD T,101,11/
      DATA ZNAM(  3),NAM(  3),INO(  3)/7H   CD P,102,11/
      DATA ZNAM(  4),NAM(  4),INO(  4)/7H   CR F,103,11/
      DATA ZNAM(  5),NAM(  5),INO(  5)/7H   CR T,104,11/
      DATA ZNAM(  6),NAM(  6),INO(  6)/7H   CR P,105,11/
      DATA ZNAM(  7),NAM(  7),INO(  7)/7H   CU F,106,11/
      DATA ZNAM(  8),NAM(  8),INO(  8)/7H   CU T,107,11/
      DATA ZNAM(  9),NAM(  9),INO(  9)/7H   CU P,108,11/
      DATA ZNAM( 10),NAM( 10),INO( 10)/7H   FE F,109, 1/
      DATA ZNAM( 11),NAM( 11),INO( 11)/7H   FE T,110, 1/
      DATA ZNAM( 12),NAM( 12),INO( 12)/7H   FE P,111,11/
      DATA ZNAM( 13),NAM( 13),INO( 13)/7H   PB F,112, 1/
      DATA ZNAM( 14),NAM( 14),INO( 14)/7H   PB T,113, 1/
      DATA ZNAM( 15),NAM( 15),INO( 15)/7H   PB P,114,11/
      DATA ZNAM( 16),NAM( 16),INO( 16)/7H   NI F,115, 1/
      DATA ZNAM( 17),NAM( 17),INO( 17)/7H   NI T,116, 1/
      DATA ZNAM( 18),NAM( 18),INO( 18)/7H   NI P,117,11/
      DATA ZNAM( 19),NAM( 19),INO( 19)/7H   ZN F,118,11/
      DATA ZNAM( 20),NAM( 20),INO( 20)/7H   ZN T,119, 1/
      DATA ZNAM( 21),NAM( 21),INO( 21)/7H   ZN P,120,11/
      DATA ZNAM( 22),NAM( 22),INO( 22)/7H     C P,121,33/
      DATA ZNAM( 23),NAM( 23),INO( 23)/7H  SPCON,122, 9/
      DATA ZNAM( 24),NAM( 24),INO( 24)/7H     PH,123,27/
      DATA ZNAM( 25),NAM( 25),INO( 25)/7H      F,124,11/
      DATA ZNAM( 26),NAM( 26),INO( 26)/7H     CL,125, 1/
      DATA ZNAM( 27),NAM( 27),INO( 27)/7H     BR,126,11/
      DATA ZNAM( 28),NAM( 28),INO( 28)/7H    SO4,127,23/
      DATA ZNAM( 29),NAM( 29),INO( 29)/7H T PART,128,35/
      DATA ZNAM( 30),NAM( 30),INO( 30)/7H   CO F,129,11/
      DATA ZNAM( 31),NAM( 31),INO( 31)/7H   CO T,130,11/
      DATA ZNAM( 32),NAM( 32),INO( 32)/7H   CO P,131,11/
      DATA ZNAM( 33),NAM( 33),INO( 33)/7H  CD FA,201,11/
      DATA ZNAM( 34),NAM( 34),INO( 34)/7H  CR FA,202,11/
      DATA ZNAM( 35),NAM( 35),INO( 35)/7H  CU FA,203,11/
      DATA ZNAM( 36),NAM( 36),INO( 36)/7H  NI FA,204, 1/
      DATA ZNAM( 37),NAM( 37),INO( 37)/7H  ZN FA,205,11/
      DATA ZNAM( 38),NAM( 38),INO( 38)/7H  CO FA,206,11/
      DATA ZNAM( 39),NAM( 39),INO( 39)/7H  FE FA,207, 1/
      DATA ZNAM( 40),NAM( 40),INO( 40)/7H  PB FA,208, 1/
      DATA ZNAM( 41),NAM( 41),INO( 41)/7H   MN F,209,11/
      DATA ZNAM( 42),NAM( 42),INO( 42)/7H  MN FA,210,11/
      DATA ZNAM( 43),NAM( 43),INO( 43)/7H   MO F,211,11/
      DATA ZNAM( 44),NAM( 44),INO( 44)/7H  MO FA,212,11/
      DATA ZNAM( 45),NAM( 45),INO( 45)/7H    V F,213,11/
      DATA ZNAM( 46),NAM( 46),INO( 46)/7H   V FA,214,11/
      DATA ZNAM( 47),NAM( 47),INO( 47)/7H   AS T,215,11/
      DATA ZNAM( 48),NAM( 48),INO( 48)/7H   AS F,216,11/
      DATA ZNAM( 49),NAM( 49),INO( 49)/7H  AS FA,217,11/
      DATA ZNAM( 50),NAM( 50),INO( 50)/7H   SE F,218,21/
      DATA ZNAM( 51),NAM( 51),INO( 51)/7H   HG T,219,21/
      DATA ZNAM( 52),NAM( 52),INO( 52)/7H   HG F,220,21/
      DATA ZNAM( 53),NAM( 53),INO( 53)/7H   NA T,221,23/
```

```
DATA ZNAM( 54),NAM( 54),INO( 54)/7H   NA F,222,23/
DATA ZNAM( 55),NAM( 55),INO( 55)/7H    K T,223,23/
DATA ZNAM( 56),NAM( 56),INO( 56)/7H    K F,224,23/
DATA ZNAM( 57),NAM( 57),INO( 57)/7H   MG T,225,13/
DATA ZNAM( 58),NAM( 58),INO( 58)/7H   MG F,226,13/
DATA ZNAM( 59),NAM( 59),INO( 59)/7H   CA T,227,23/
DATA ZNAM( 60),NAM( 60),INO( 60)/7H   CA F,228,23/
DATA ZNAM( 61),NAM( 61),INO( 61)/7H HARD T,229,13/
DATA ZNAM( 62),NAM( 62),INO( 62)/7H HARD F,230,13/
DATA ZNAM( 63),NAM( 63),INO( 63)/7HSOLID F,231,13/
DATA ZNAM( 64),NAM( 64),INO( 64)/7H  ALK F,232,13/
DATA ZNAM( 65),NAM( 65),INO( 65)/7H  P T F,233, 1/
DATA ZNAM( 66),NAM( 66),INO( 66)/7H   N KJ,234,23/
DATA ZNAM( 67),NAM( 67),INO( 67)/7H   ACID,235,13/
DATA ZNAM( 68),NAM( 68),INO( 68)/7H SIO2 F,236,23/
DATA ZNAM( 69),NAM( 69),INO( 69)/7H  NO3 F,237,23/
DATA ZNAM( 70),NAM( 70),INO( 70)/7H   MN P,238,11/
DATA ZNAM( 71),NAM( 71),INO( 71)/7H  SE FA,239,21/
DATA ZNAM( 72),NAM( 72),INO( 72)/7H    P T,240, 1/
DATA ZNAM( 73),NAM( 73),INO( 73)/7H    P R,241, 1/
DATA ZNAM( 74),NAM( 74),INO( 74)/7H    N T,242,33/
DATA ZNAM( 75),NAM( 75),INO( 75)/7H  NO3 R,243,33/
DATA ZNAM( 76),NAM( 76),INO( 76)/7H  NH3 R,244,33/
DATA ZNAM( 77),NAM( 77),INO( 77)/7H   CD R,245,11/
DATA ZNAM( 78),NAM( 78),INO( 78)/7H   CU R,246,11/
DATA ZNAM( 79),NAM( 79),INO( 79)/7H   FE R,247,11/
DATA ZNAM( 80),NAM( 80),INO( 80)/7H   PB R,248,11/
DATA ZNAM( 81),NAM( 81),INO( 81)/7H   NI R,249,11/
DATA ZNAM( 82),NAM( 82),INO( 82)/7H   ZN R,250,11/
DATA ZNAM( 83),NAM( 83),INO( 83)/7H  NH3 F,252,33/
DATA ZNAM( 84),NAM( 84),INO( 84)/7H  P R F,253, 1/
DATA ZNAM( 85),NAM( 85),INO( 85)/7H   AG F,261,11/
DATA ZNAM( 86),NAM( 86),INO( 86)/7H   AG T,262,11/
DATA ZNAM( 87),NAM( 87),INO( 87)/7H   AL F,263,11/
DATA ZNAM( 88),NAM( 88),INO( 88)/7H   AL T,264,11/
DATA ZNAM( 89),NAM( 89),INO( 89)/7H    B F,265,11/
DATA ZNAM( 90),NAM( 90),INO( 90)/7H    B T,266,11/
DATA ZNAM( 91),NAM( 91),INO( 91)/7H   BA F,267,11/
DATA ZNAM( 92),NAM( 92),INO( 92)/7H   BA T,268,11/
DATA ZNAM( 93),NAM( 93),INO( 93)/7H   SN F,269,11/
DATA ZNAM( 94),NAM( 94),INO( 94)/7H   SN T,270,11/
DATA ZNAM( 95),NAM( 95),INO( 95)/7H   TI F,271,11/
DATA ZNAM( 96),NAM( 96),INO( 96)/7H   TI T,272,11/
DATA ZNAM( 97),NAM( 97),INO( 97)/7H    V T,274,11/
DATA ZNAM( 98),NAM( 98),INO( 98)/7H    Y F,275,11/
DATA ZNAM( 99),NAM( 99),INO( 99)/7H    Y T,276,11/
DATA ZNAM(100),NAM(100),INO(100)/7H   MN T,277,11/
DATA ZNAM(101),NAM(101),INO(101)/7H   MO T,278,11/
DATA ZNAM(102),NAM(102),INO(102)/7HPFORTHO,279,11/
DATA ZNAM(103),NAM(103),INO(103)/7H  ALK T,280,13/
DATA ND/103/
END
```

APPENDIX B

B.1 Data, Station 12

The raw data and trends are displayed graphically in the Figures to follow. The data after the trends are removed are not given, but can be visualized if one considers the trend line as the time-axis. (Compare Figures B.1.1 and B.1.2).

Table B.1.1 gives the parameters considered at this station and the length of the time series.

TABLE B.1.1: Parameters studied at station 12

Parameters	Code No.	Time Interval
Cd T	101	7210-7511
Cd T	101	7210-7511
Cu T	107	7206-7511
Fe T	110	7206-7511
Ni T	116	7210-7511
Zn T	119	7206-7511
pH	123	7206-7511
SO_4	127	7206-7511
T PART	128	7206-7511
P T	240	7210-7511

Figure B.1.1: Cd T data and trend, station 12 (7210-7511)

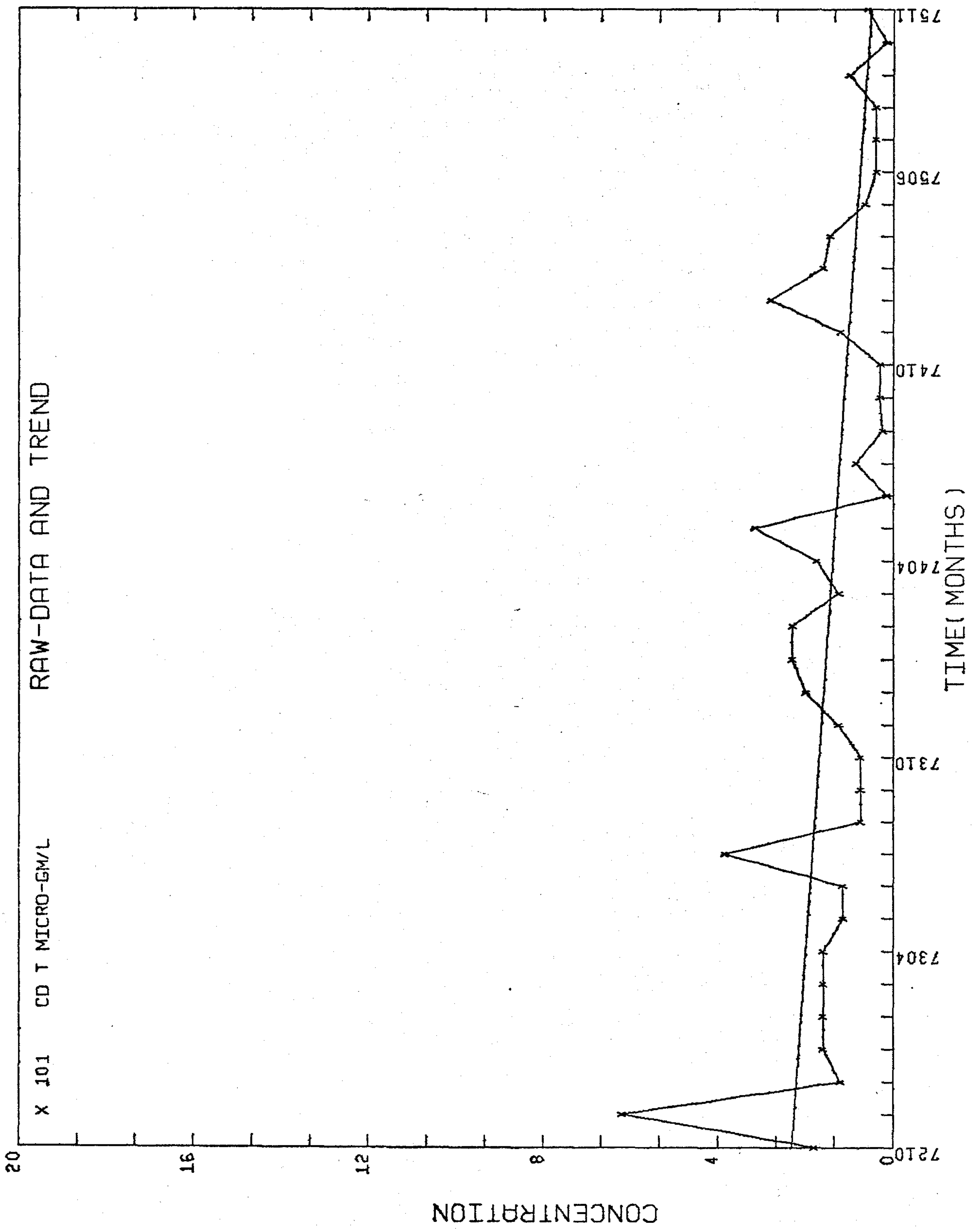


Figure B.1.2: Cd T data-trend removed, station 12 (7210-7511)

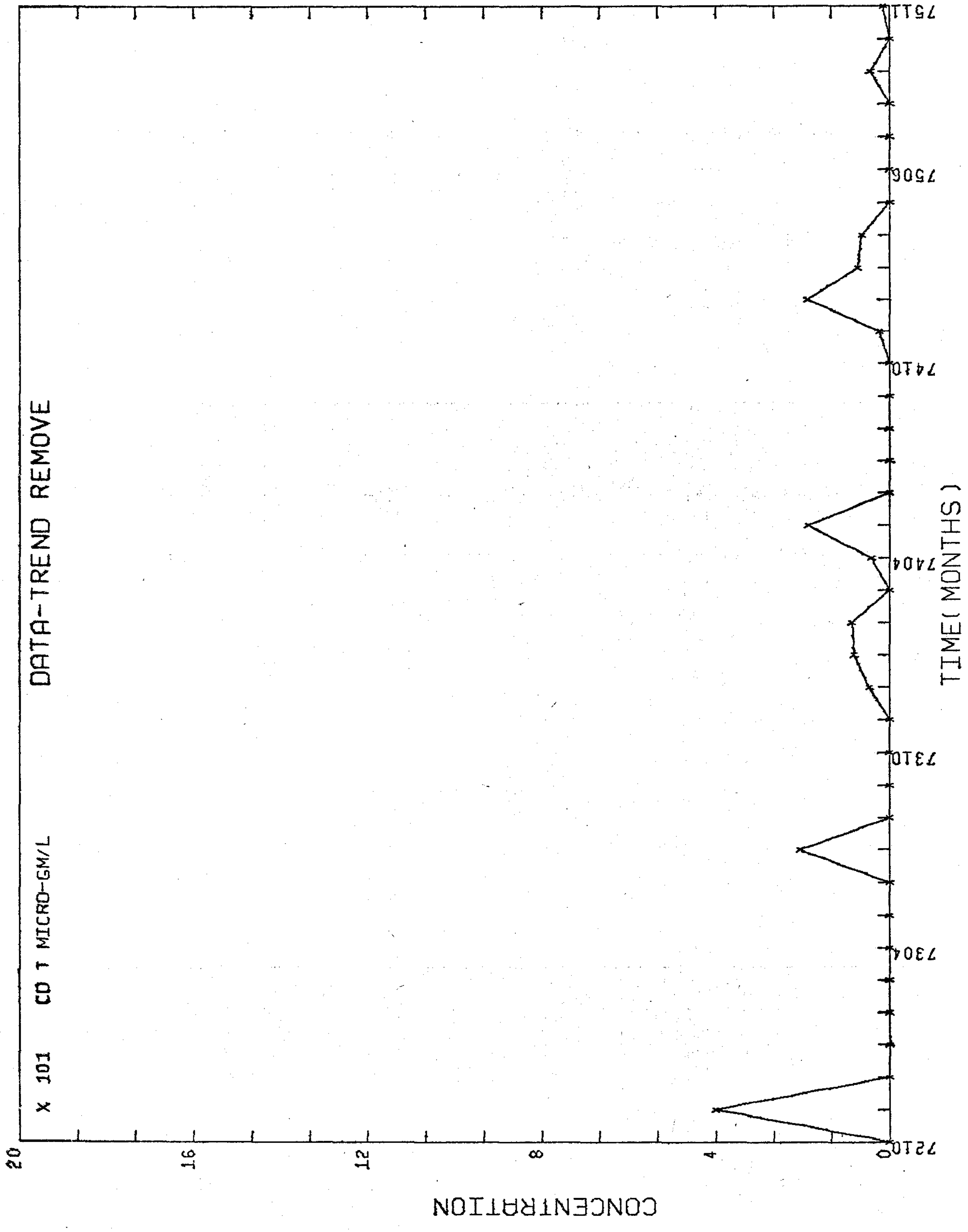


Figure B.1.3: Cu T data and trend, station 12 (7210-7511)

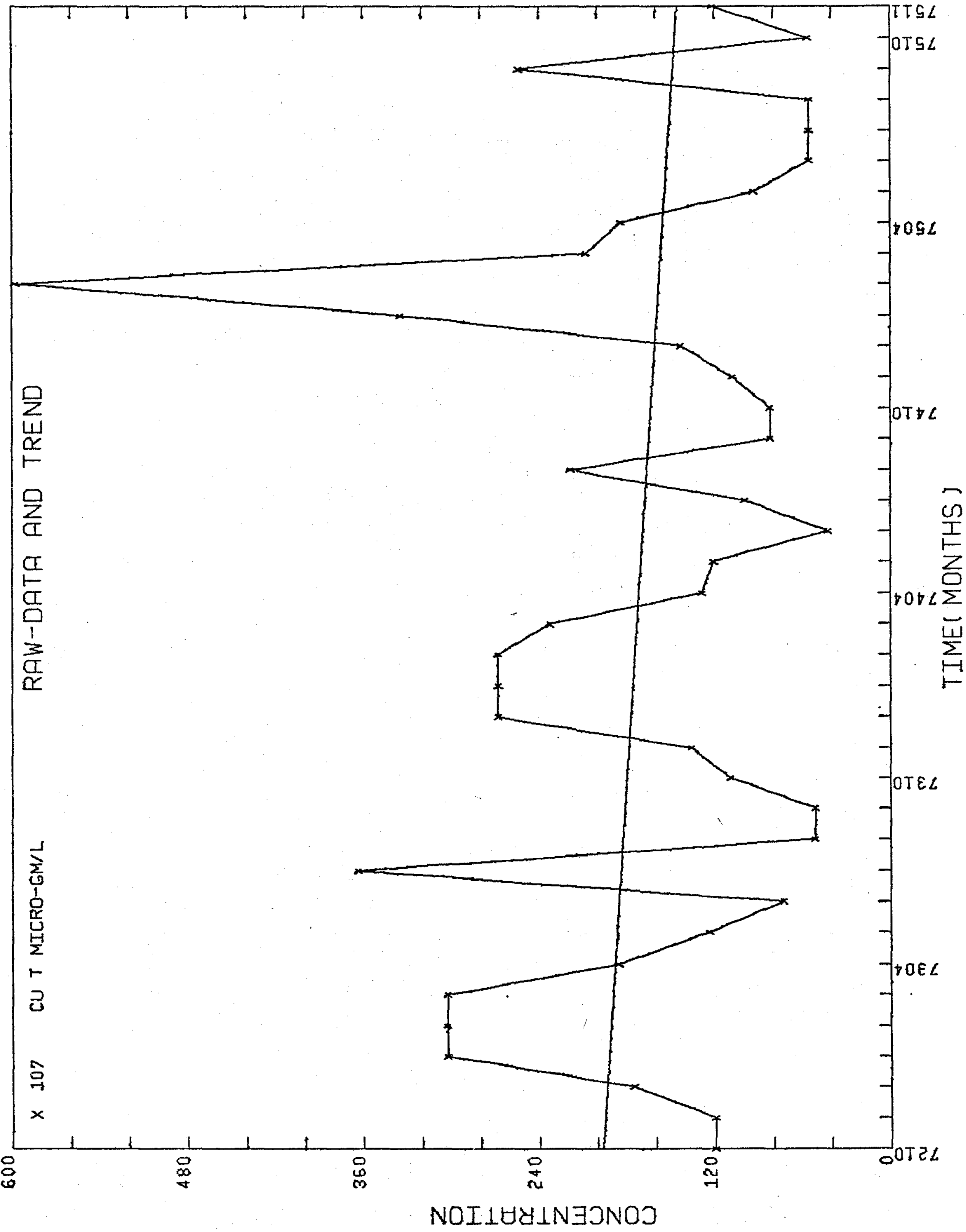


Figure B.1.4: Fe T data and trend, station 12 (7206-7511)

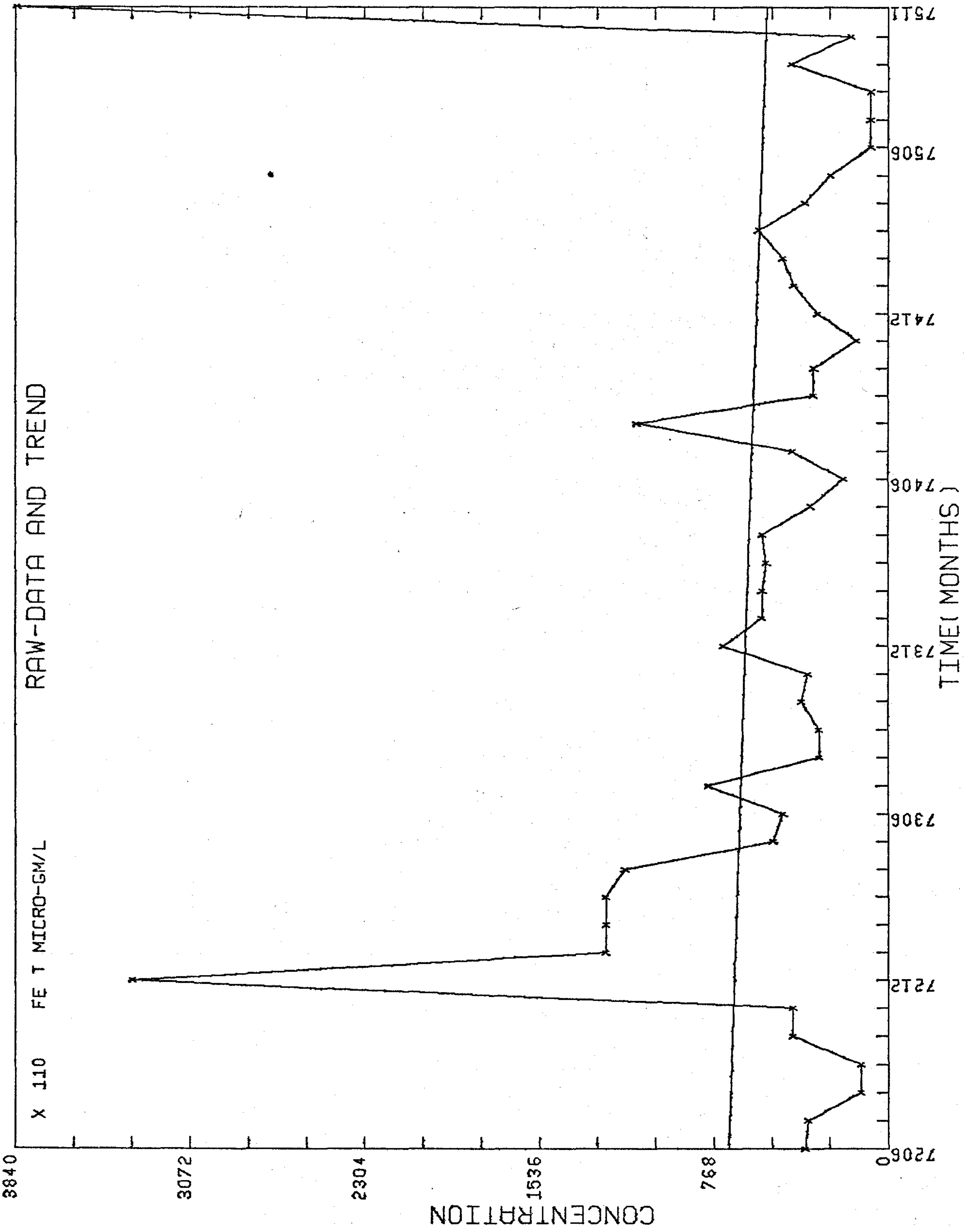


Figure B.1.5: Ni T data and trend, station 12 (7210-7511)

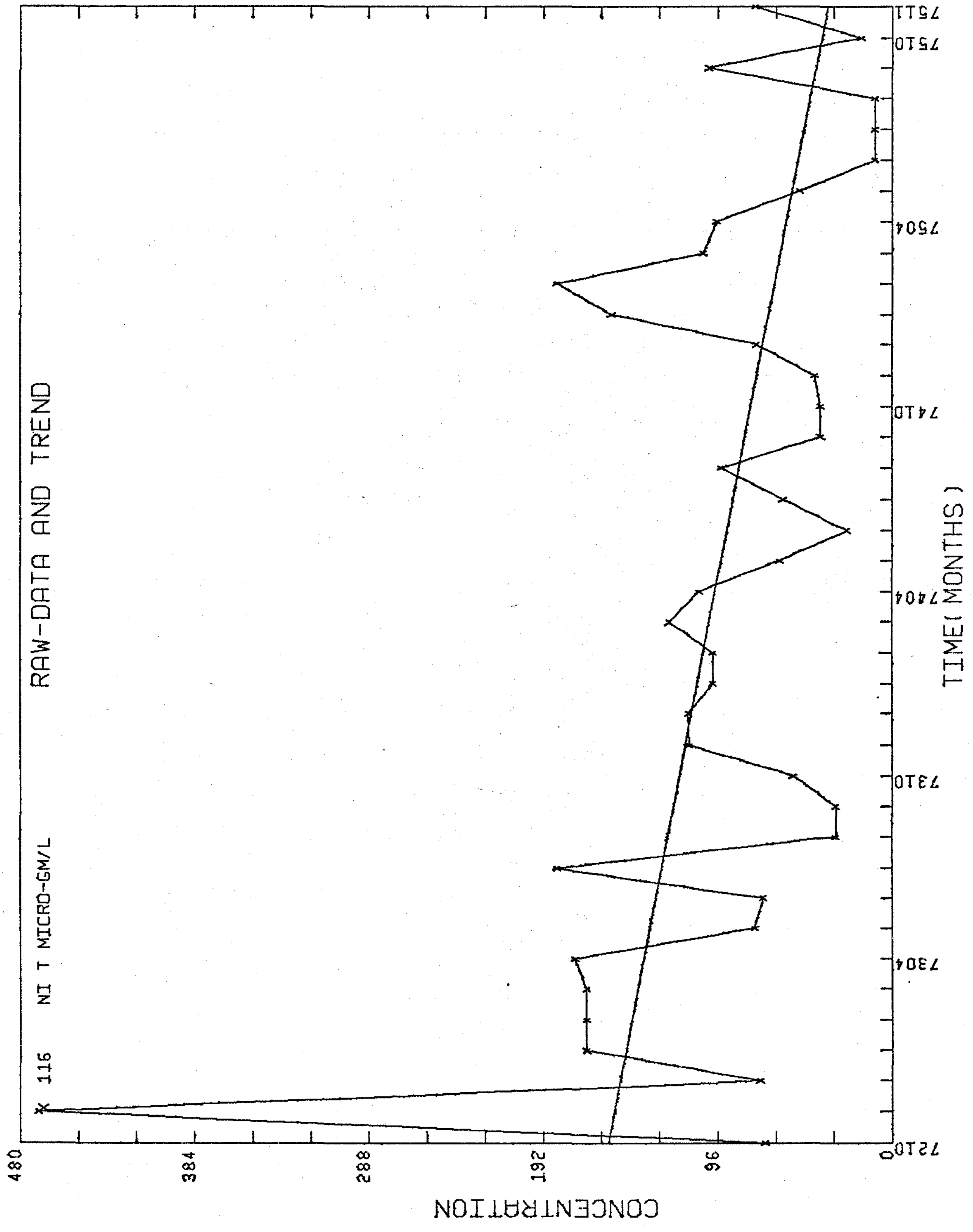


Figure B.1.6: Zn T data and trend, station 12 (7206-7511)

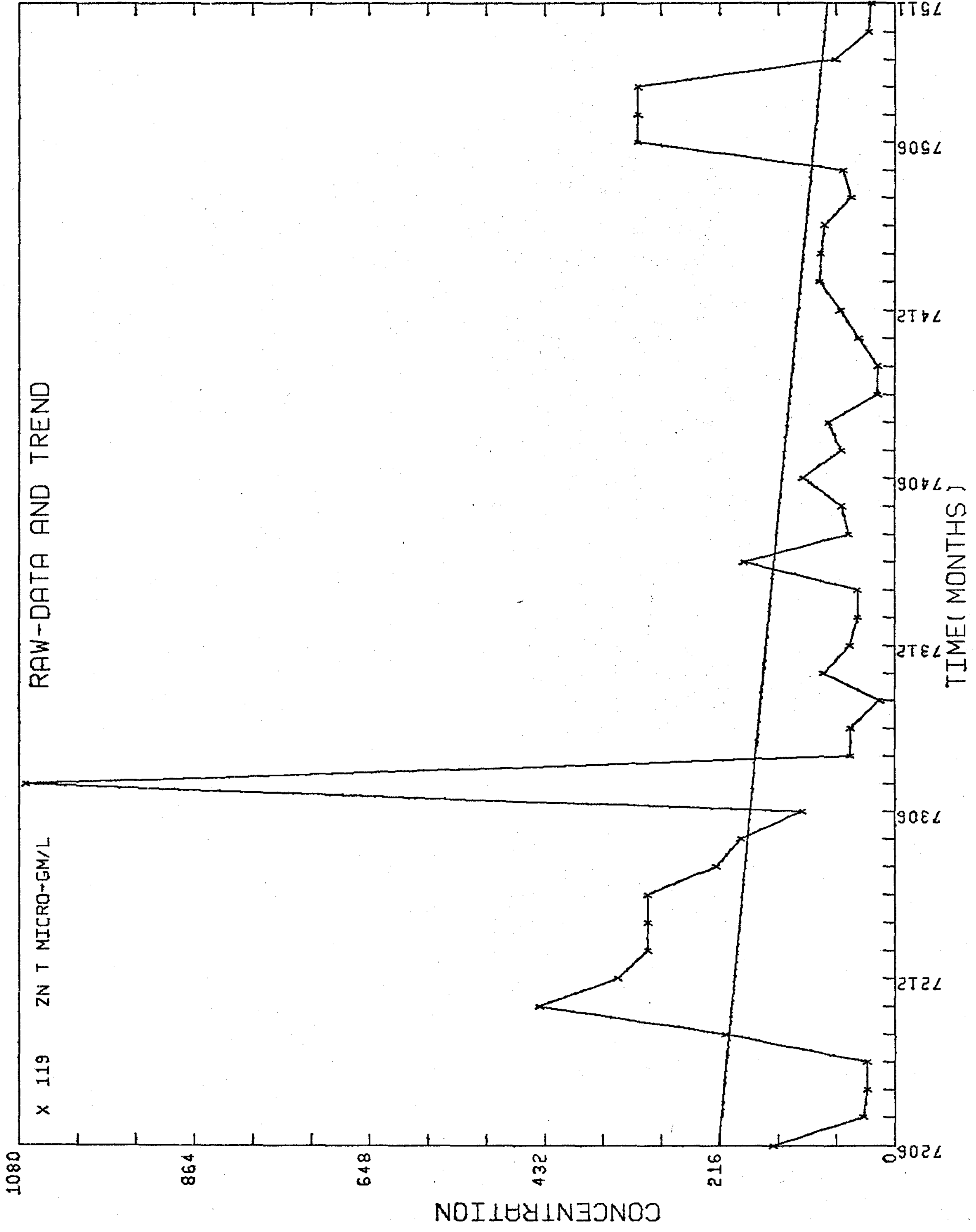


Figure B.1.7: pH data and trend, station 12 (7206-7511)

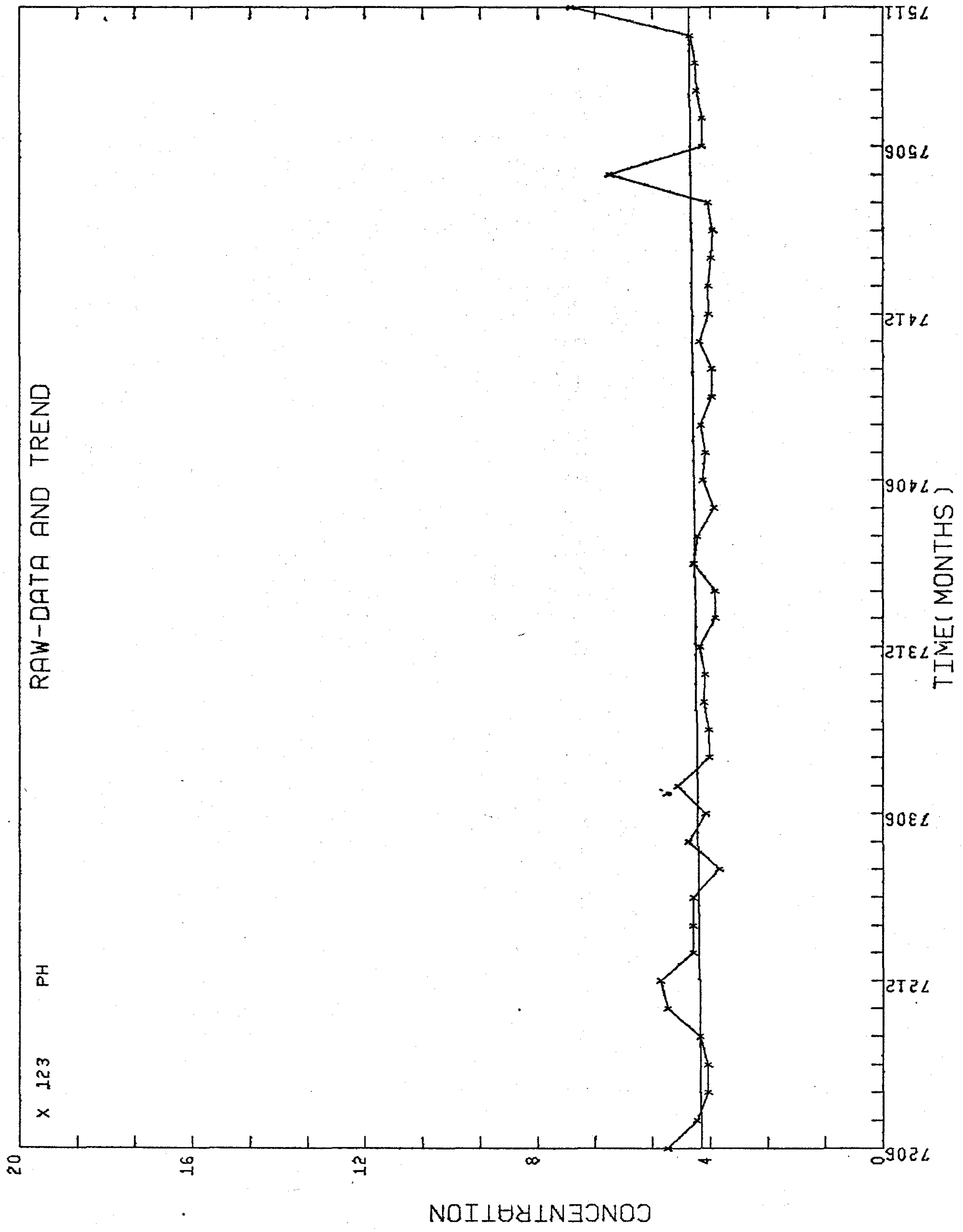


Figure B.1.8: SO_4 data and trend, station 12 (7206-7511)

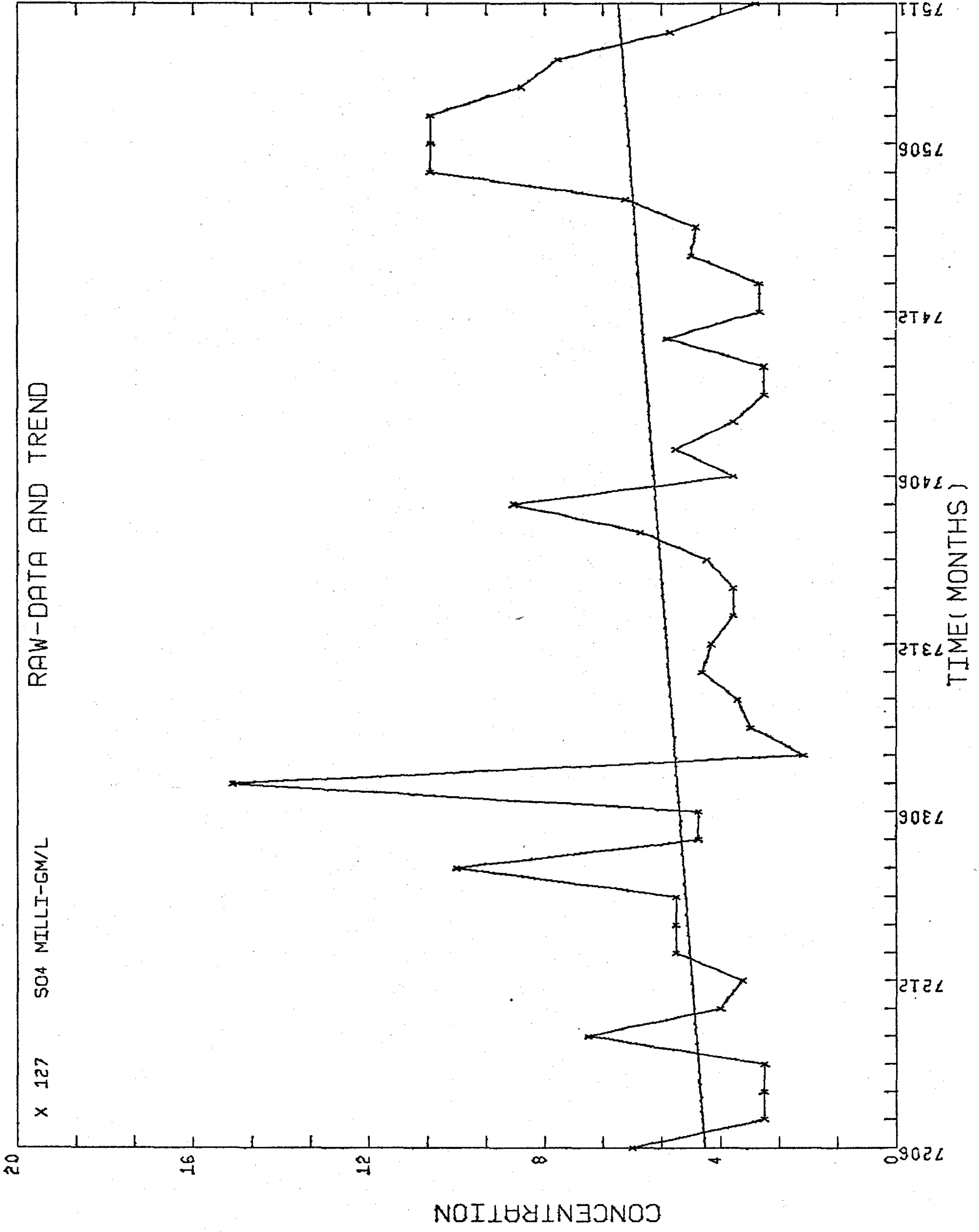


Figure B.1.9: T PART data and trend, station 12 (7206-7511)

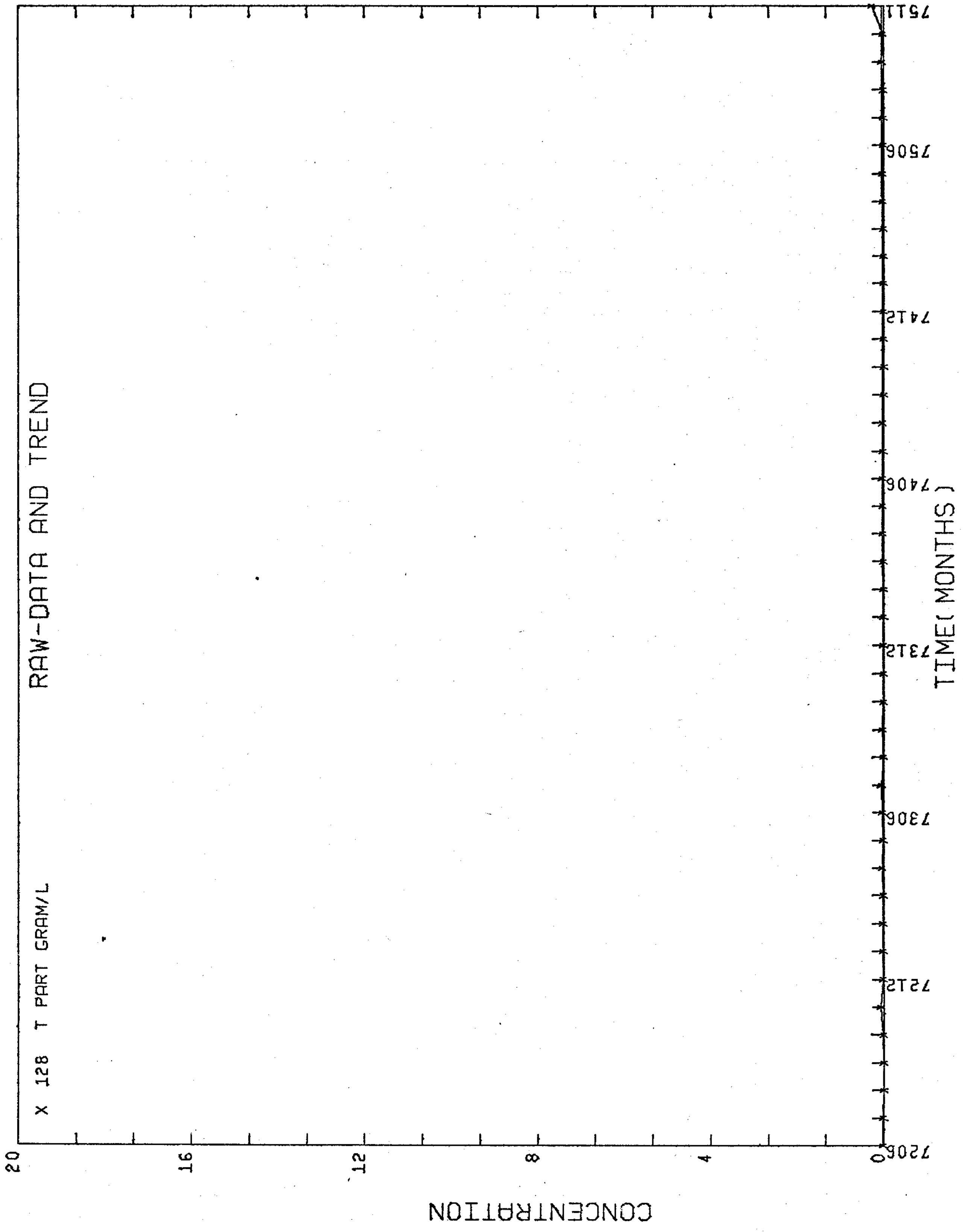
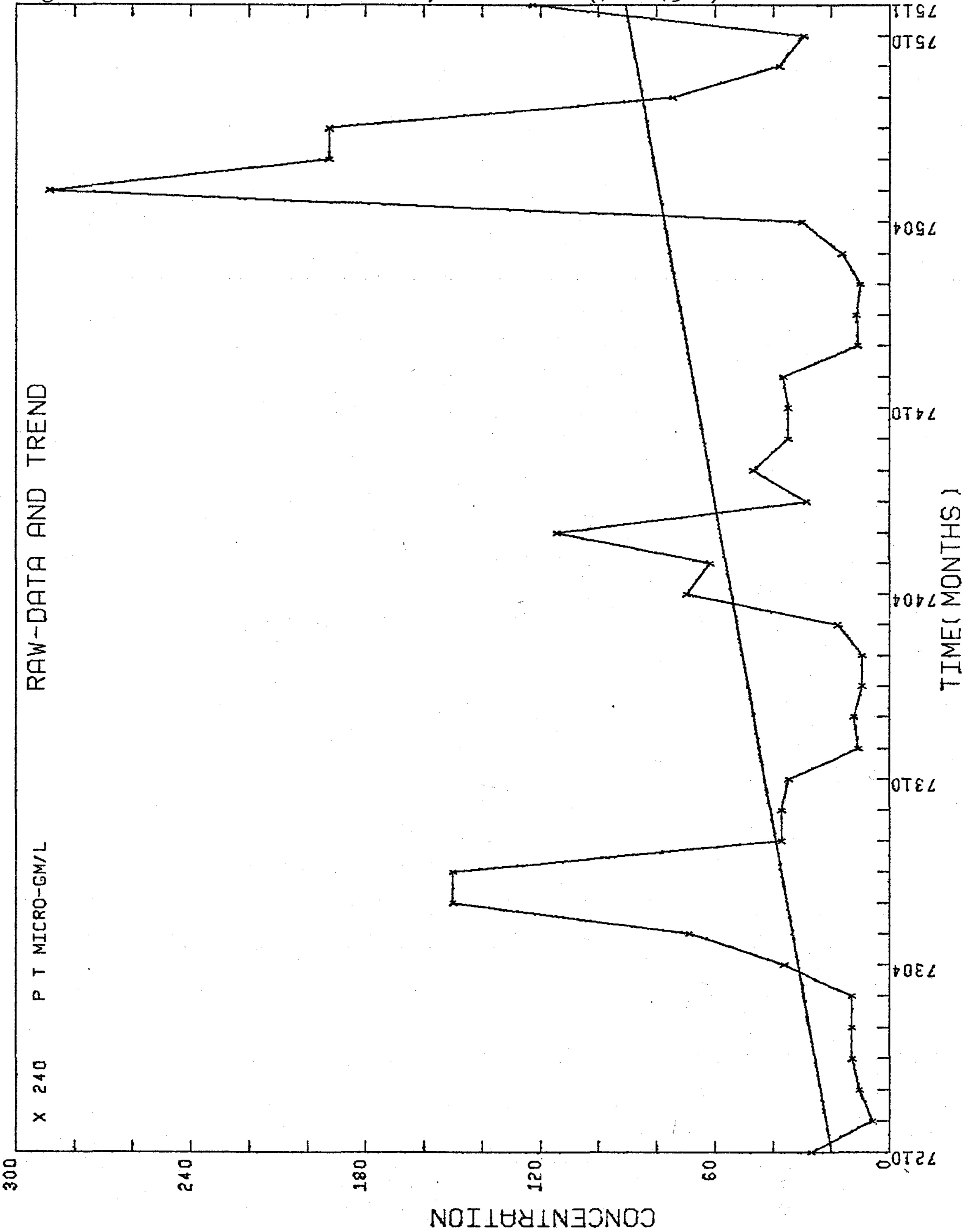


Figure B.1.10: P T data and trend, station 12 (7210-7511)



B.2 Data, Station 2

Table B.2.1 gives the parameters considered and the length of the time series. The data are displayed graphically in the figures to follow.

TABLE B.2.1: Parameters studied at station 2

Parameters	Code No.	Time Interval
Cd T	101	7206-7510
Cu T	107	7206-7510
Fe T	110	7206-7510
Ni T	116	7206-7510
Zn T	119	7206-7510
pH	123	7206-7510
SO_4	127	7206-7510
T PART	128	7206-7510
P T	240	7211-7510

Figure B.2.1: Cd T data and trend, station 2 (7206-7510)

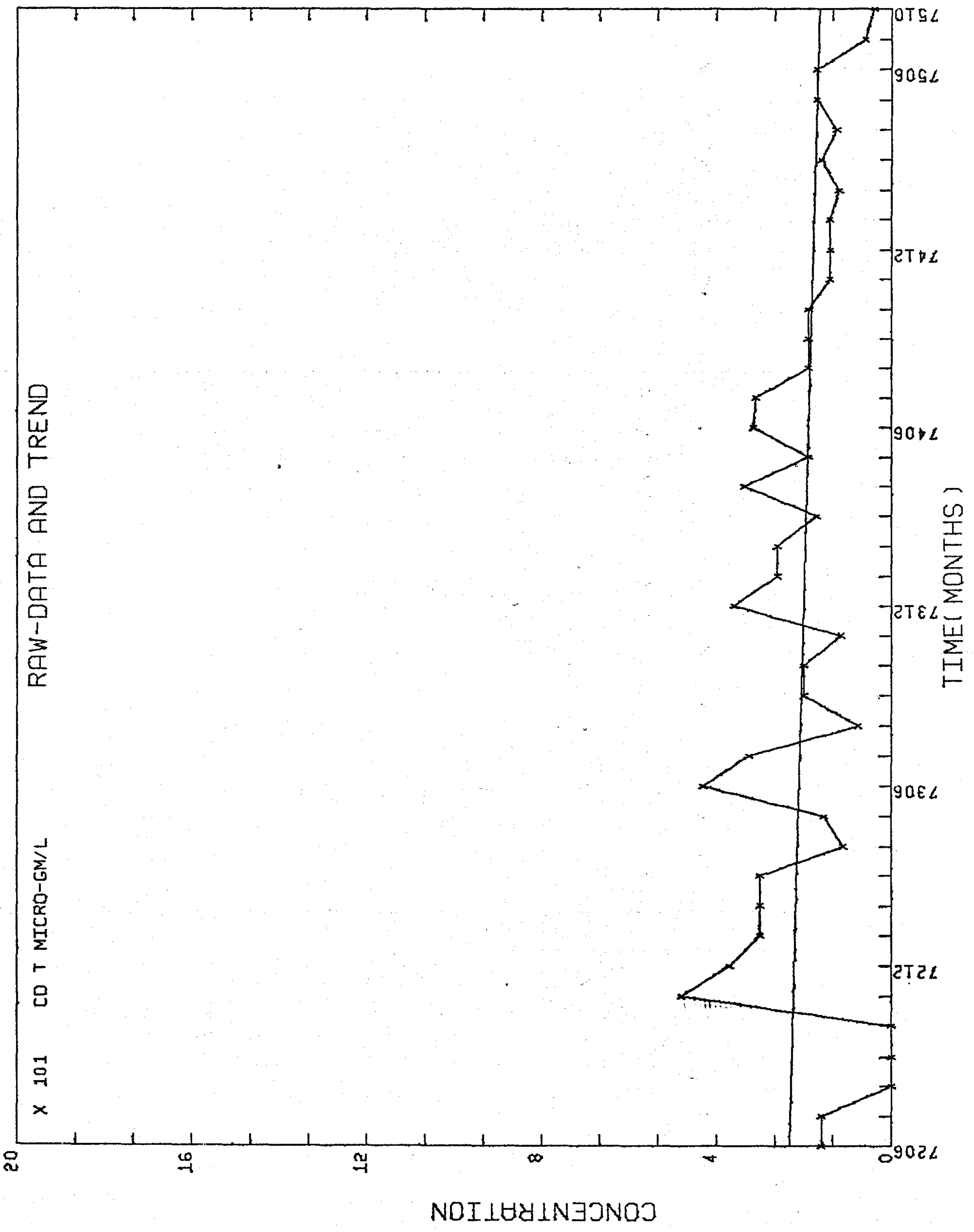


Figure B.2.2: Cu T data and trend, station 2 (7206-7510)

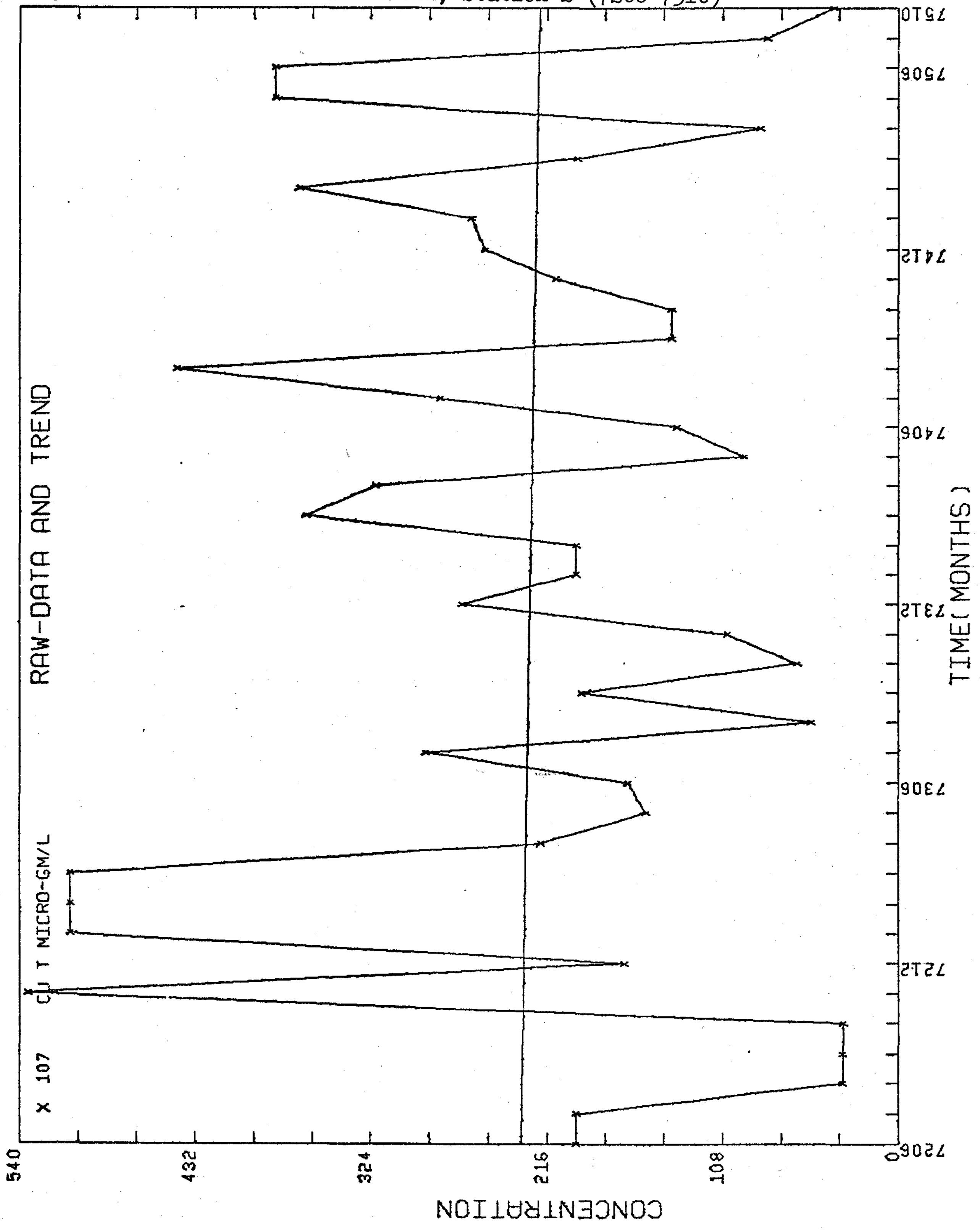


Figure B.2.3: Fe T data and trend, station 2 (7206-7510)

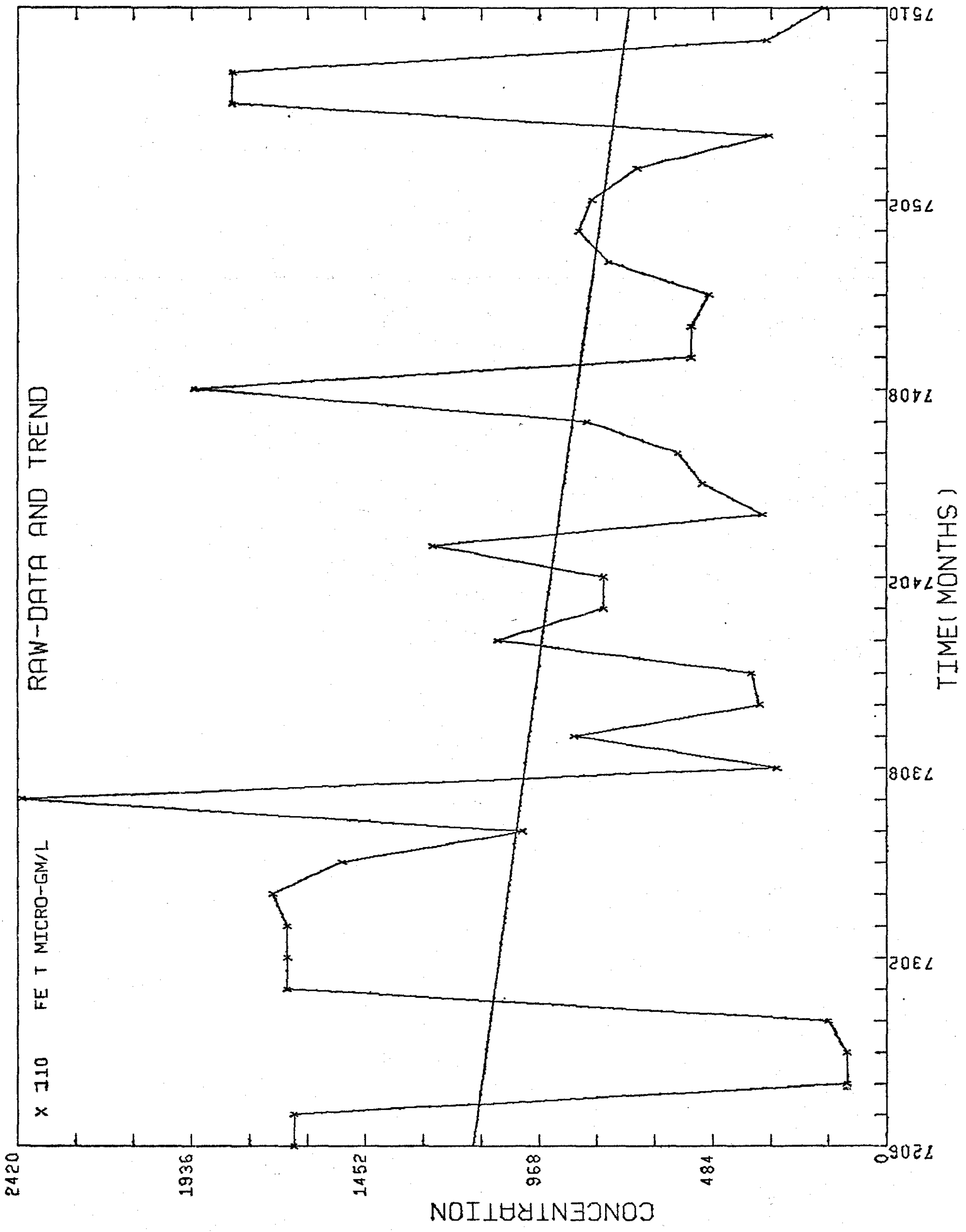


Figure B.2.4: Ni T data and trend, station 2 (7206-7510)

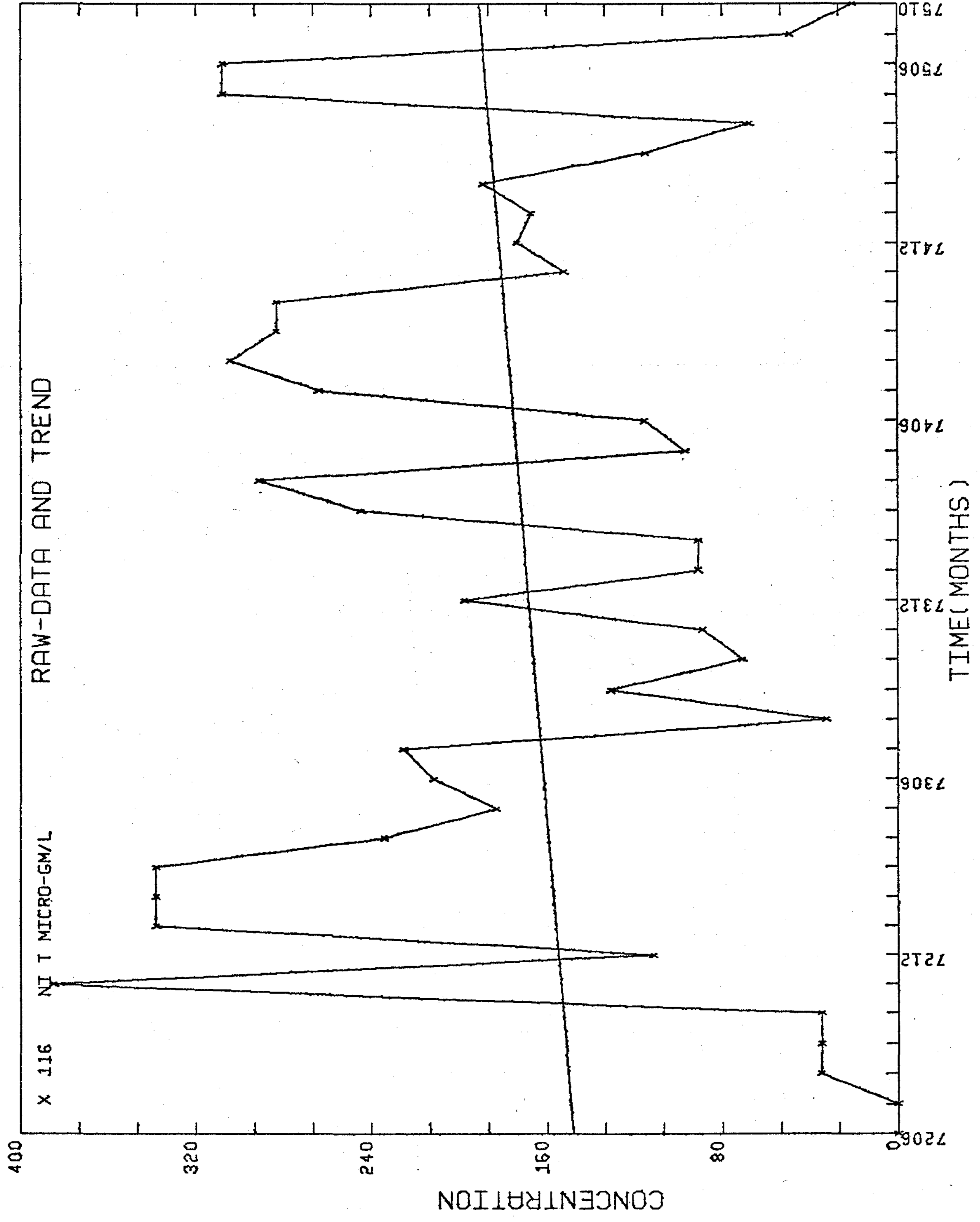


Figure B.2.5: Zn T data and trend, station 2 (7206-7510)

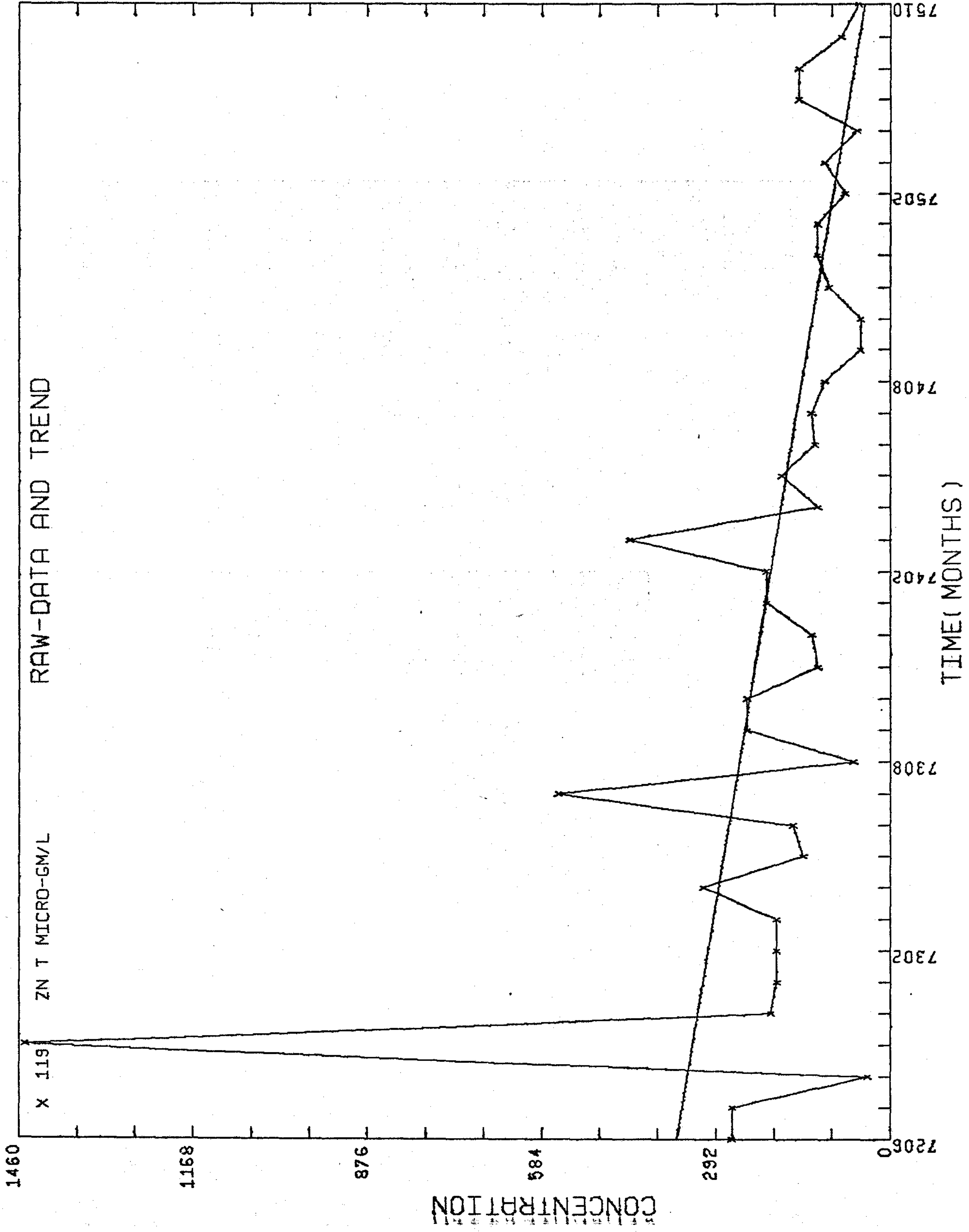


Figure B.2.6: pH data and trend, station 2 (7206-7510)

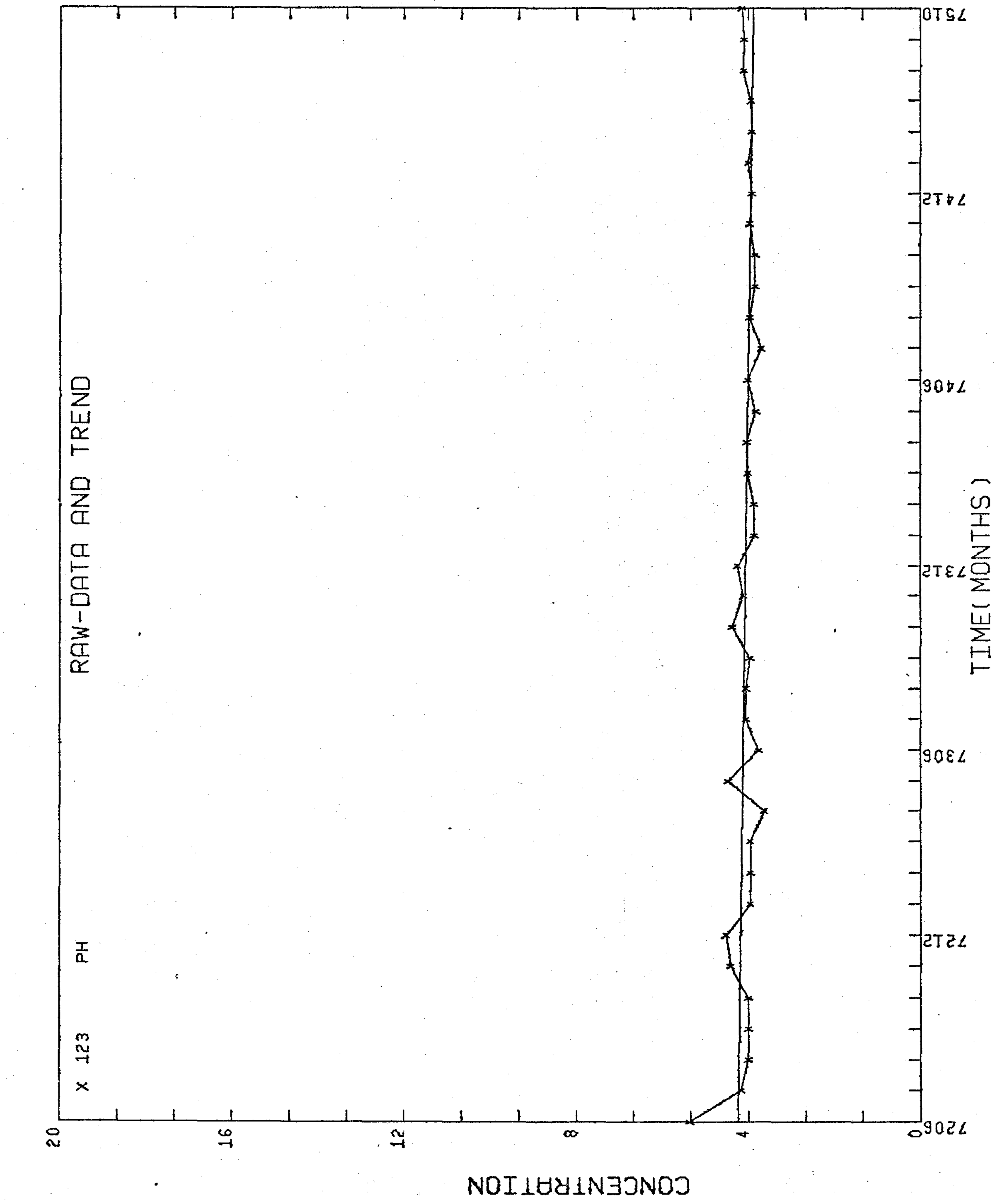


Figure B.2.7: SO_4 data and trend, station 2 (7206-7510)

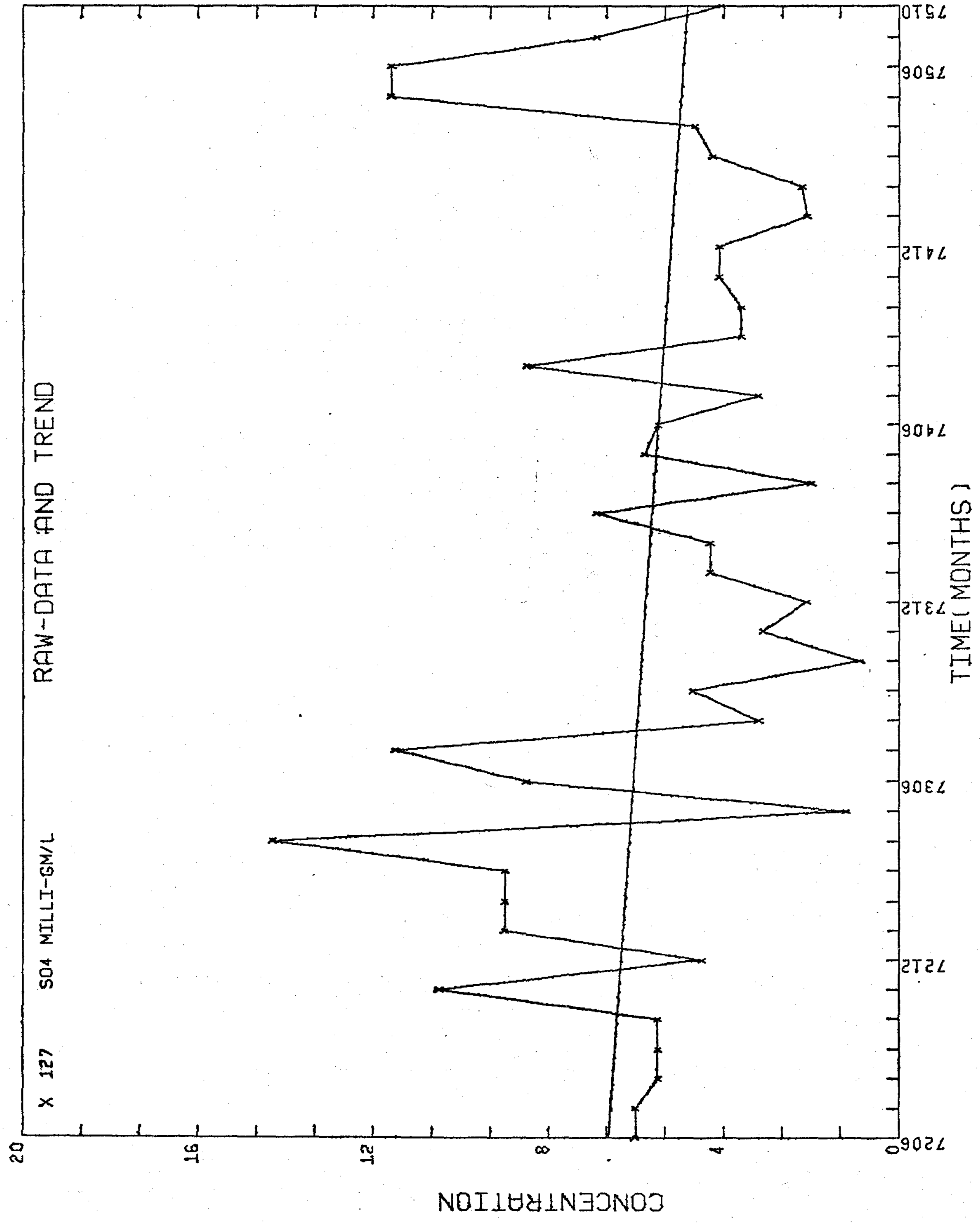


Figure B.2.8: T PART data and trend, station 2 (7206-7510)

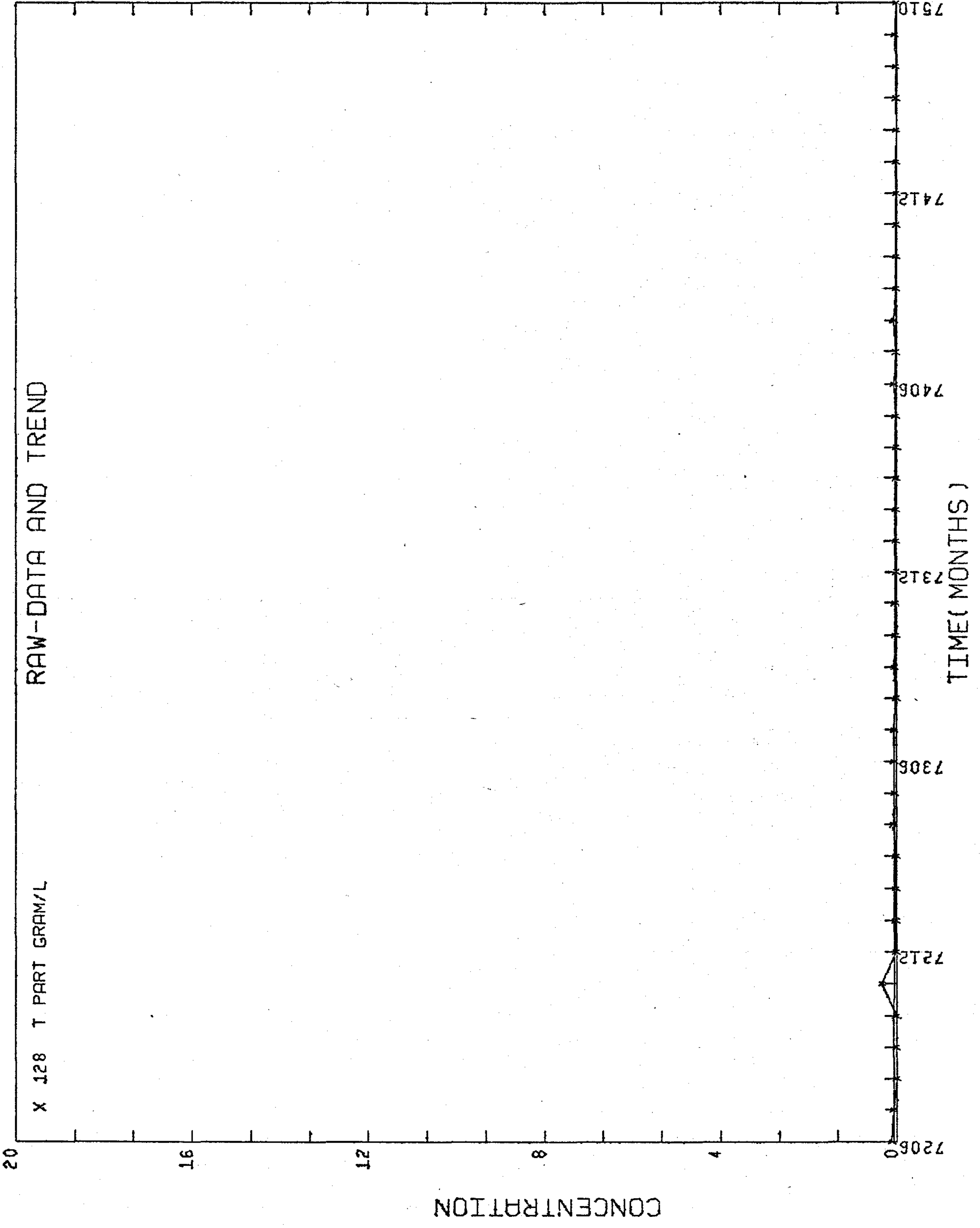
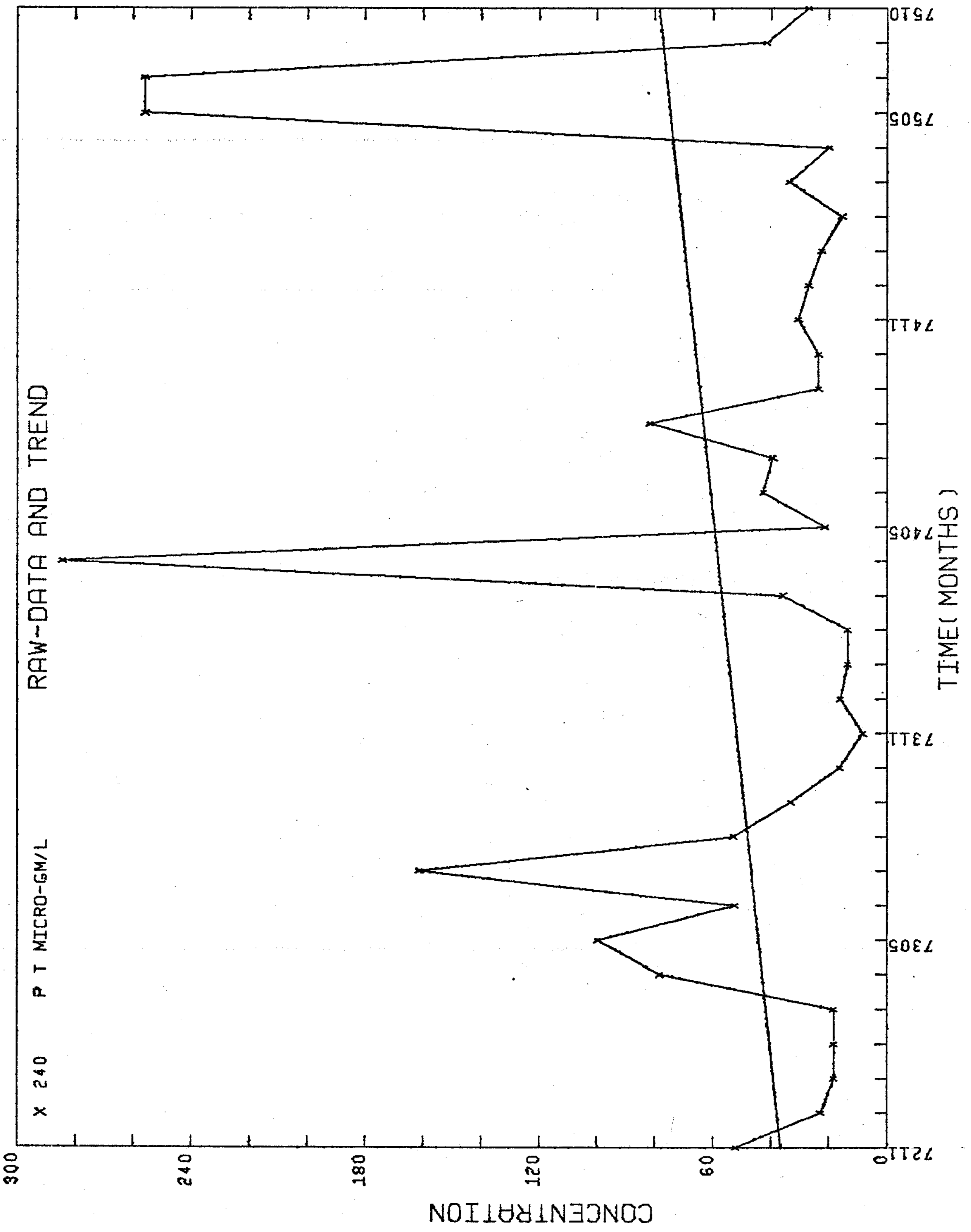


Figure B.2.9: P T data and trend, station 2 (7211-7510)



B.3 Data, Station 3

Table B.3.1 gives the parameters considered and the length of the time series. The data are displayed graphically in the figures to follow.

TABLE B.3.1: Parameters studied at station 3

Parameters	Code No.	Time Interval
Cd T	101	7211-7511
Cu T	107	7211-7511
Fe T	110	7206-7511
Ni T	116	7211-7511
Zn T	119	7206-7511
pH	128	7206-7511
SO_4	127	7206-7511

Figure B.3.1: Cd T data and trend, station 3 (7211-7511)

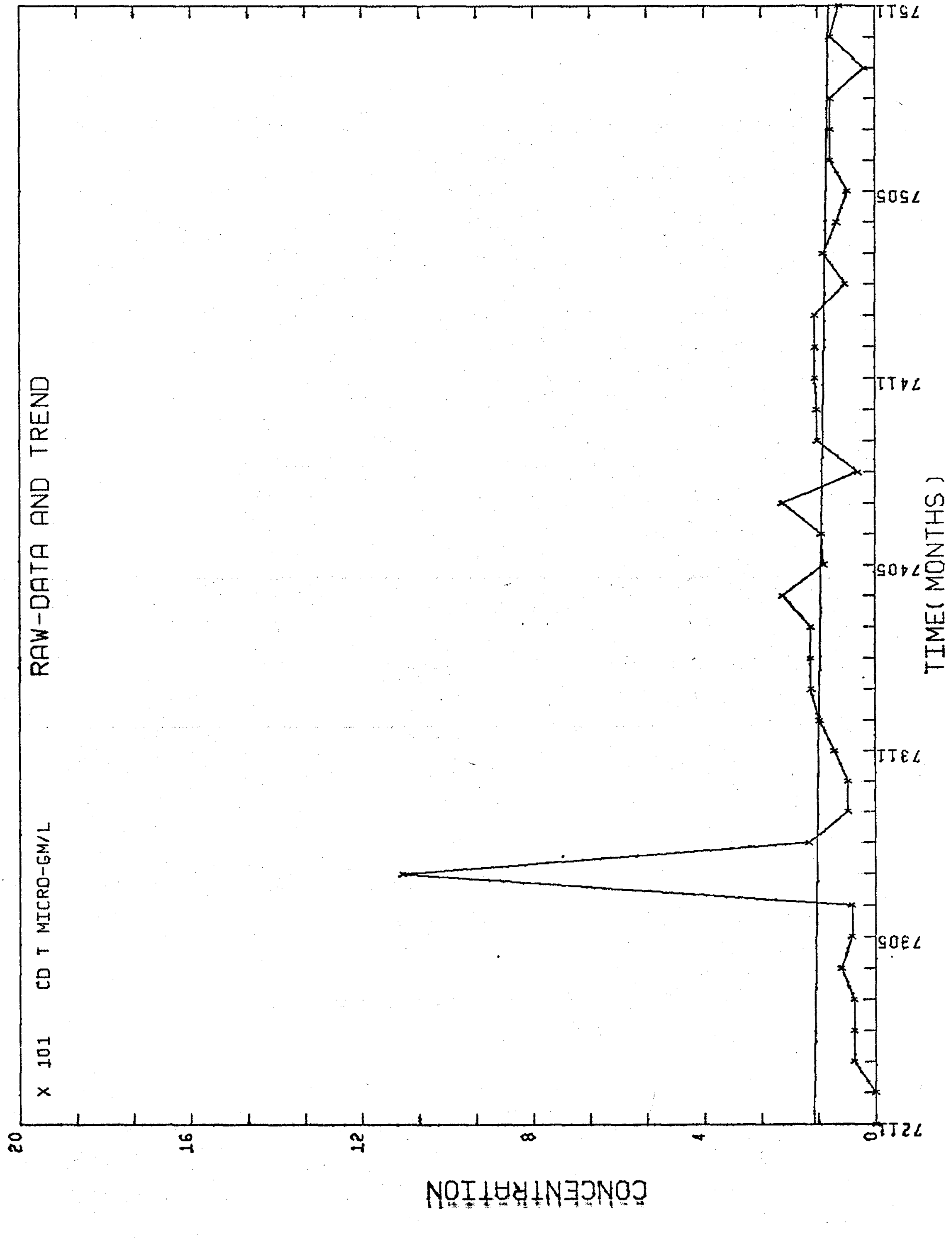


Figure B.3.2: Cu T data and trend, station 3 (7211-7511)

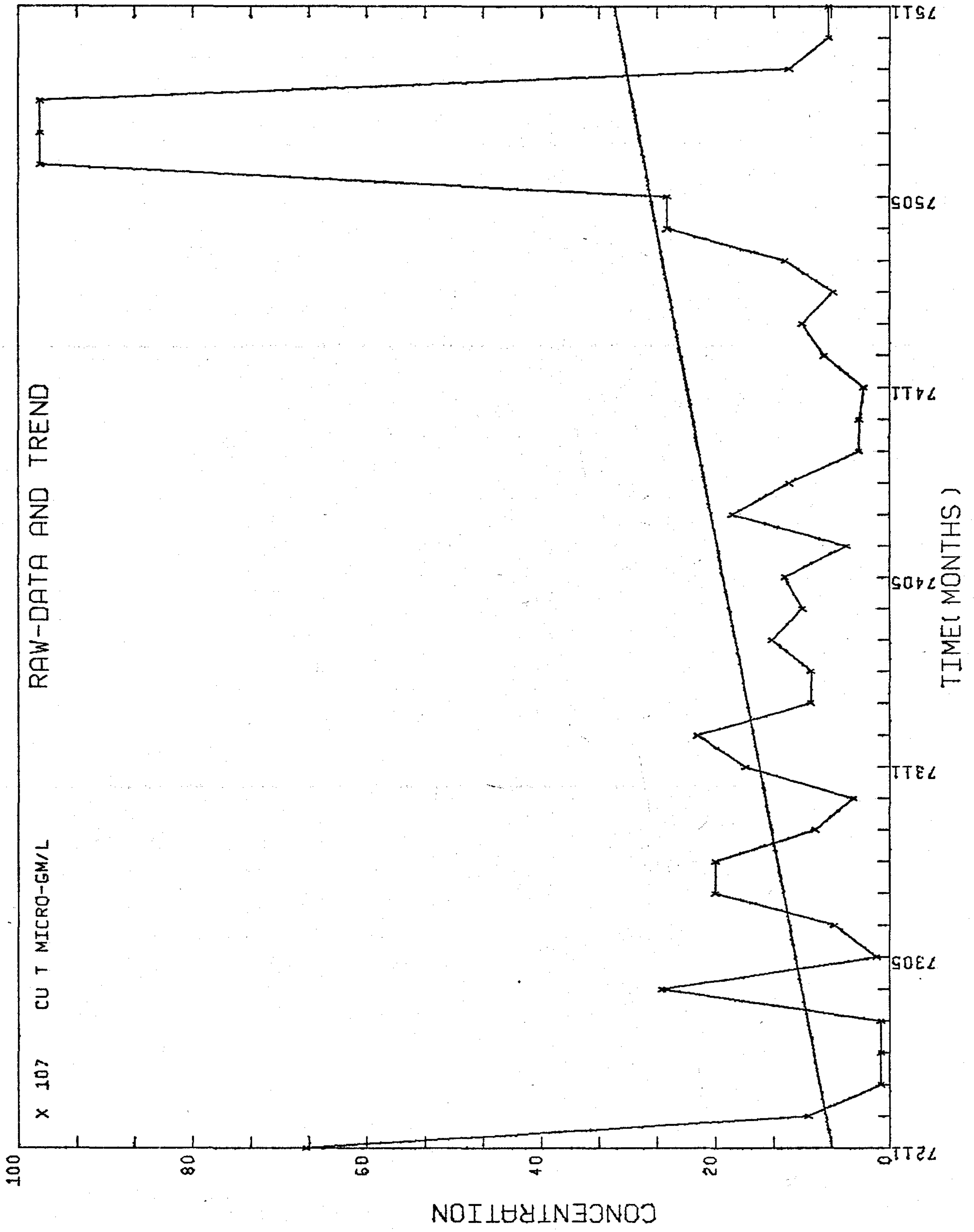


Figure B.3.3: Fe T data and trend, station 3 (7206-7511)

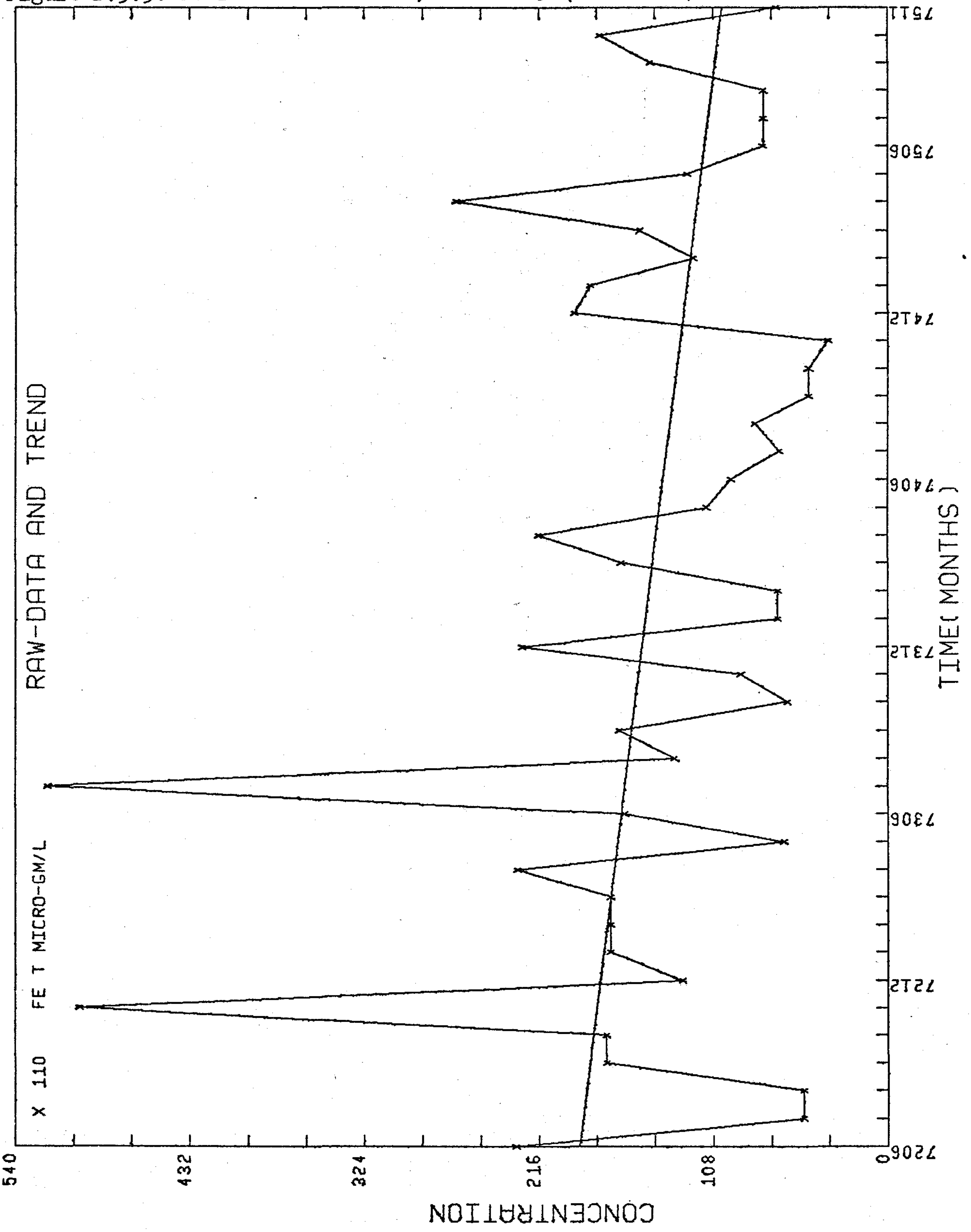


Figure B.3.4: Ni T data and trend, station 3 (7211-7511)

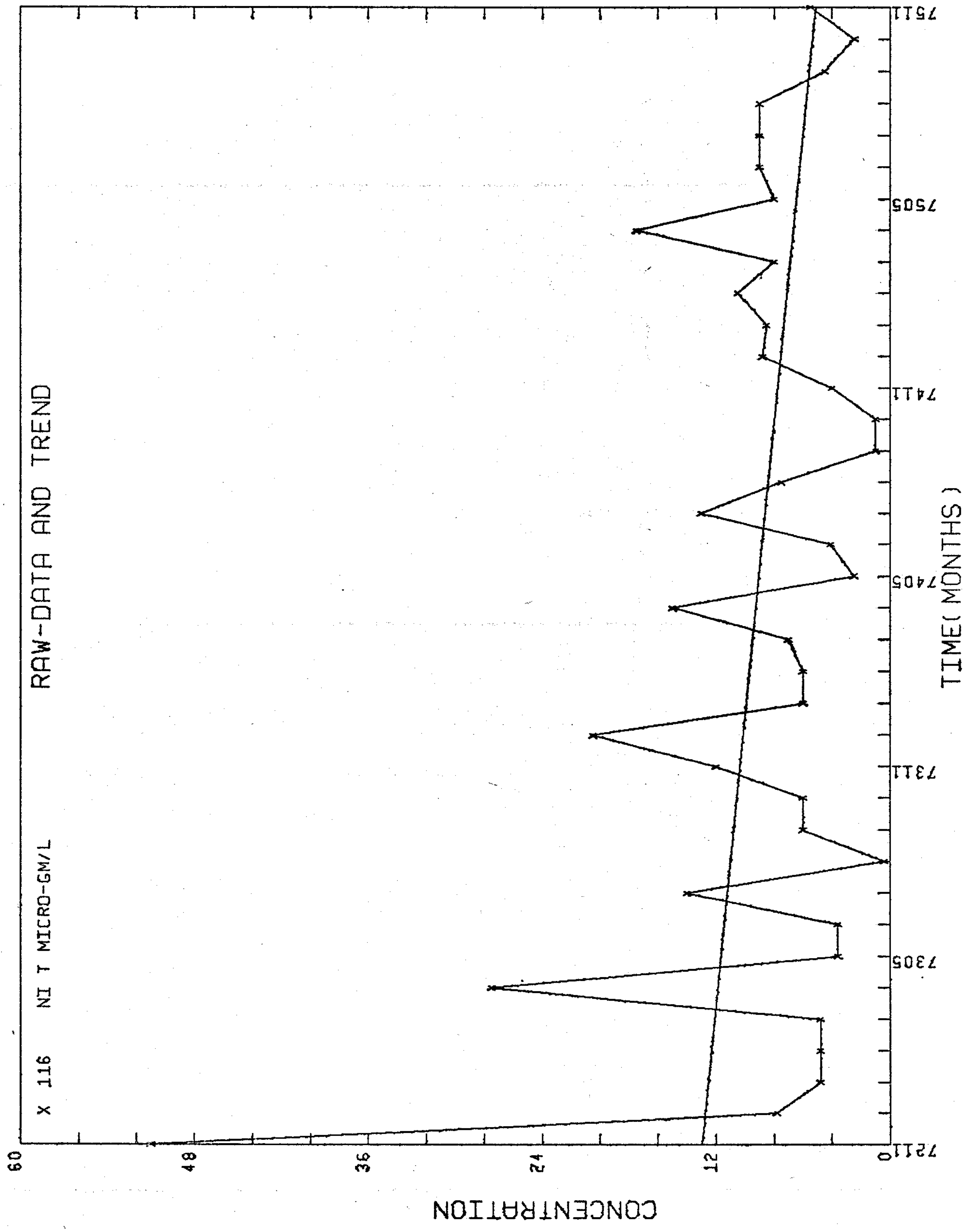


Figure B.3.5: Zn T data and trend, station 3 (7206-7511)

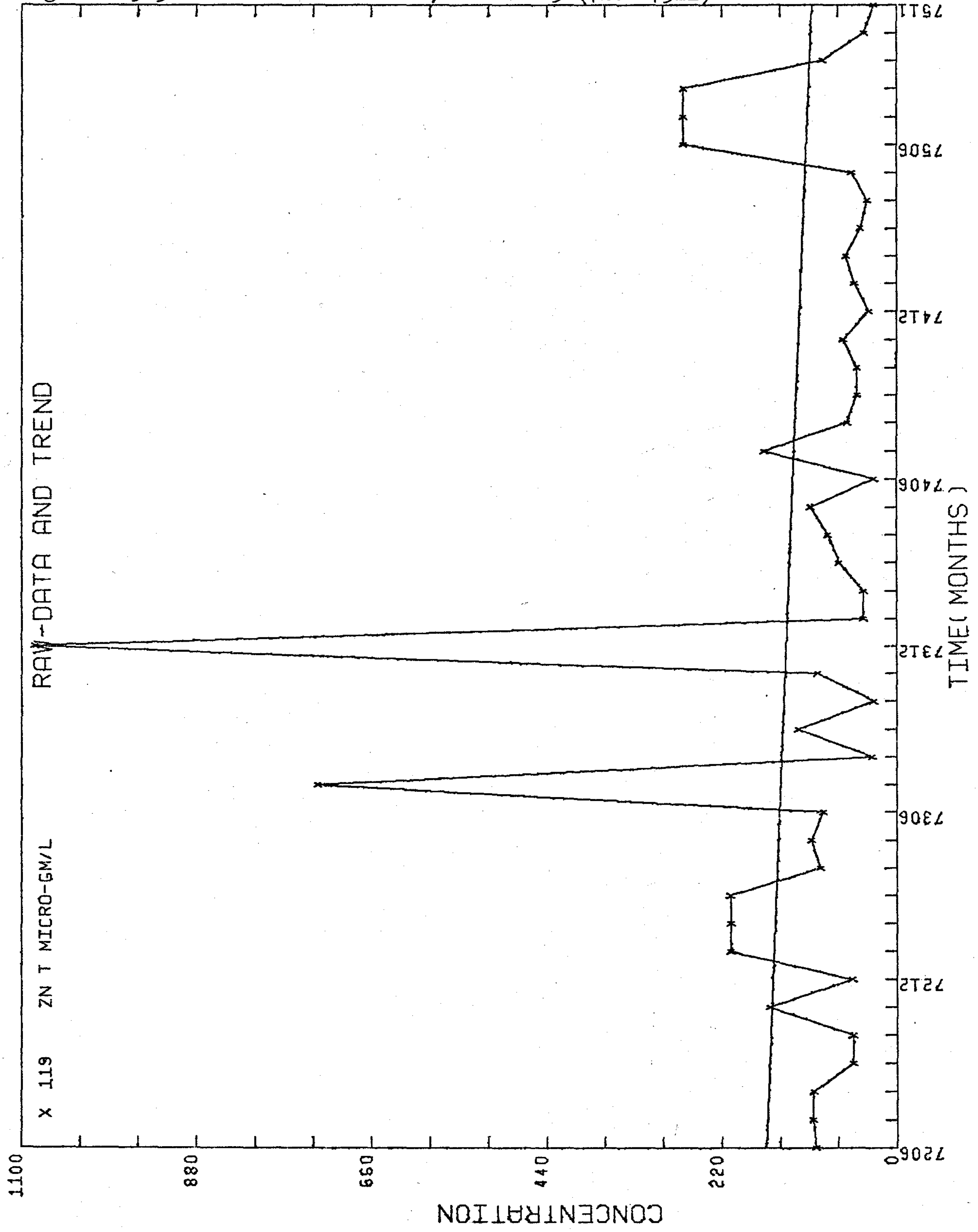


Figure B.3.6: pH data and trend, station 3 (7206-7511)

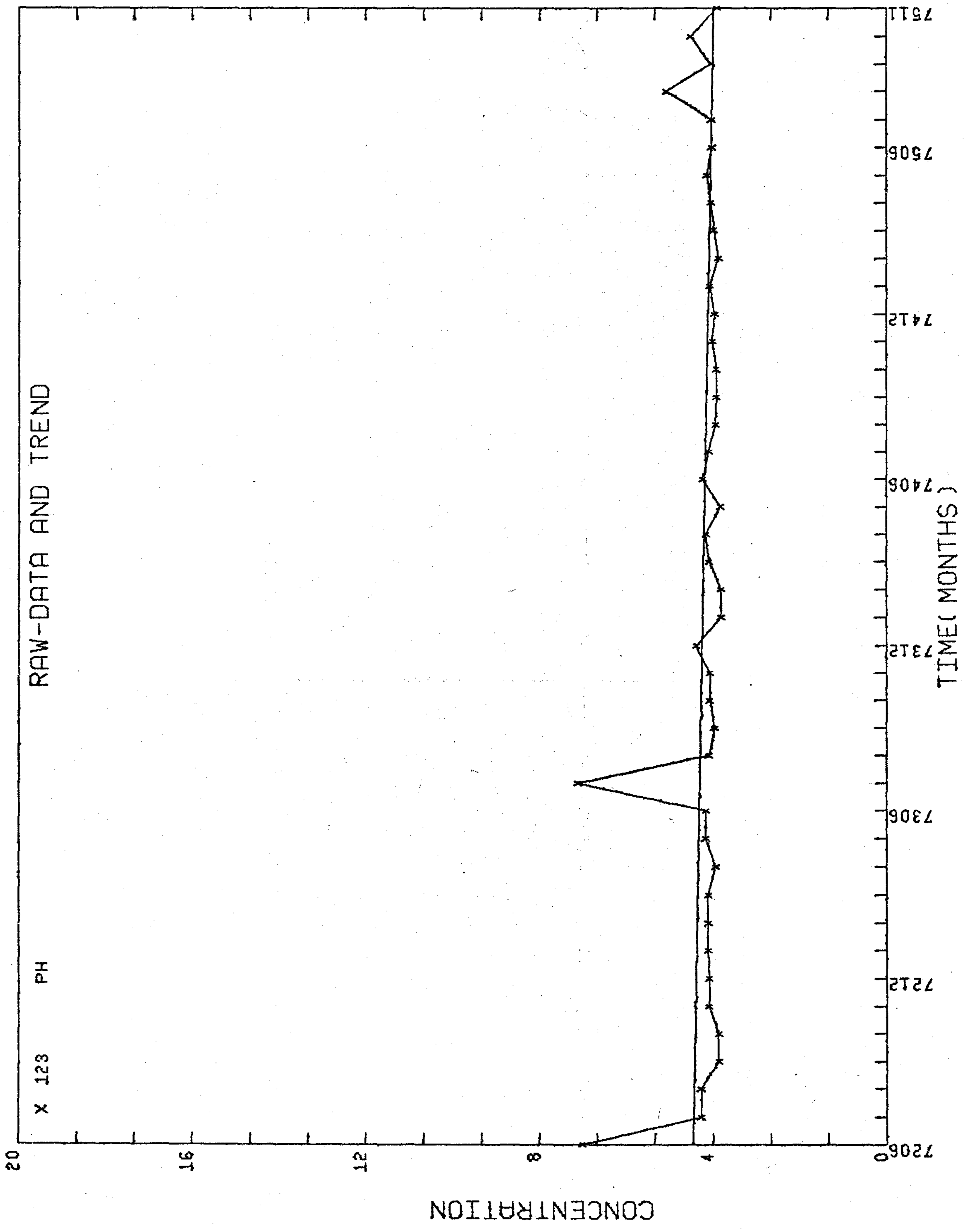
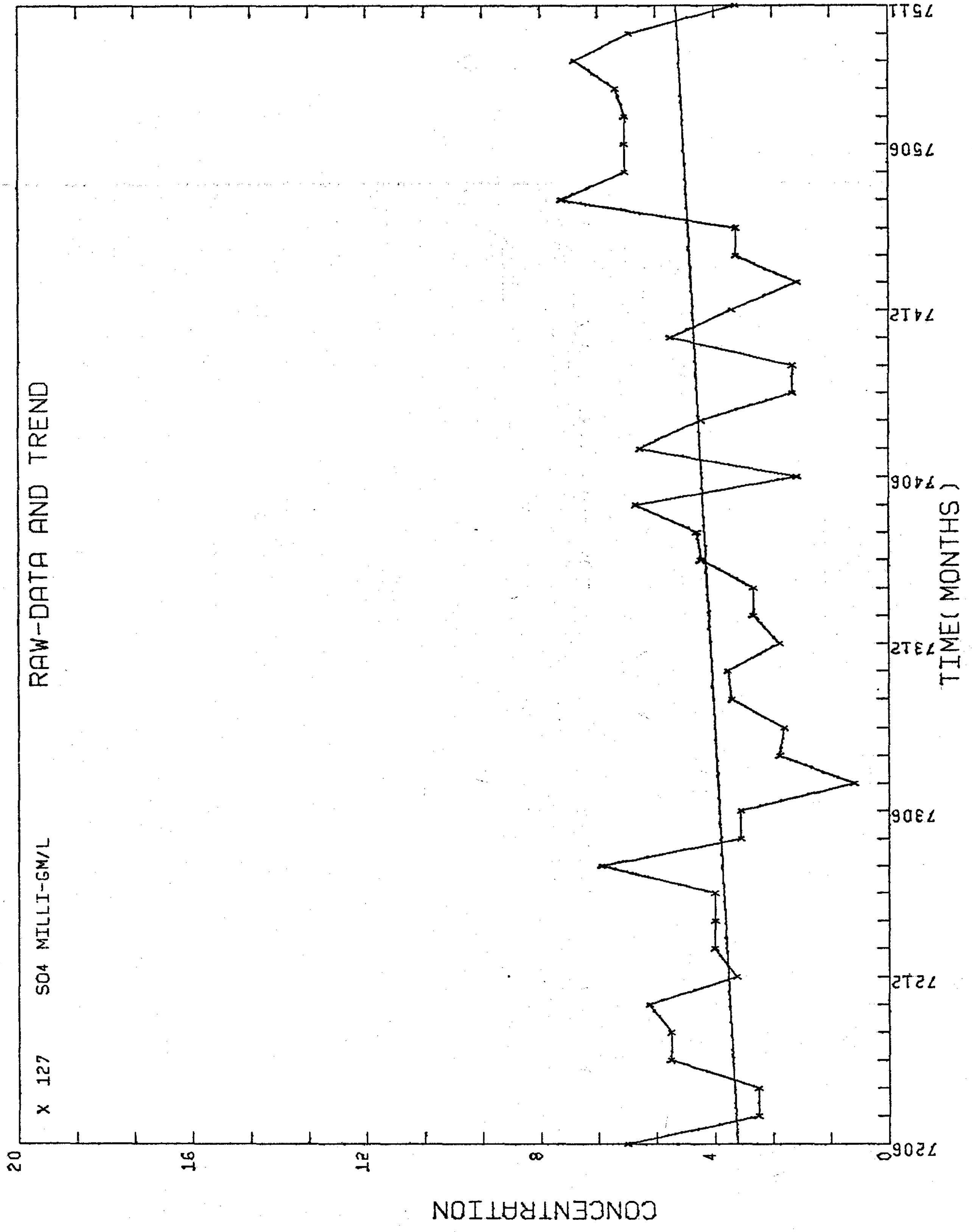


Figure B.3.7: SO_4 data and trend, station 3 (7206-7511)



B.4 Data, Station 4

Table B.4.1 gives the parameters considered and the length of the time series. The data are displayed graphically in the figures to follow.

TABLE B.4.1: Parameters studied at station 4

Parameters	Code No.	Time Interval
Cd T	101	7204-7507
Cu T	107	7204-7509
Fe T	110	7204-7509
Ni T	116	7204-7509
Zn T	119	7204-7509
pH	123	7203-7509
SO_4	127	7203-7509

Figure B.4.1: Cd T data and trend, station 4 (7204-7507)

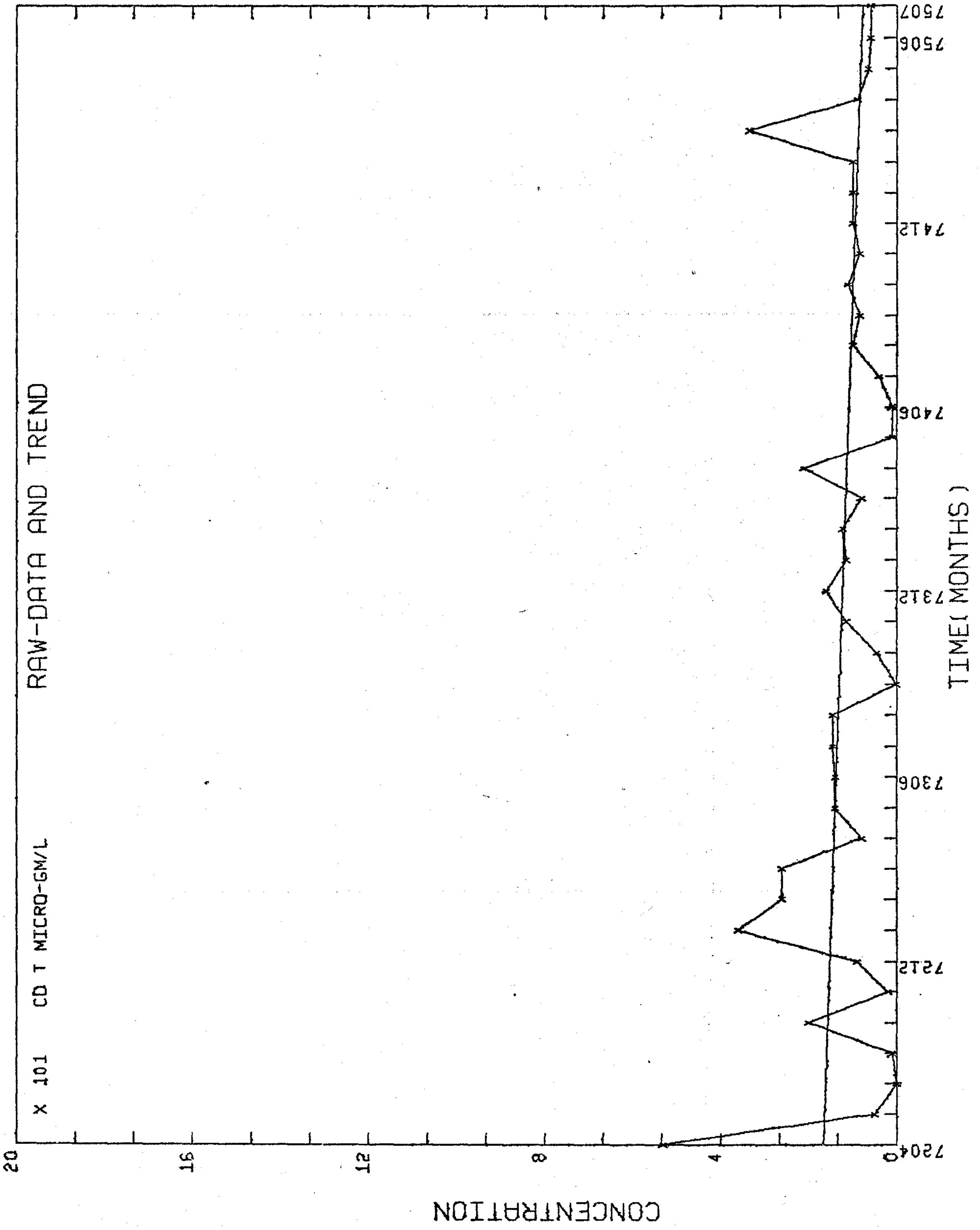


Figure B.4.2: Cu T data and trend, station 4 (7204-7509)

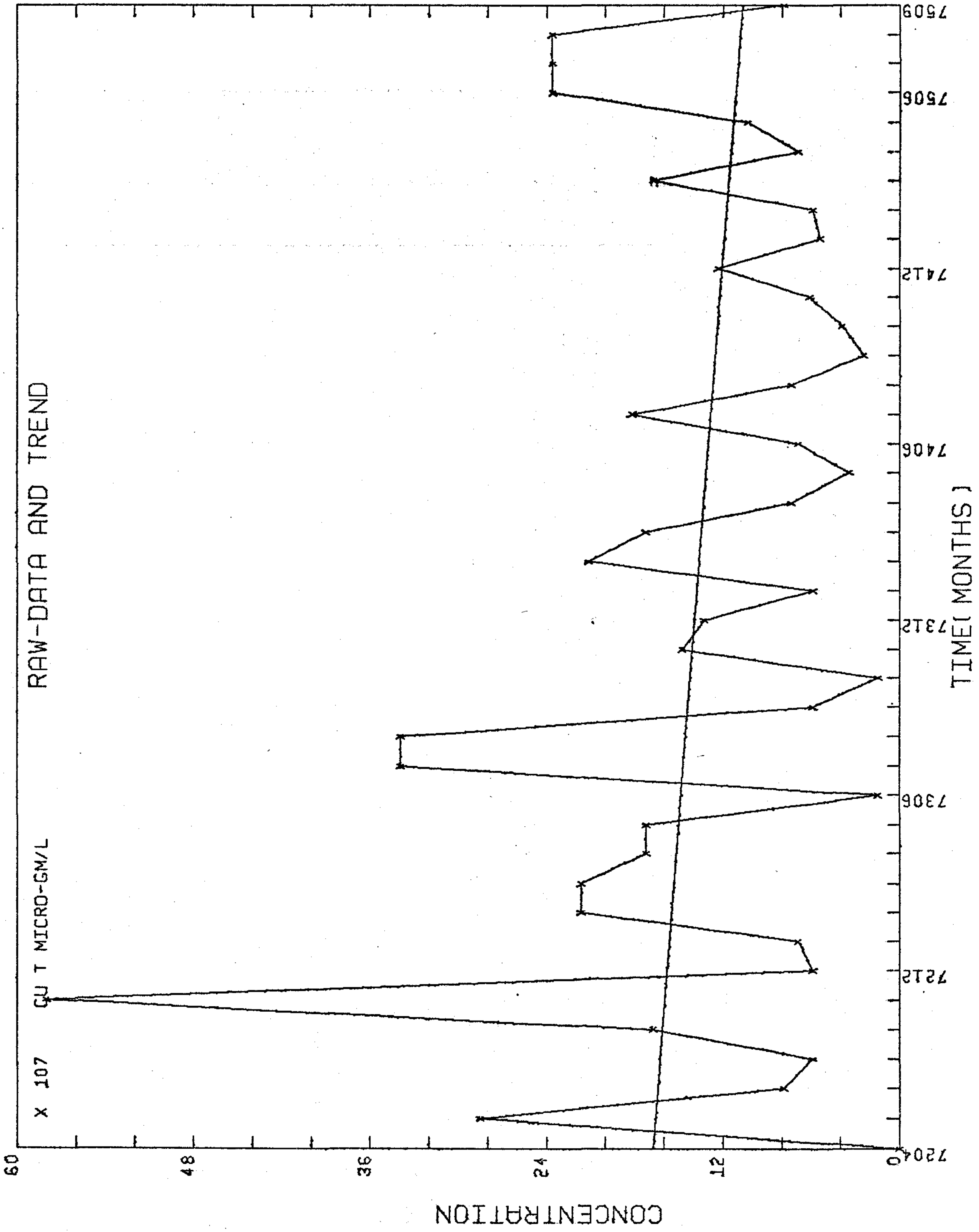


Figure B.4.3: Fe T data and trend, station 4 (7204-7509)

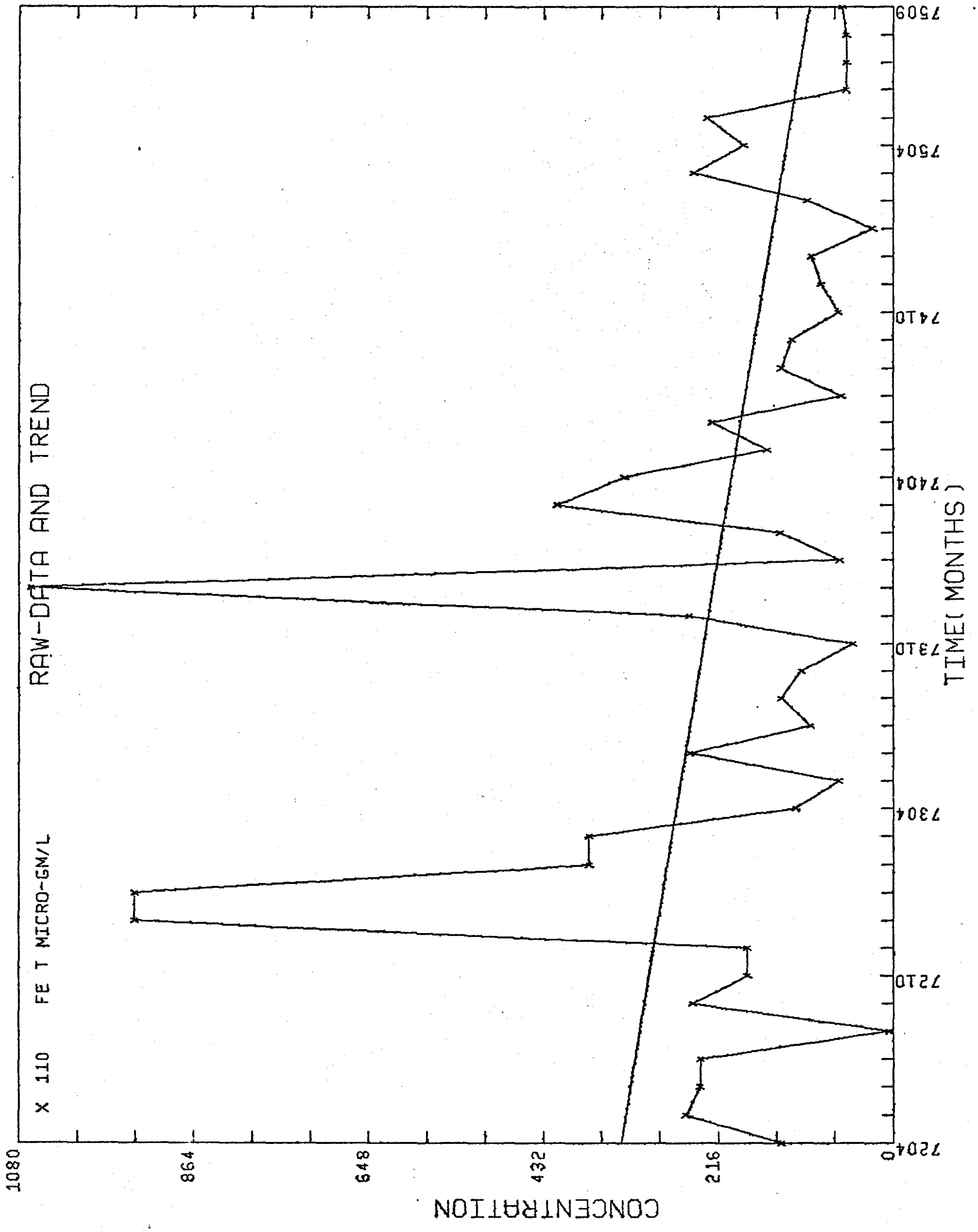


Figure B.4.4: Ni T data and trend, station 4 (7204-7509)

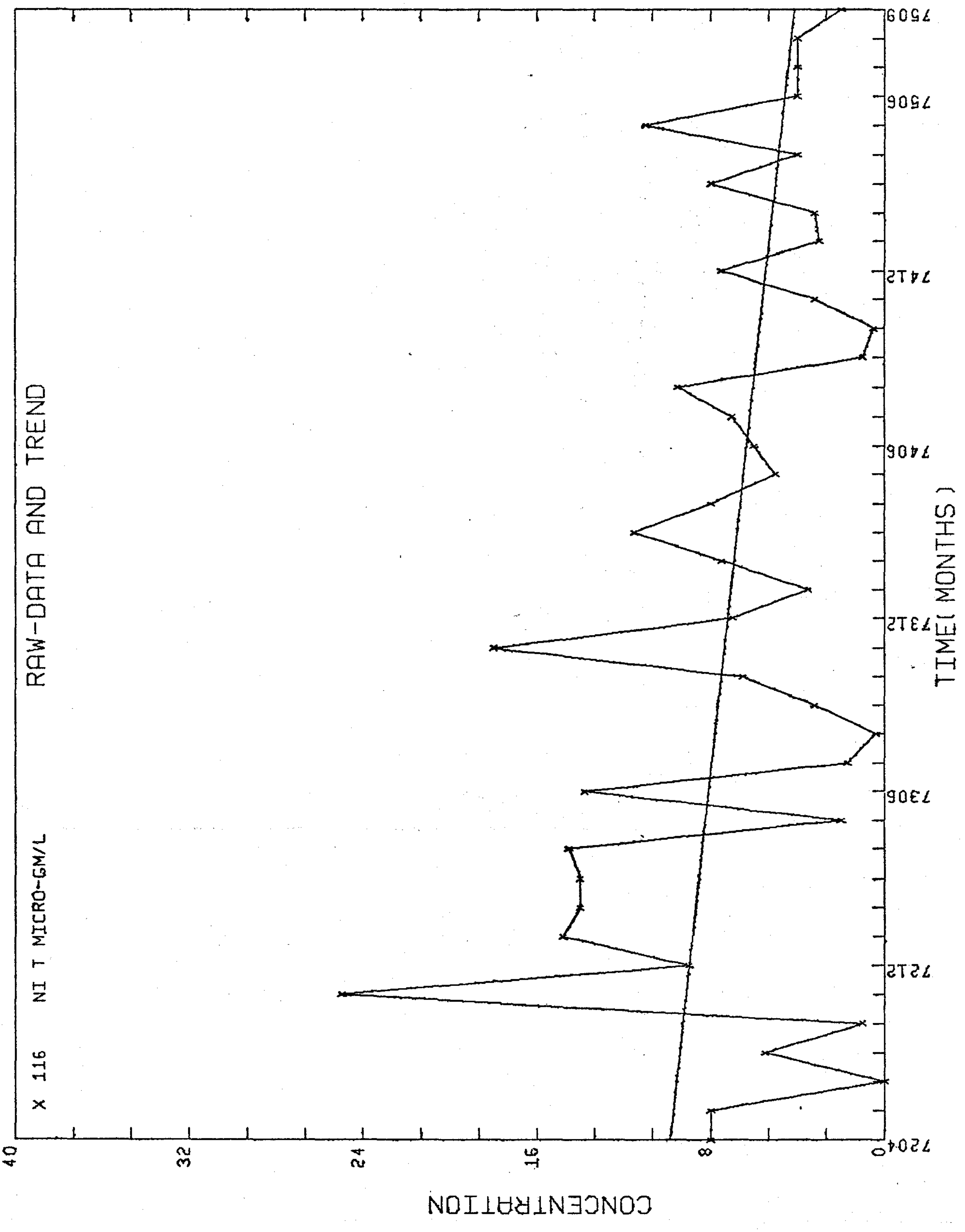


Figure B.4.5: Zn T data and trend, station 4 (7204-7509)

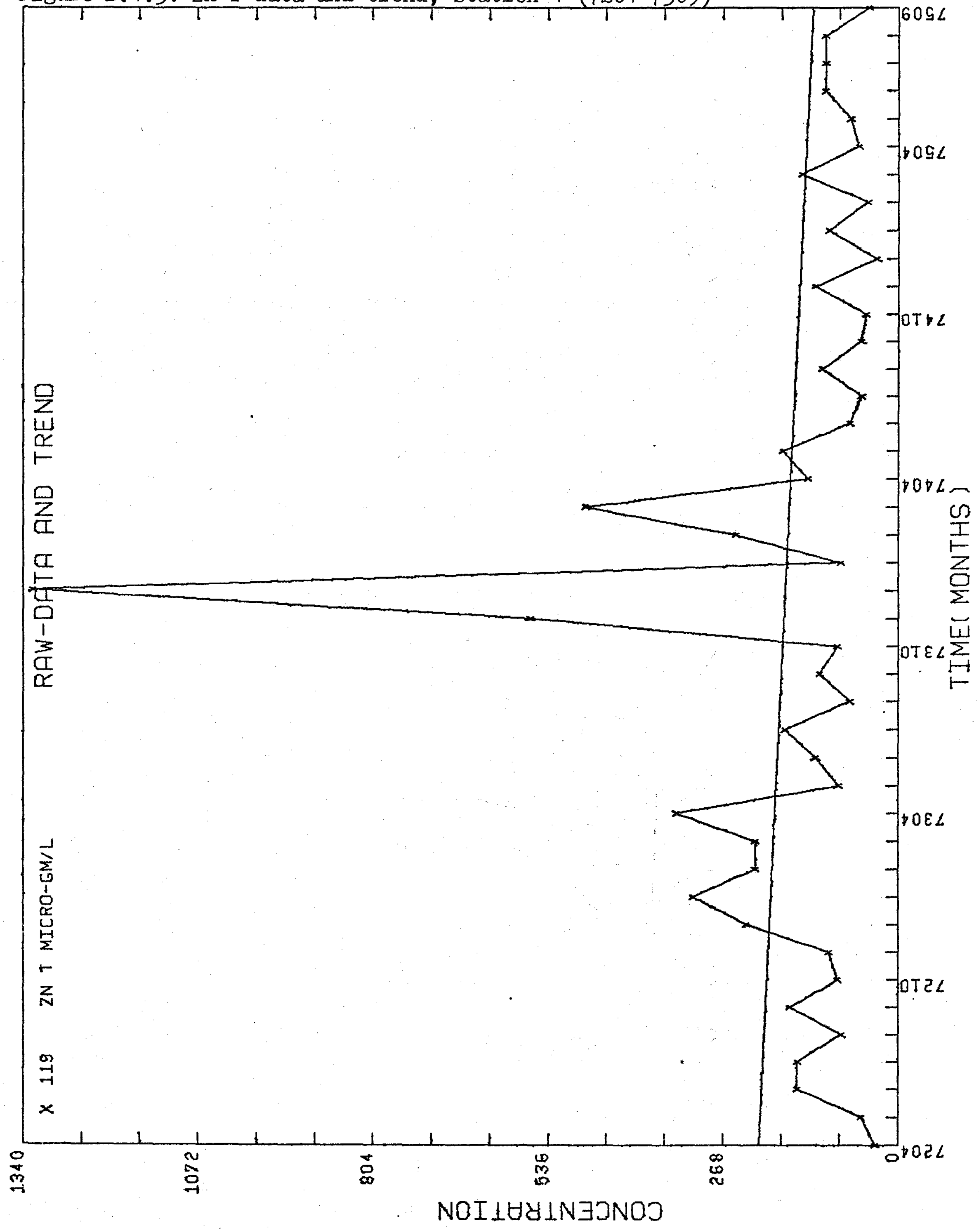


Figure B.4.6: pH data and trend, station 4 (7203-7509)

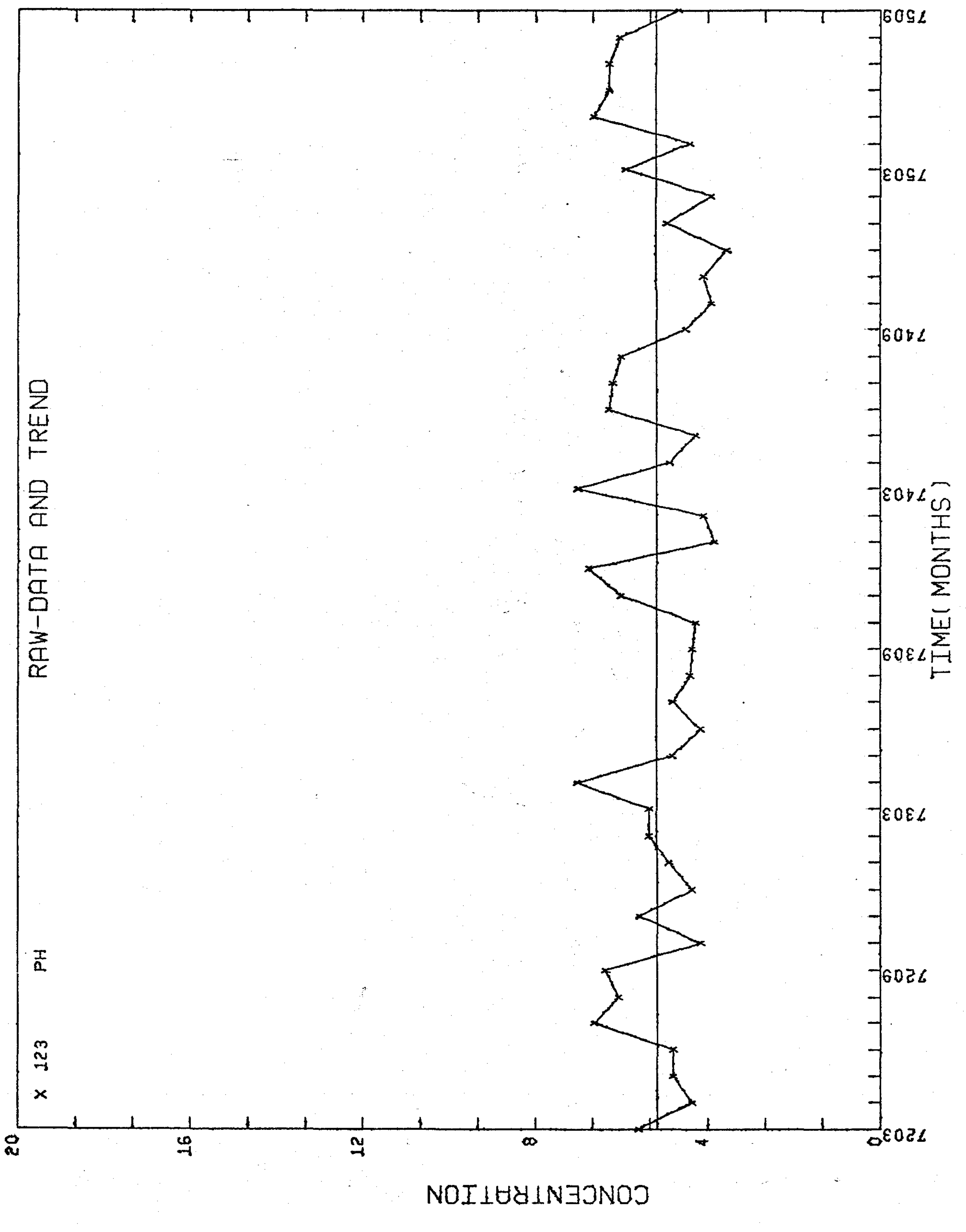
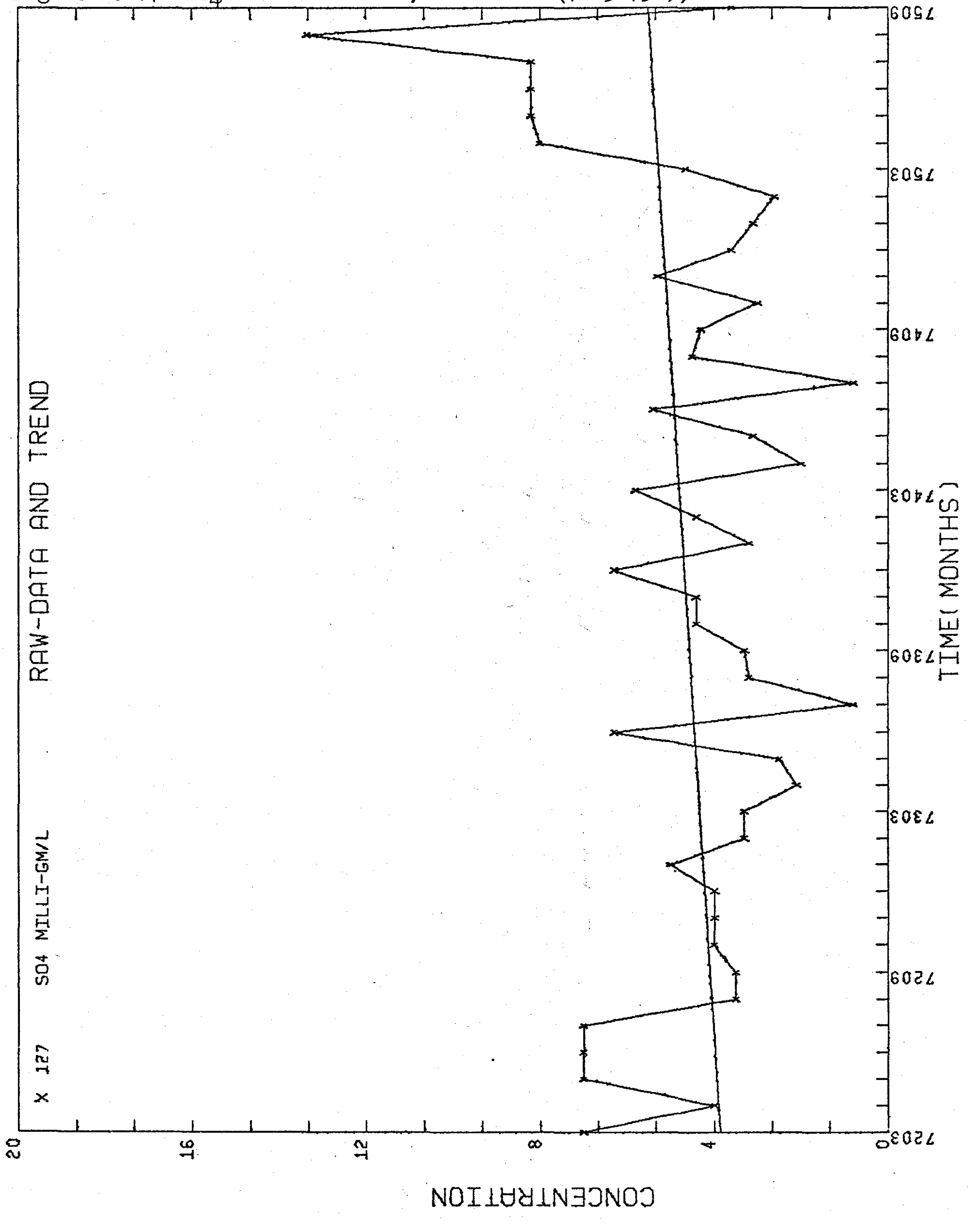


Figure B.4.7: SO_4 data and trend, station 4 (7203-7509)



B.5 Data, Sudbury Area

Table B.5.1 gives the parameters considered and the length of the time series. The data are displayed graphically in the figures to follow.

TABLE B.5.1: Parameters studied for the Sudbury area

Parameters	Code No.	Time Interval
Cd T	101	7204-7511
Cu F	106	7204-7511
Cu T	107	7208-7511
Fe T	109	7204-7511
Pb T	113	7201-7511
Ni T	116	7208-7511
Zn F	118	7204-7511
Zn T	119	7204-7511
SP CON	122	7108-7511
pH	123	7108-7511
SO_4	127	7108-7511

Figure B.5.1: Cd T data and trend, Sudbury area (7204-7511)

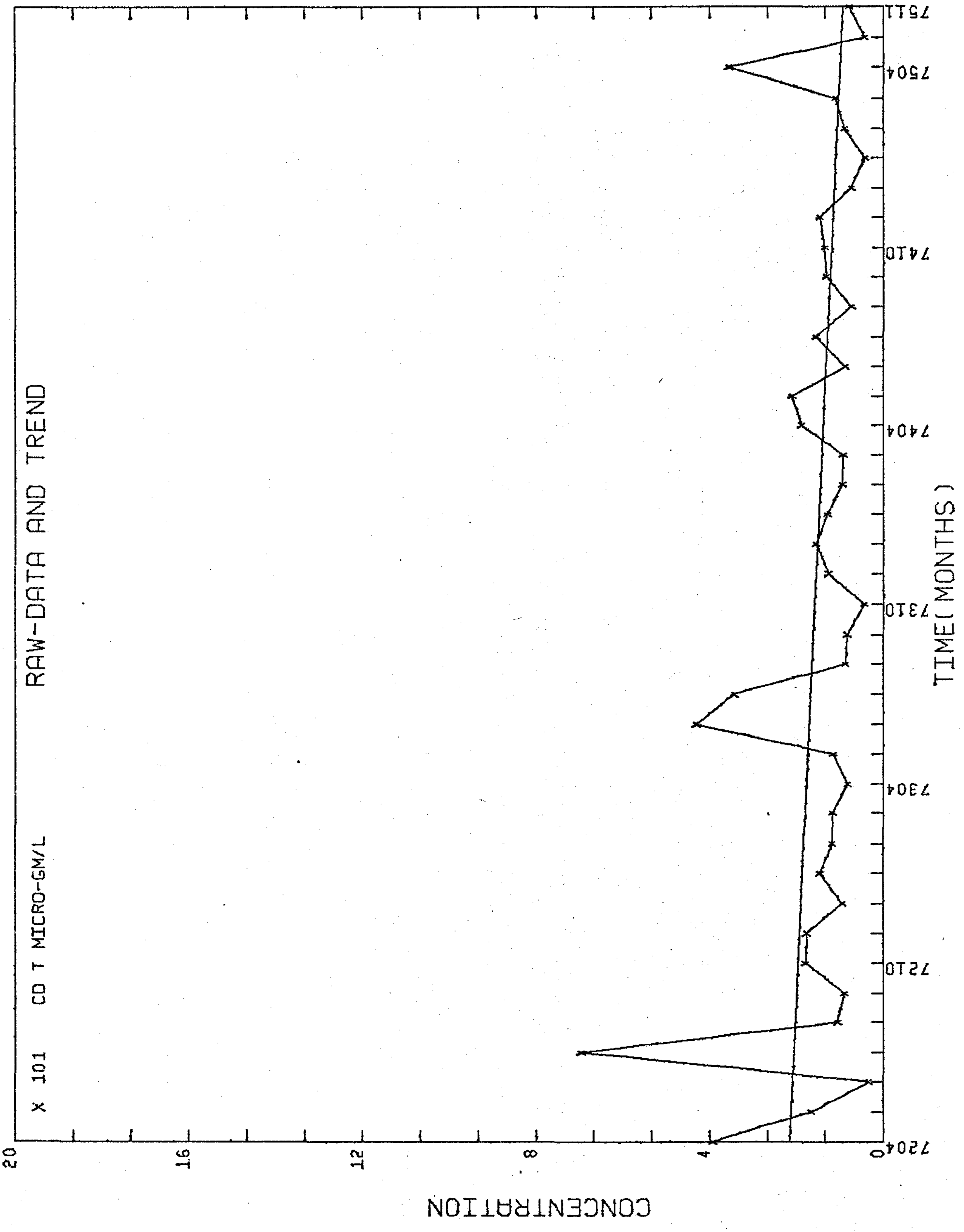


Figure B.5.2: Cu F data and trend, Sudbury area (7204-7511)

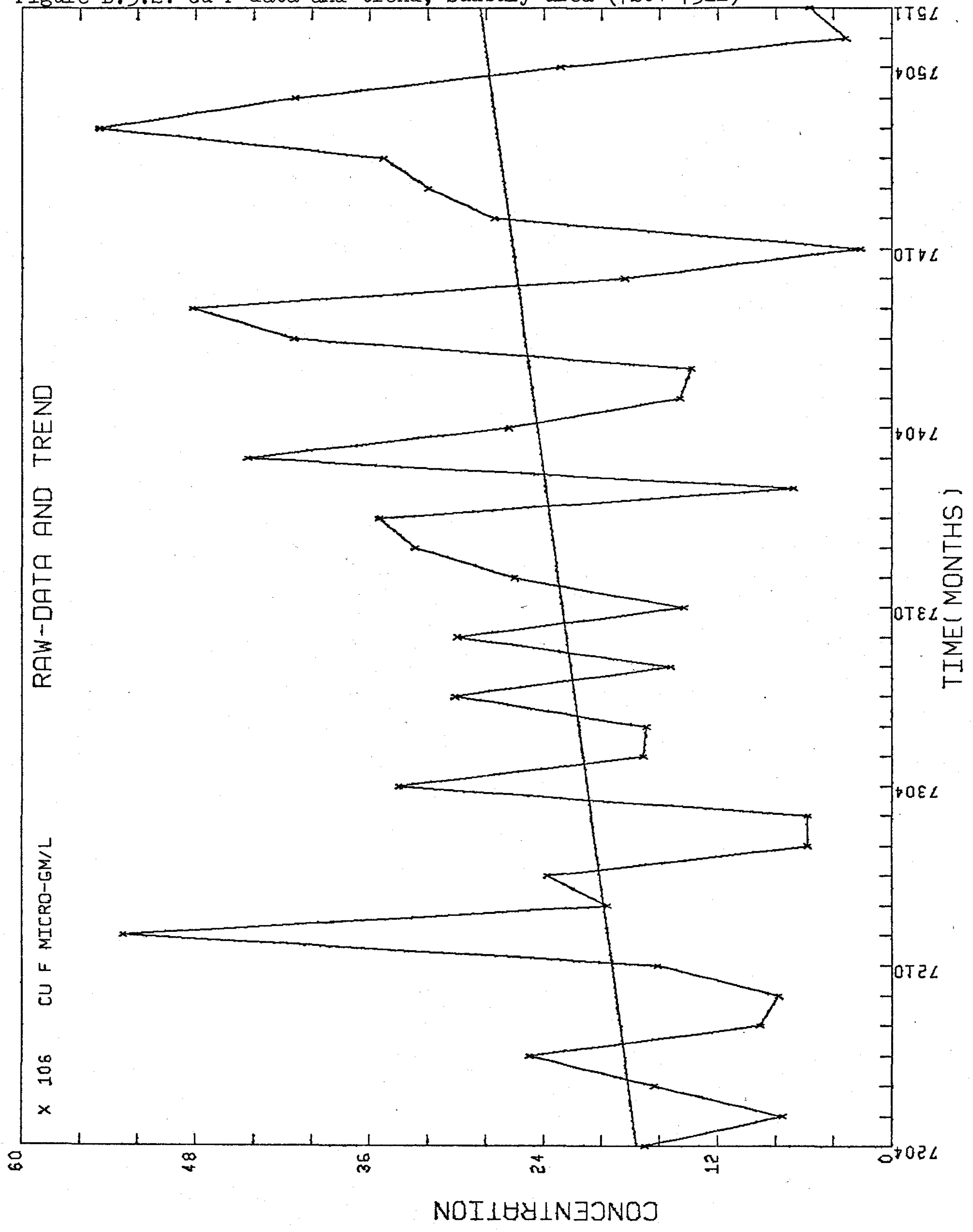


Figure B.5.3: Cu T data and trend, Sudbury area (7208-7511)

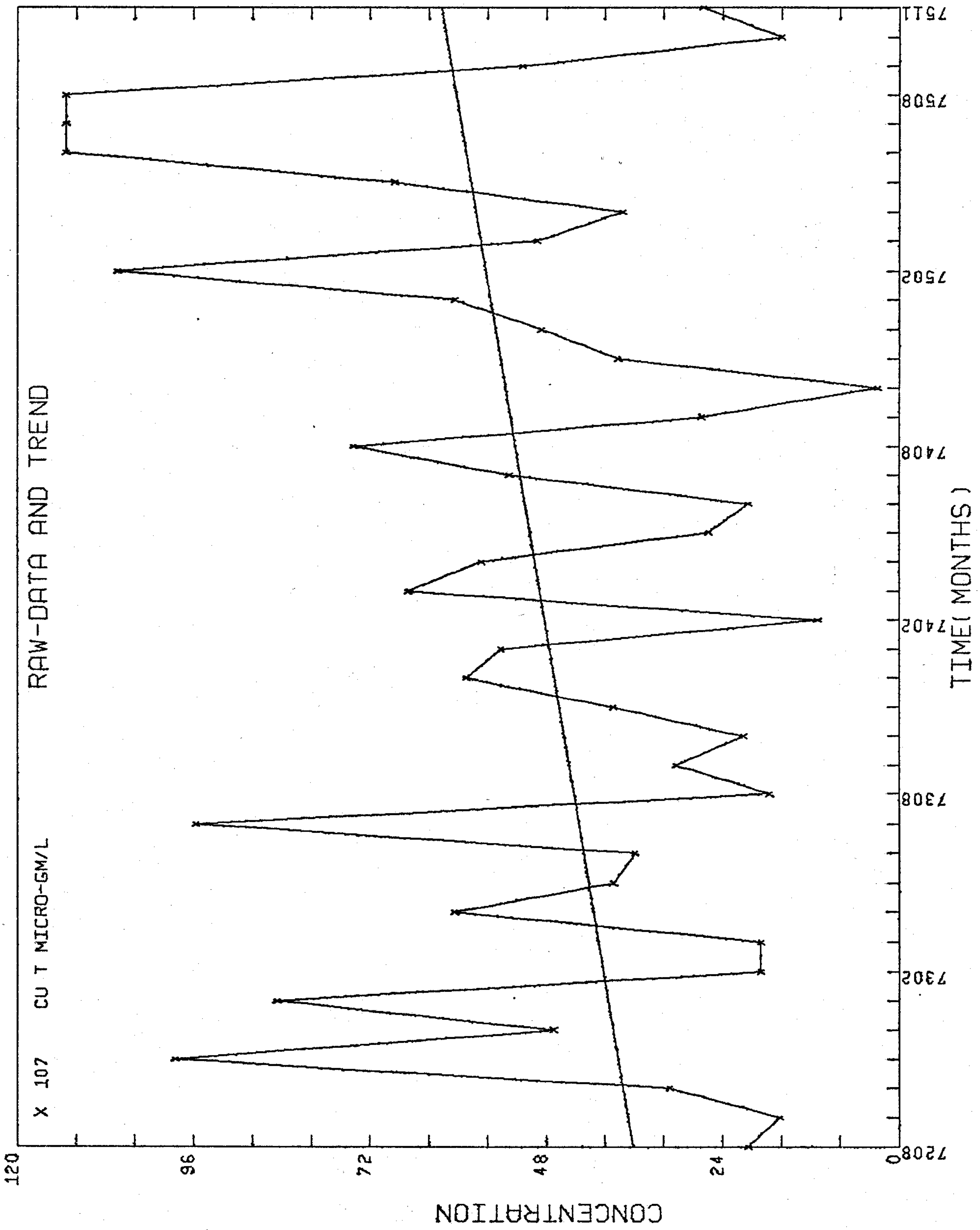


Figure B.5.4: Fe F data and trend, Sudbury area (7204-7511)

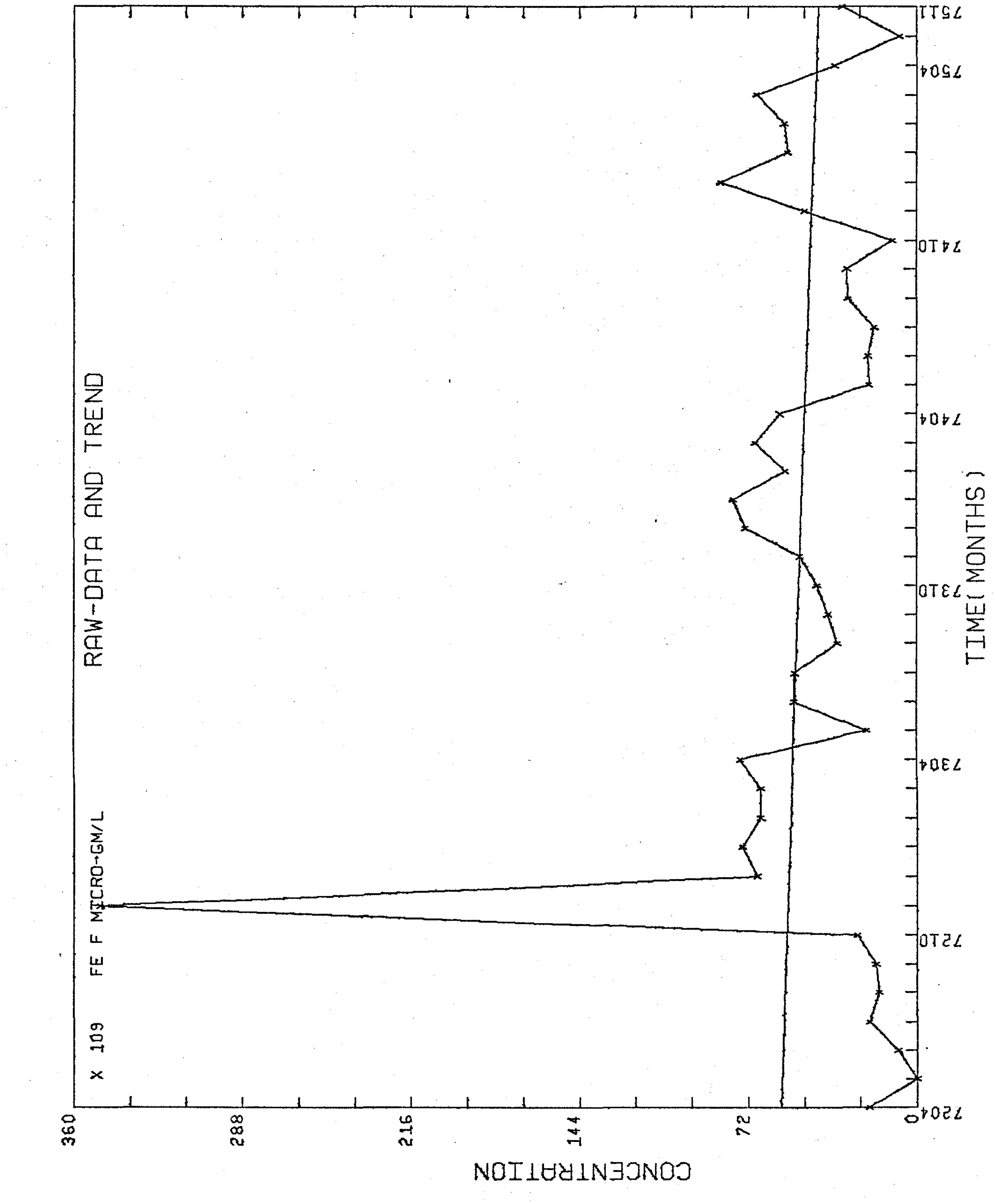


Figure B.5.5: Pb T data and trend, Sudbury area (7201-7511)

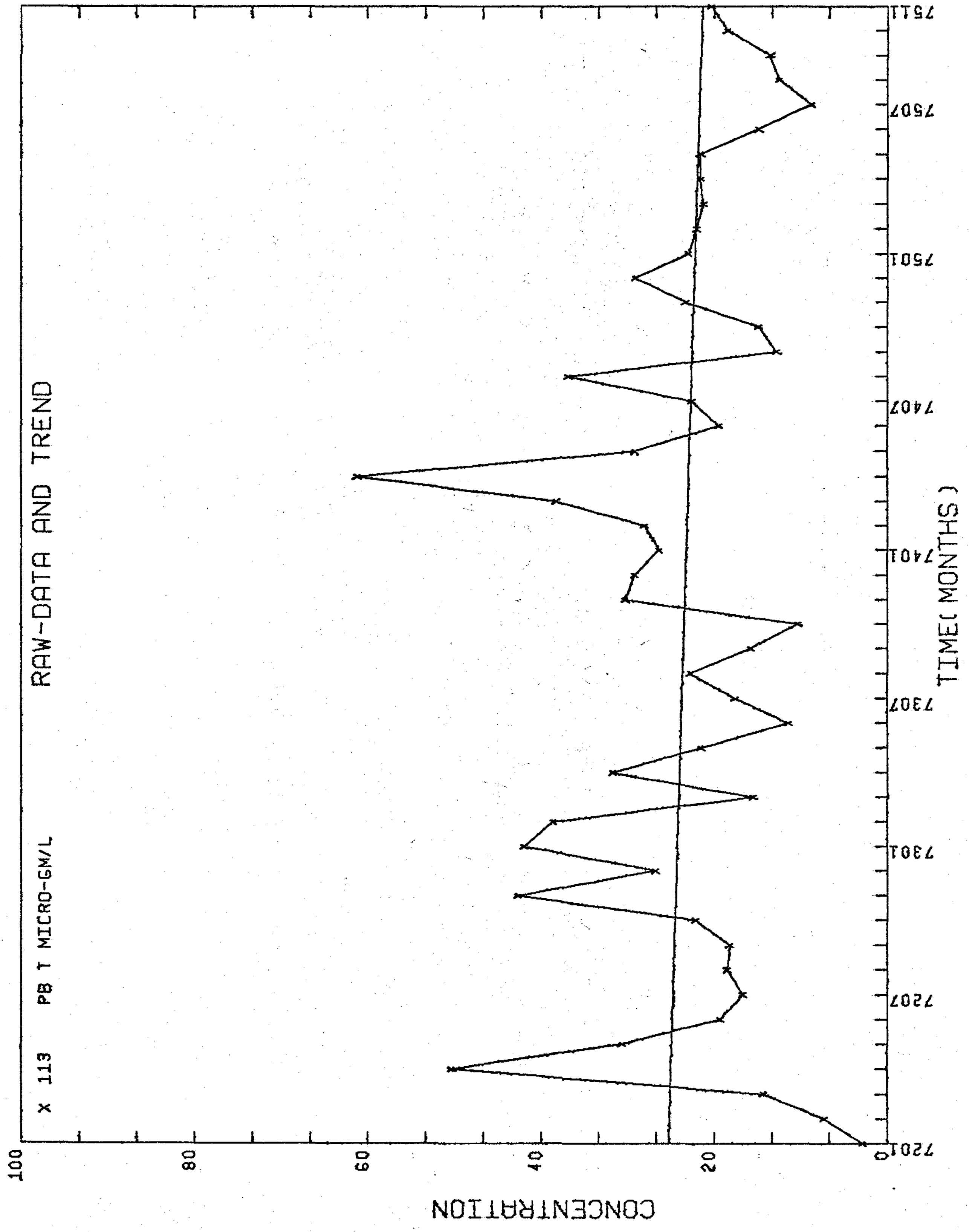


Figure B.5.6: Ni T data and trend, Sudbury area (7208-7511)

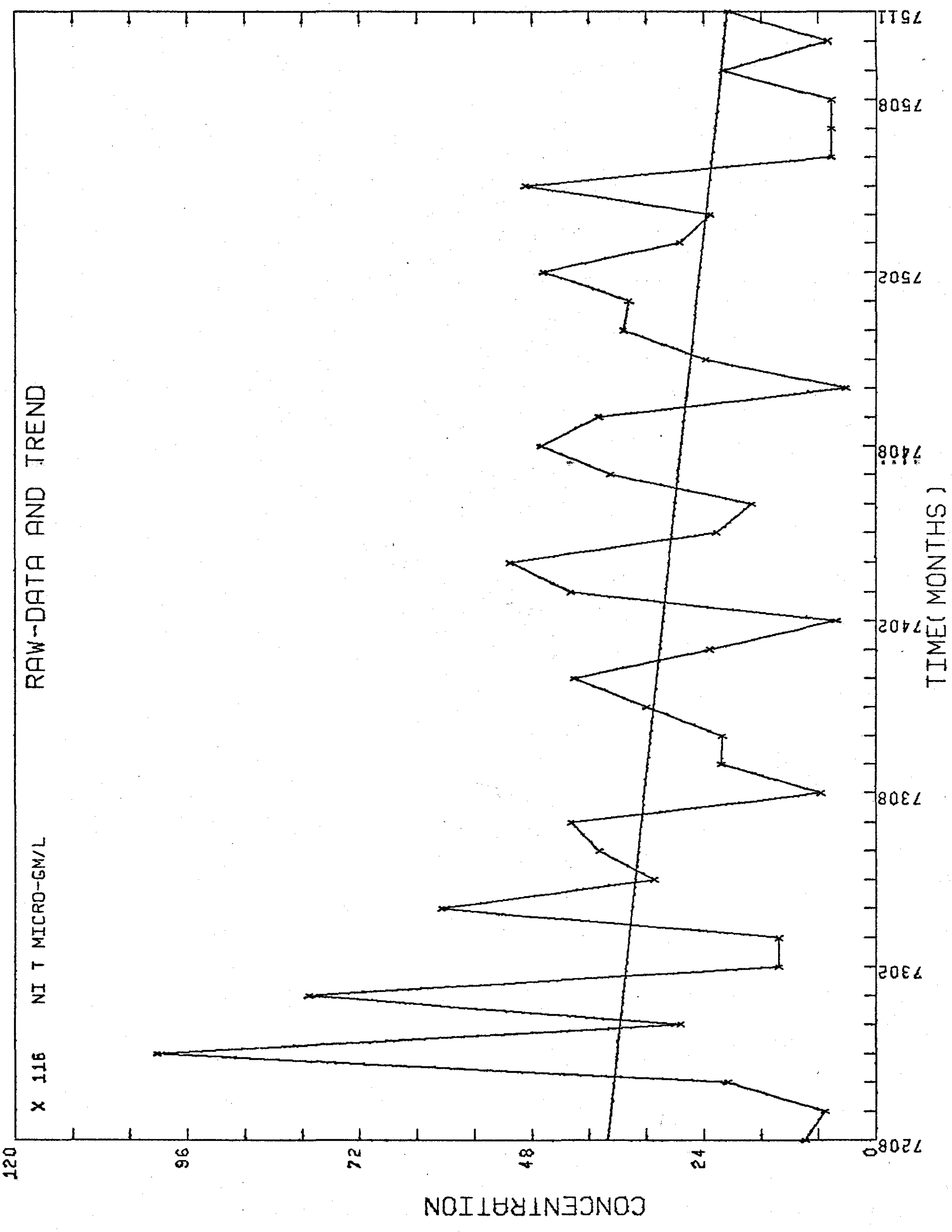


Figure B.5.7: Zn F data and trend, Sudbury area (7204-7511)

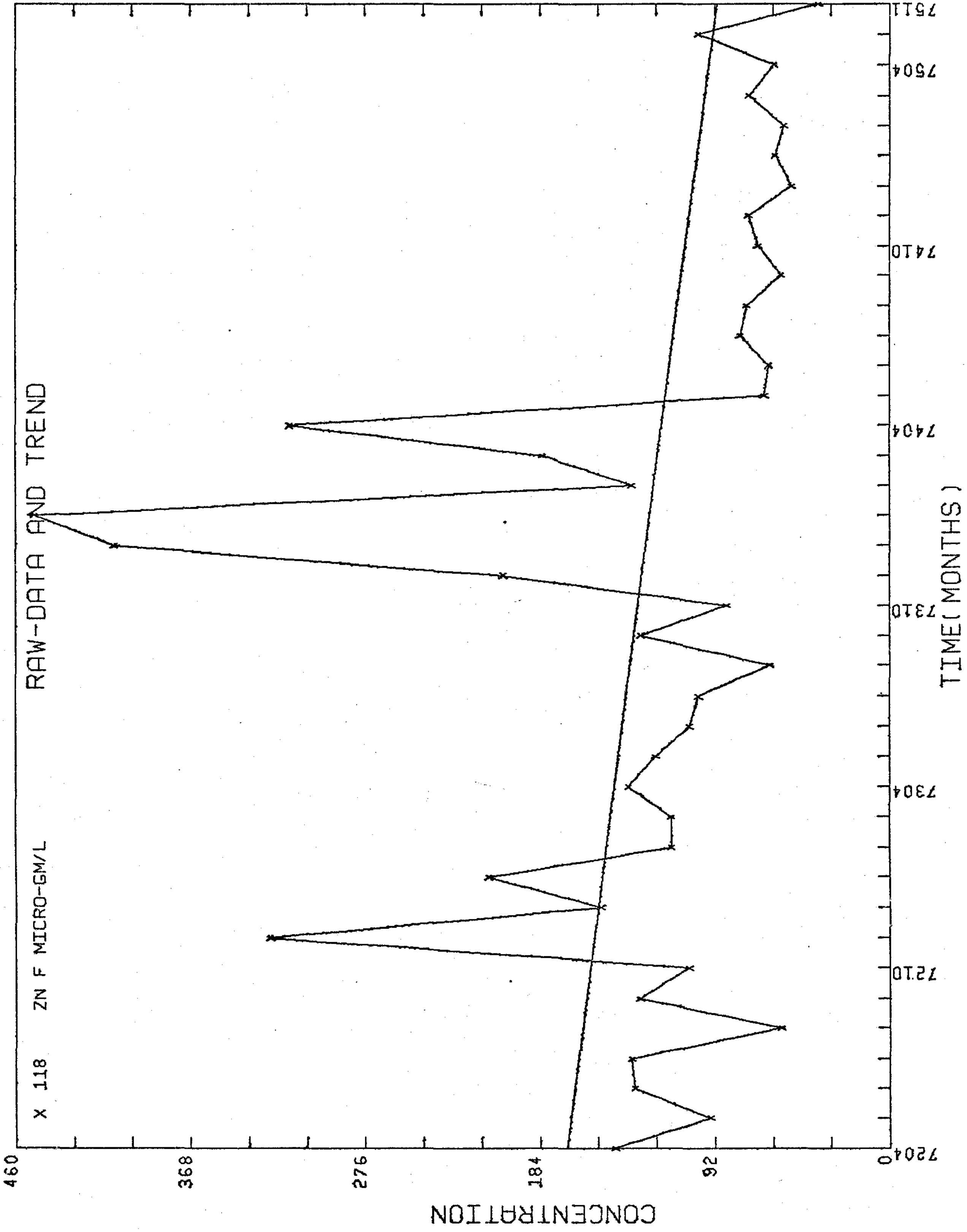


Figure B.5.8: Zn T data and trend, Sudbury area (7204-7511)

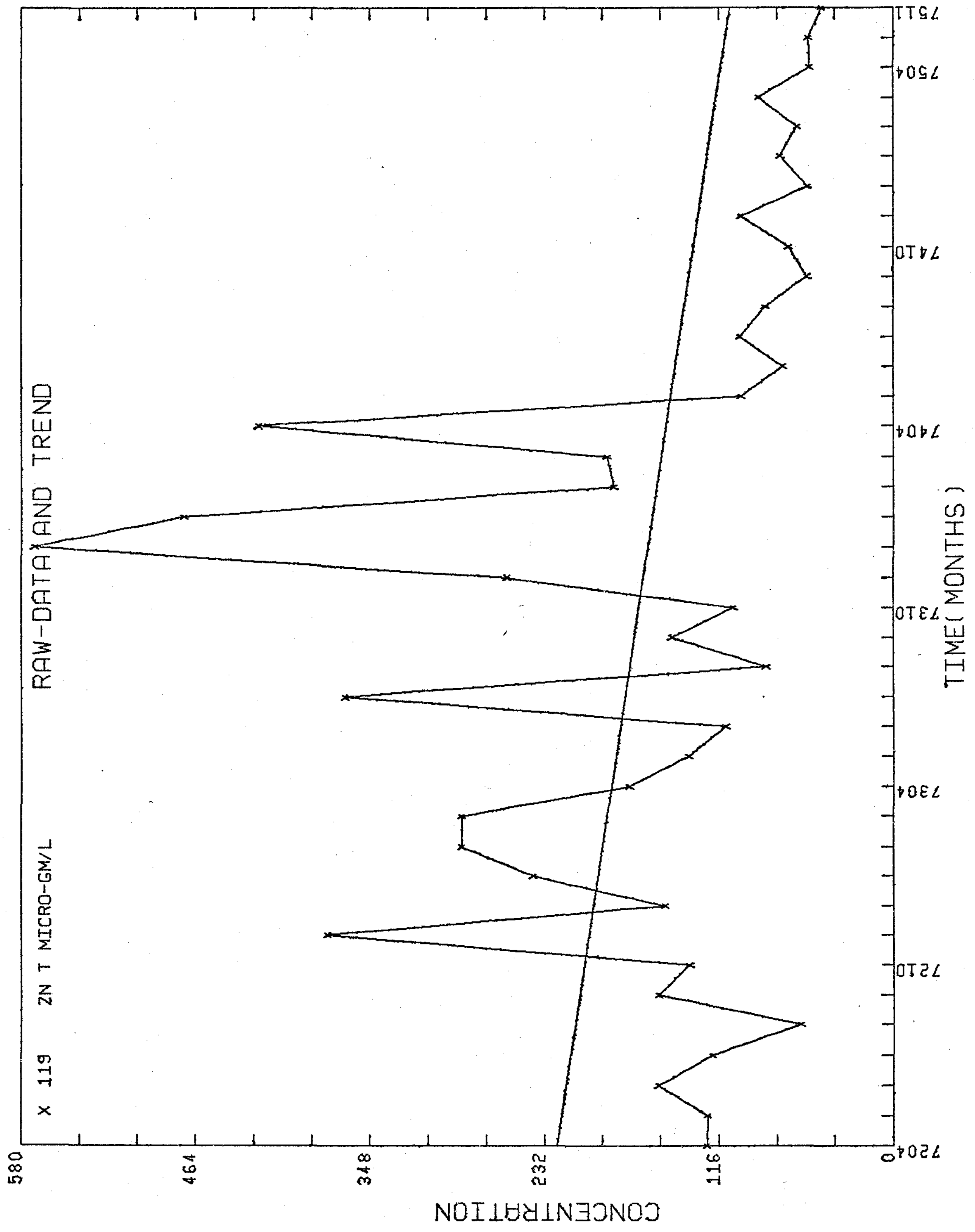


Figure B.5.9: SP CON data and trend, Sudbury area (7108-7511)

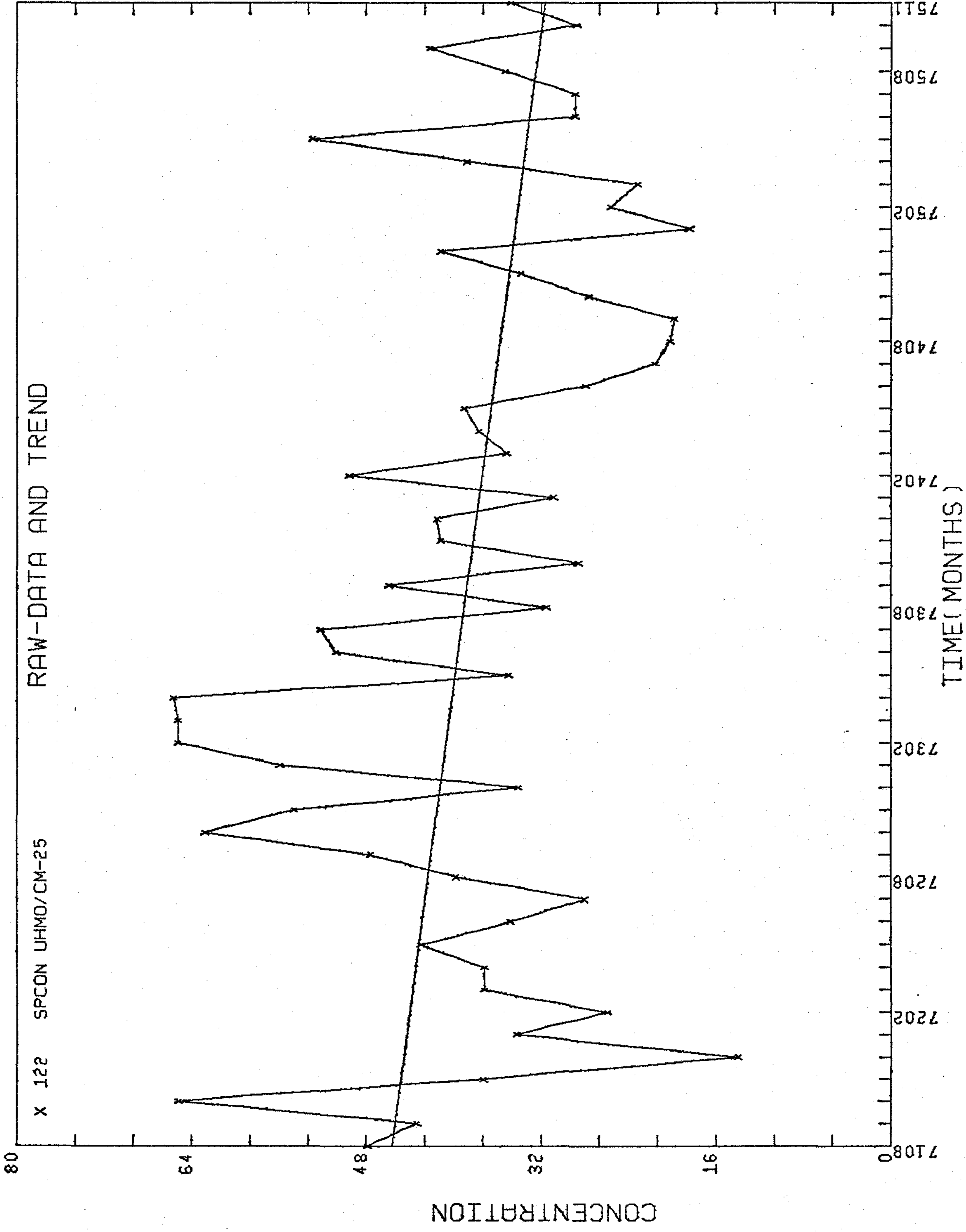


Figure B.5.10: pH data and trend, Sudbury area (7108-7511)

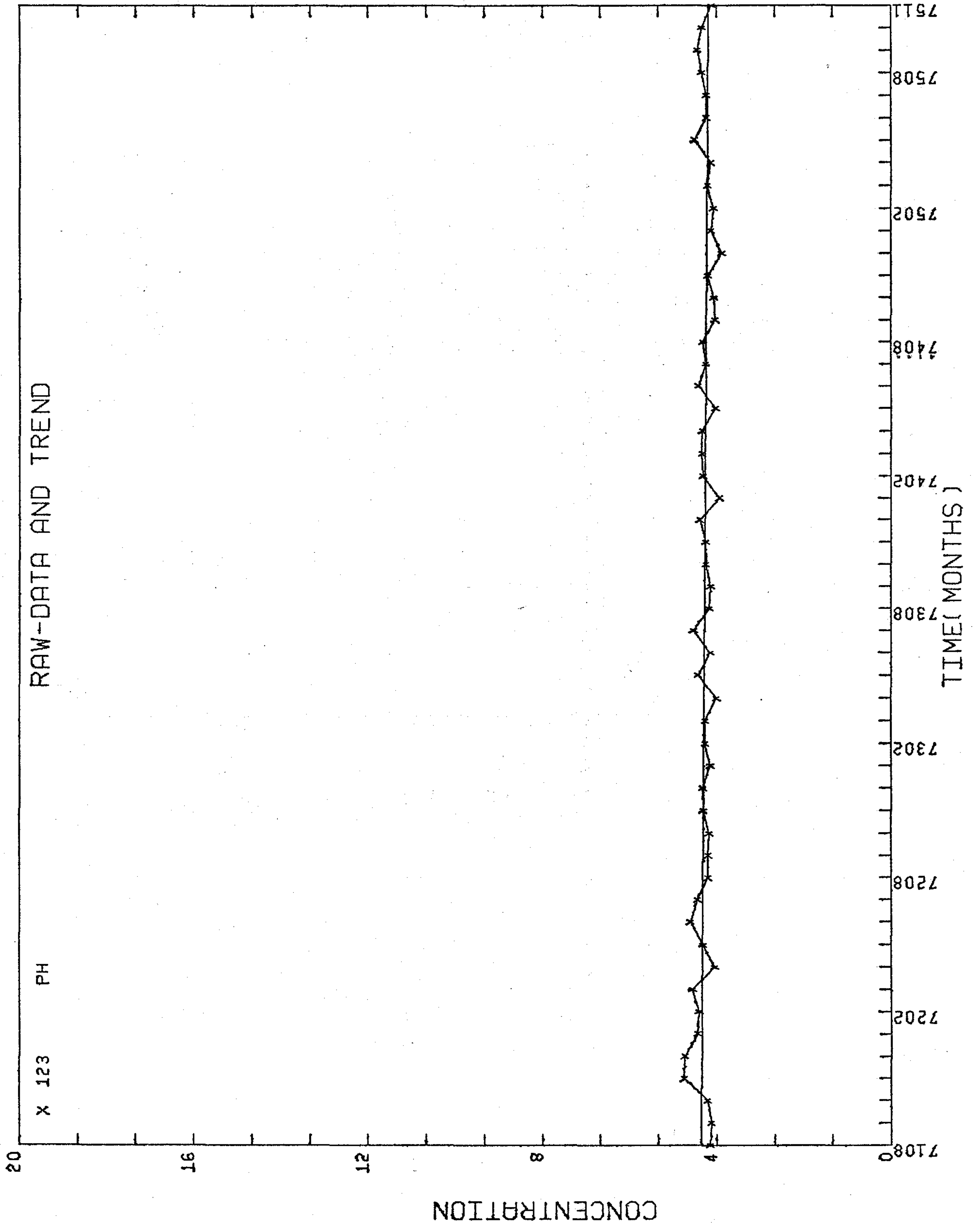
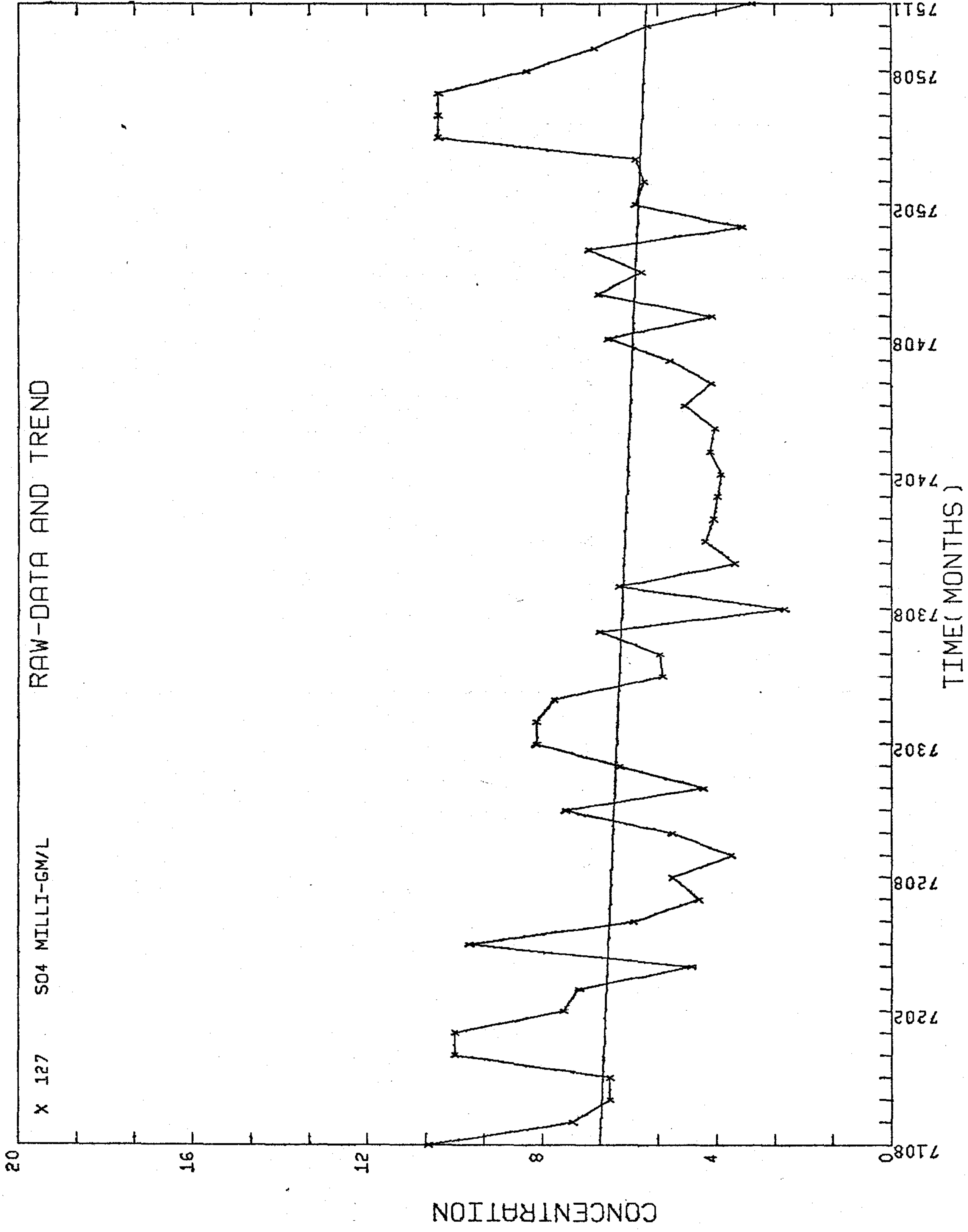


Figure B.5.11: SO_4 data and trend, Sudbury area (7108-7511)



B.6 Data, U.G.L. Area

Table B.6.1 gives the parameters considered and the length of the time series. The data are displayed graphically in the figures to follow.

TABLE B.6.1: Parameters studied for the U.G.L. area

Parameters	Code No.	Time Interval
Cd T	101	7201-7511
Cu F	106	7204-7511
Cu T	107	7201-7511
Fe T	110	7201-7511
Pb T	113	7201-7511
Ni T	116	7201-7511
Zn F	118	7204-7511
Zn T	119	7204-7511
SP CON	122	7108-7511
pH	123	7108-7511
SO_4	127	7108-7511
T PART	128	7204-7506
P T	240	7201-7511

Figure B.6.1: Cd T data and trend, U.G.L. area (7201-7511)

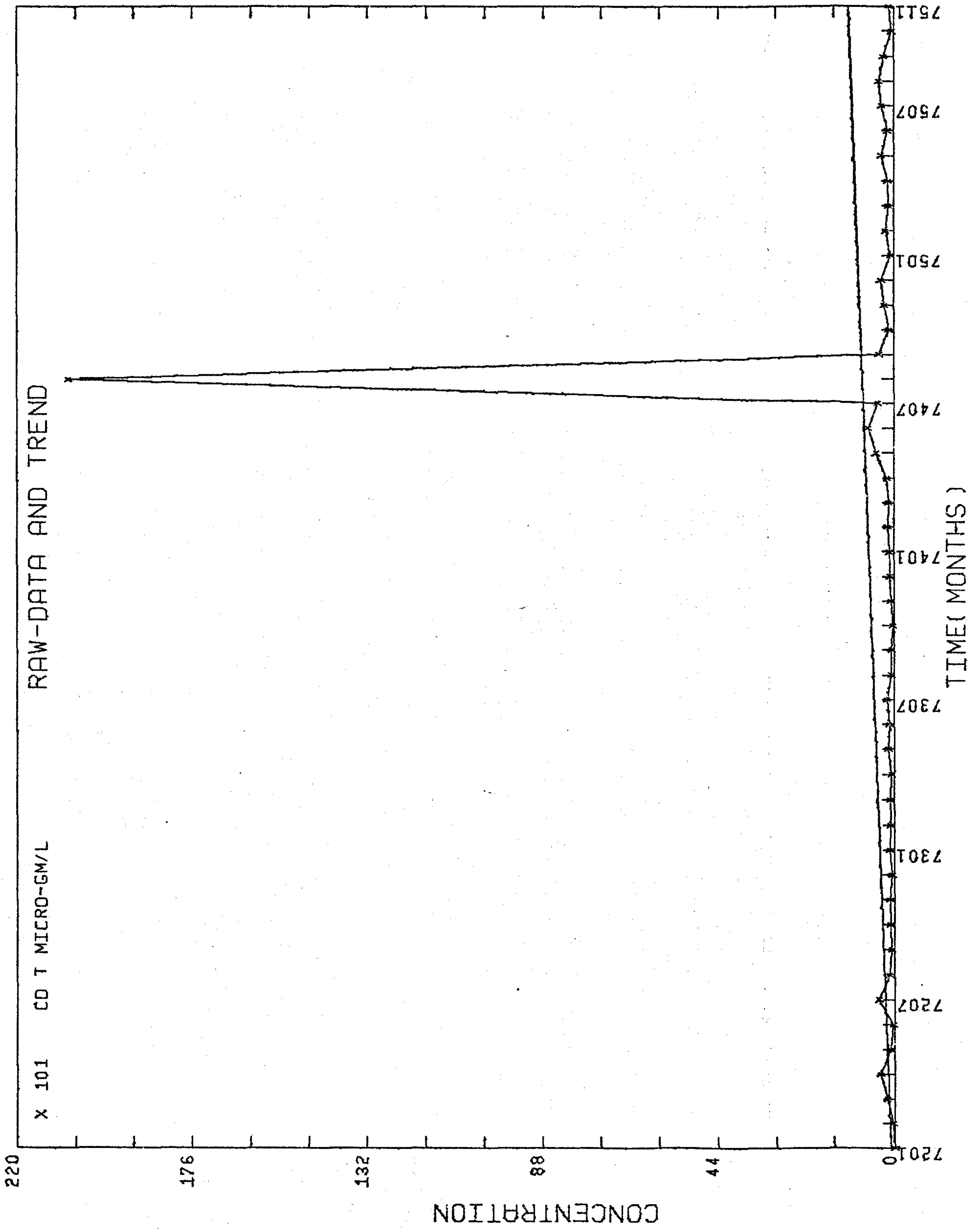


Figure B.6.2: Cu F data and trend, U.G.L. area (7204-7511)

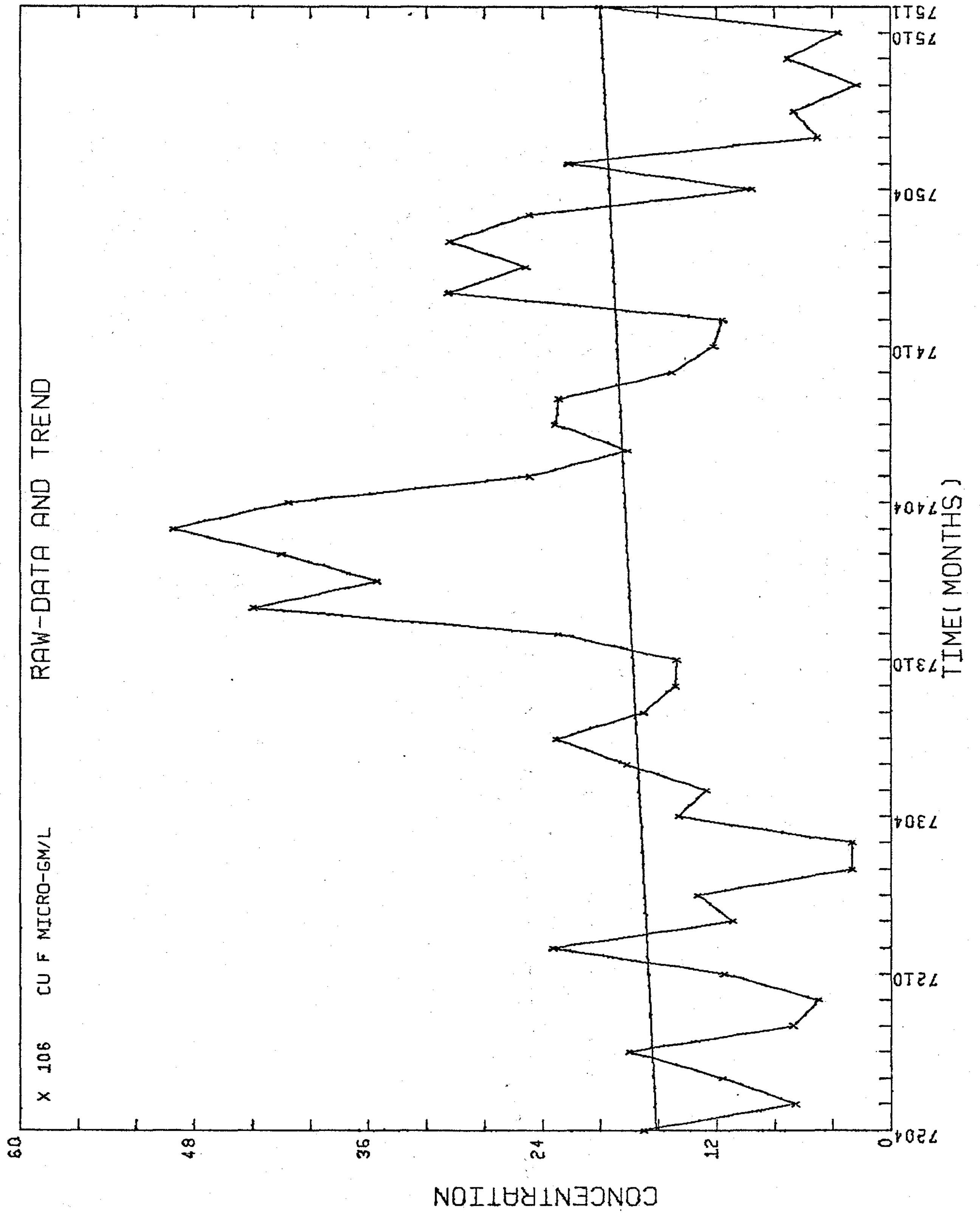


Figure B.6.3: Cu T data and trend, U.G.L. area (7201-7511)

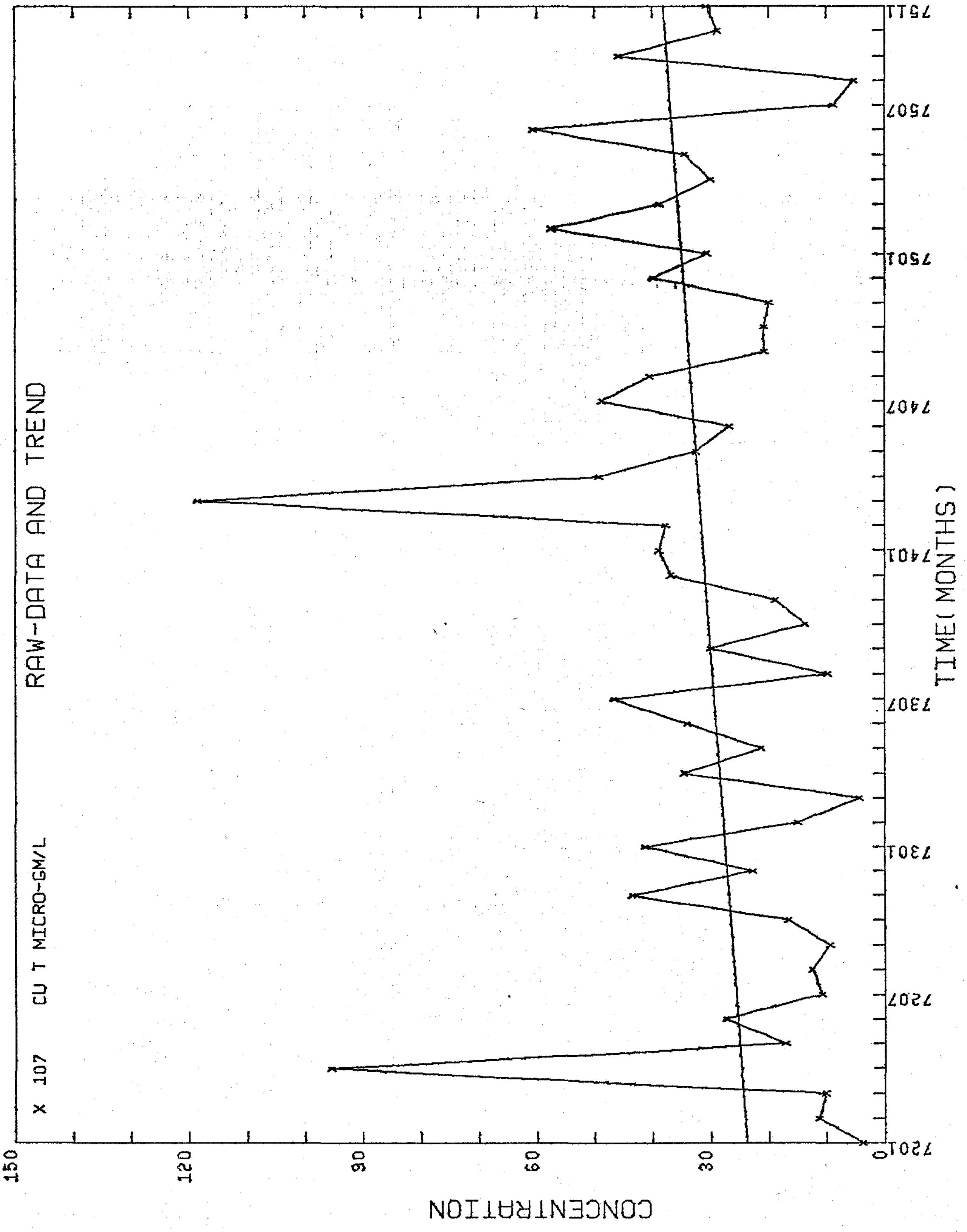


Figure B.6.4: Fe T data and trend, U.G.L. area (7201-7511)

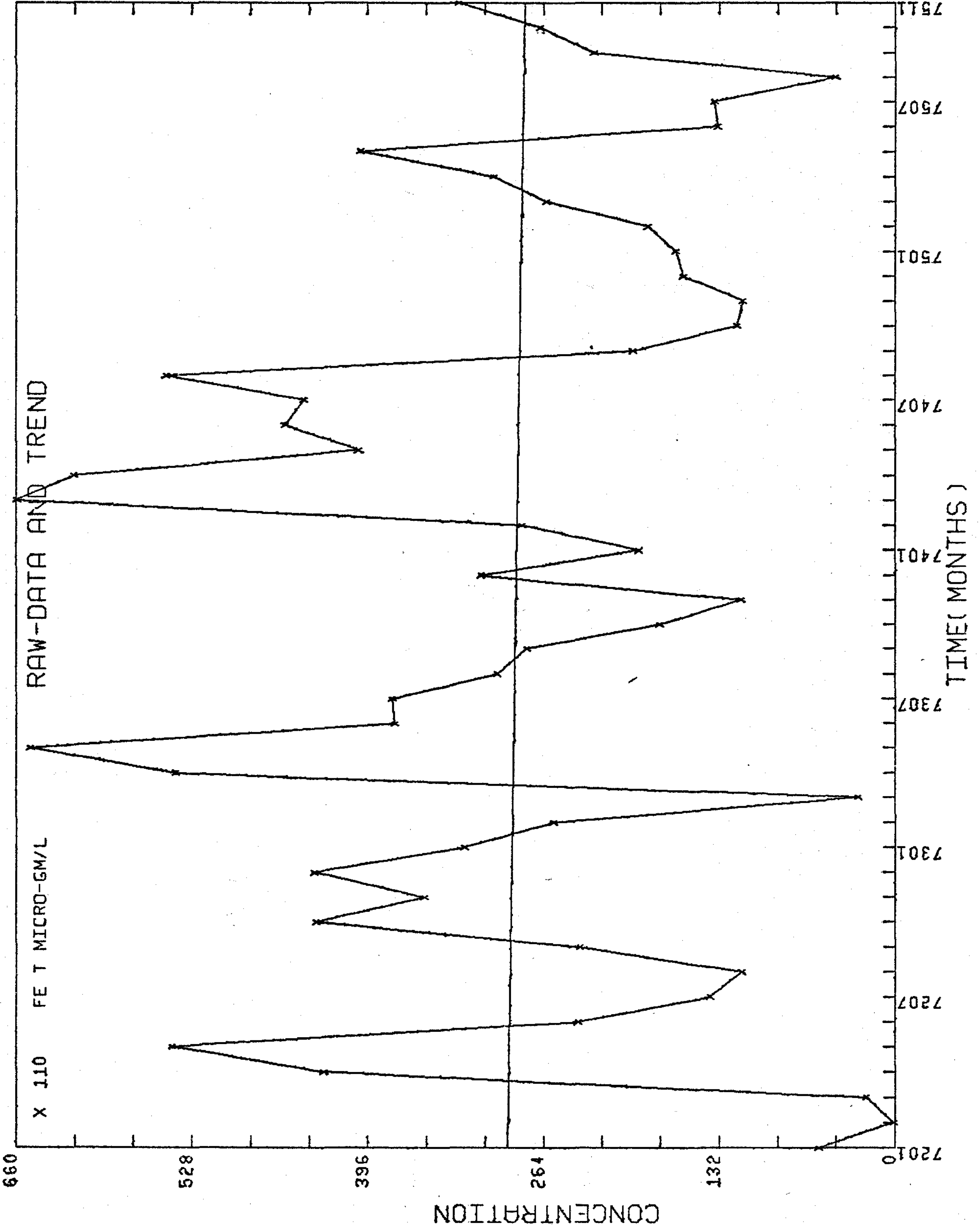


Figure B.6.5: Pb T data and trend, U.G.L. area (7201-7511)

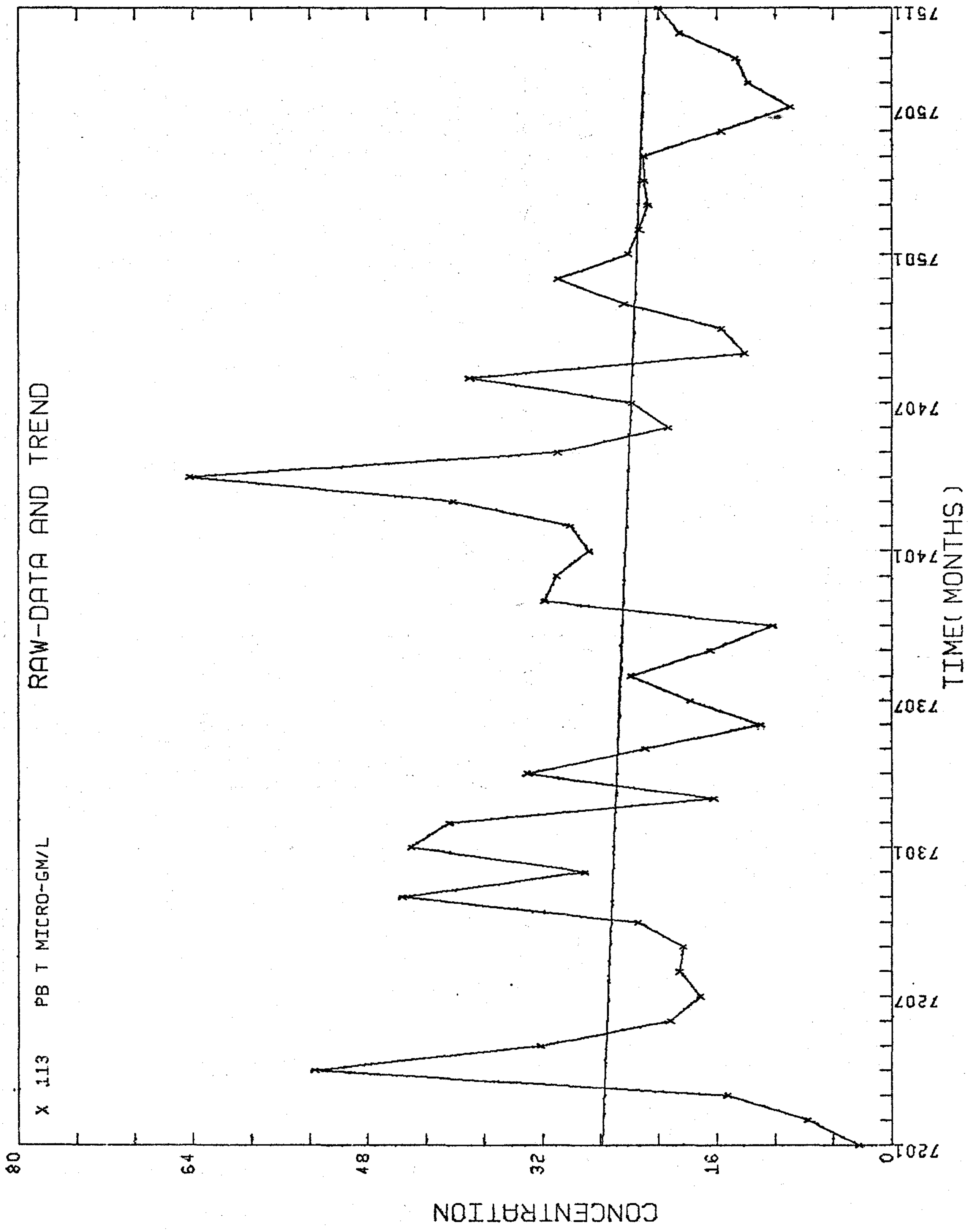


Figure B.6.6: Ni T data and trend, U.G.L. area (7201-7511)

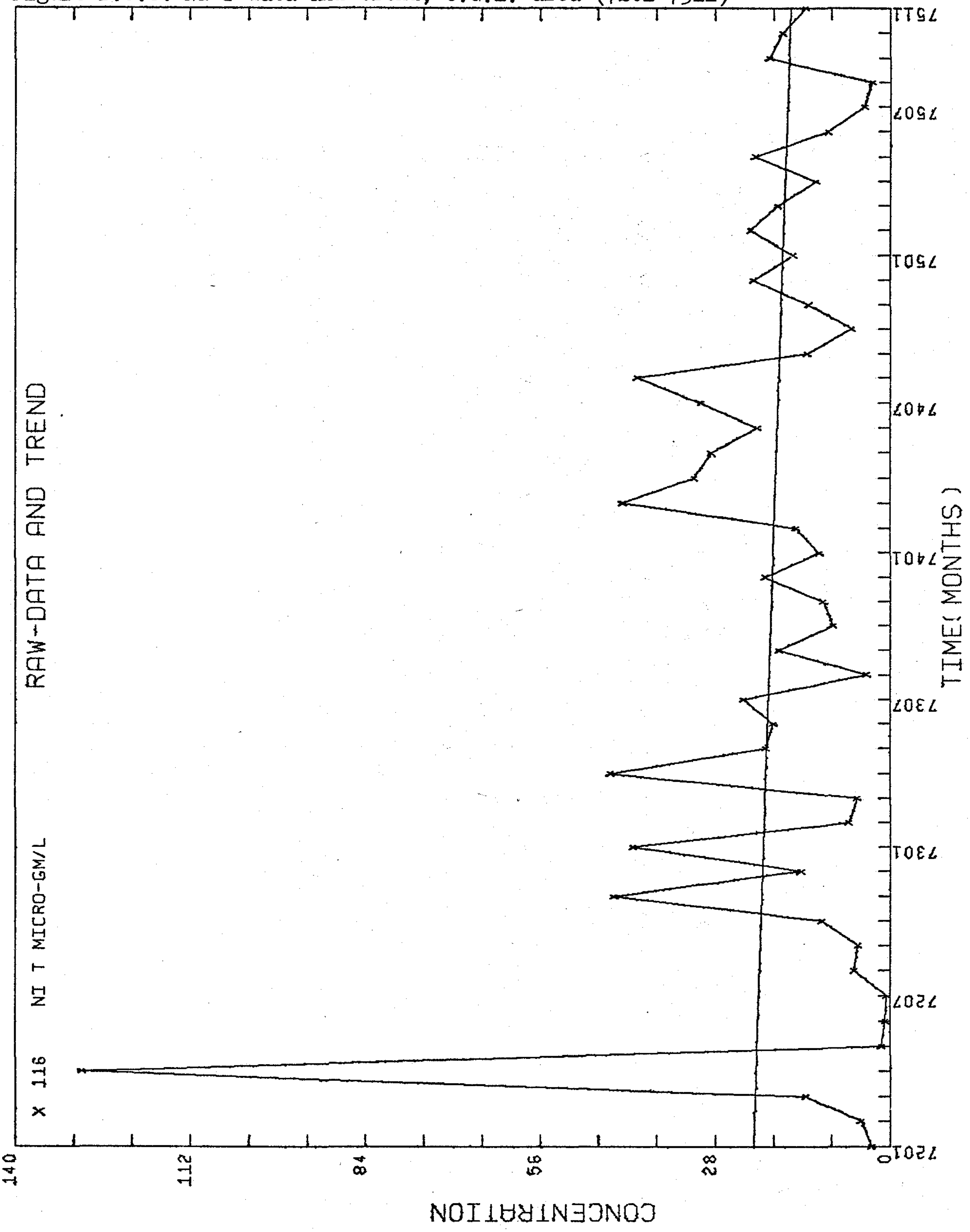


Figure B.6.7: Zn F data and trend, U.G.L. area (7204-7511)

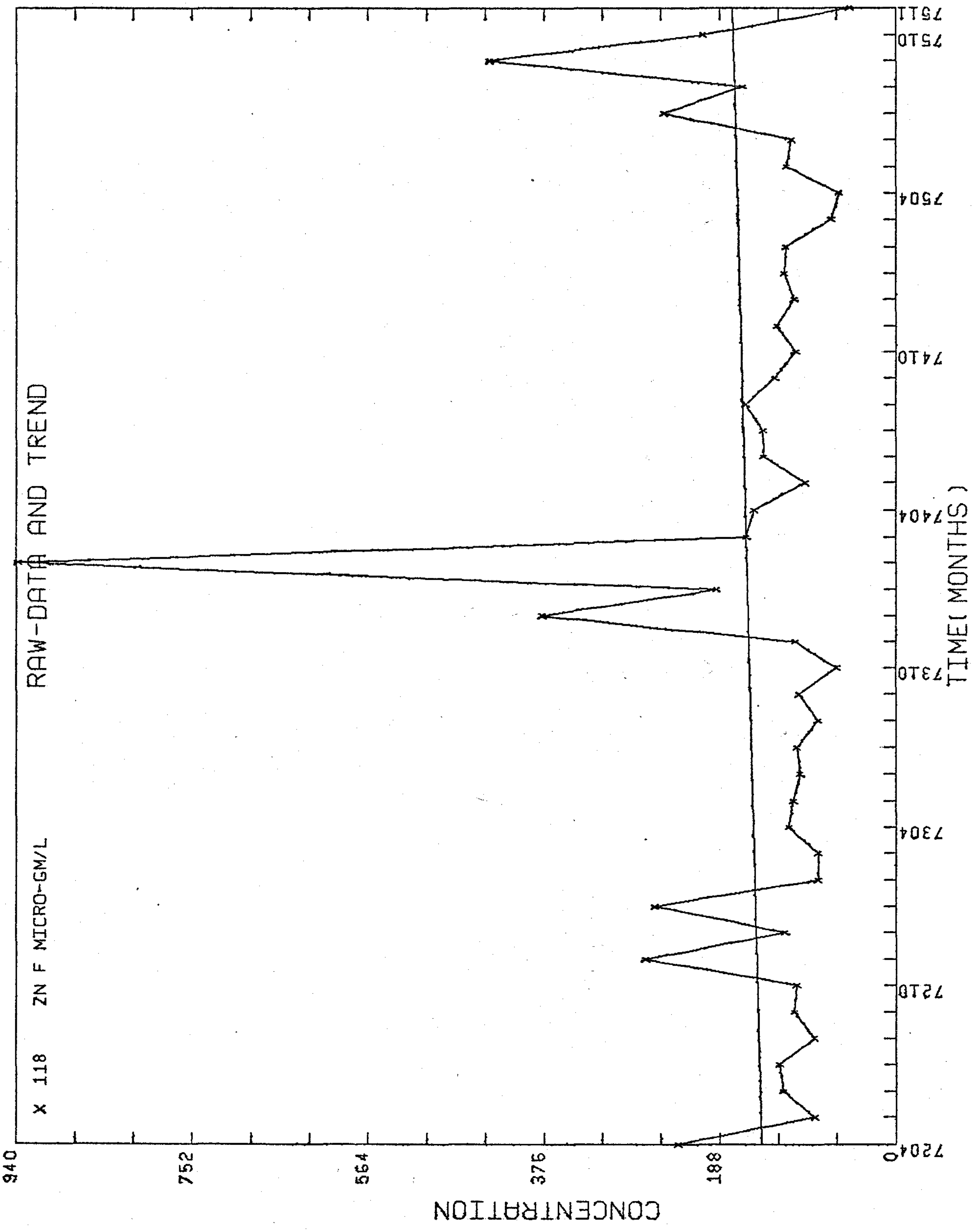


Figure B.6.8: Zn T data and trend, U.G.L. area (7204-7511)

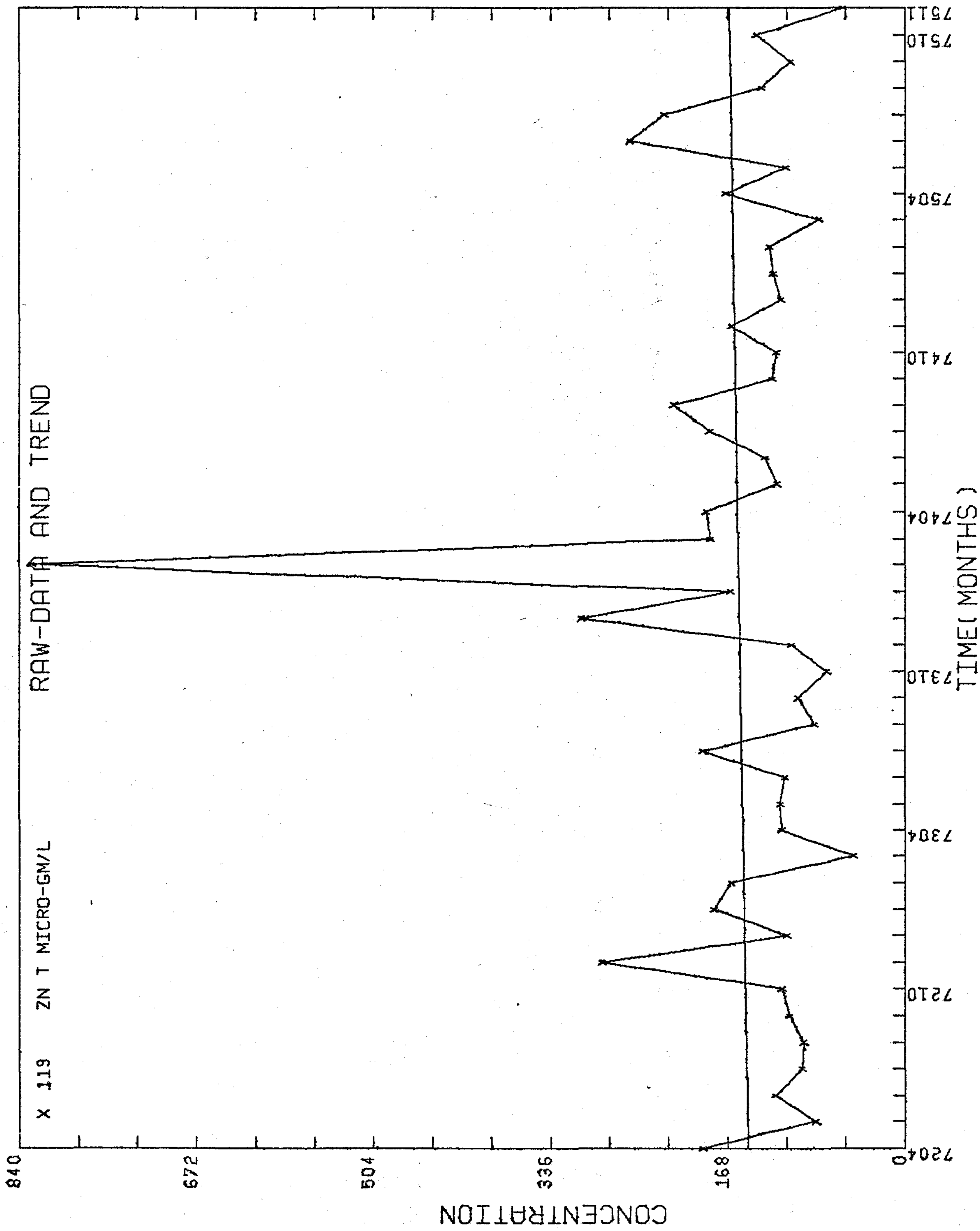


Figure B.6.9: SP CON data and trend, U.G.L. area (7108-7511)

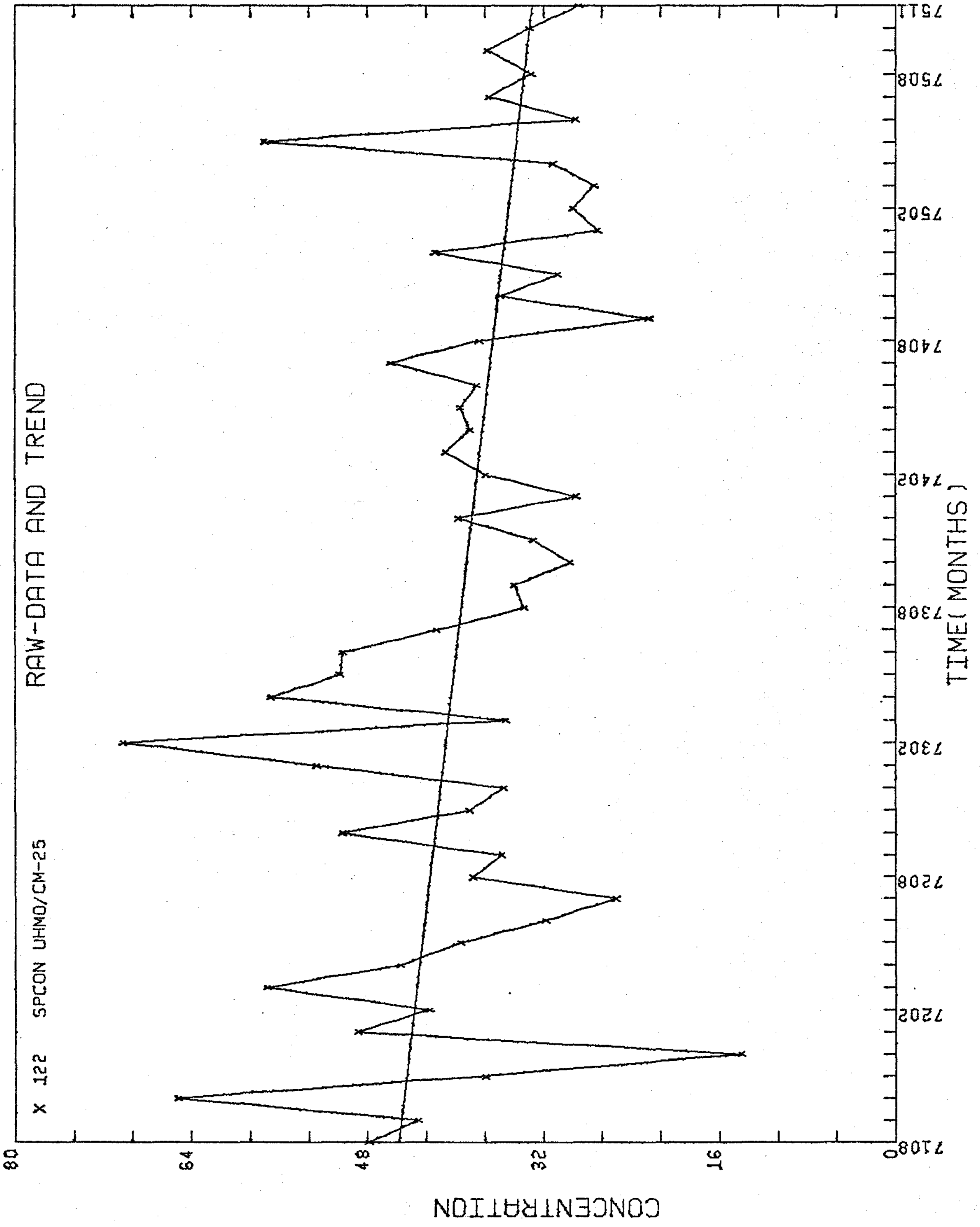


Figure B.6.10: pH data and trend, U.G.L. area (7108-7511)

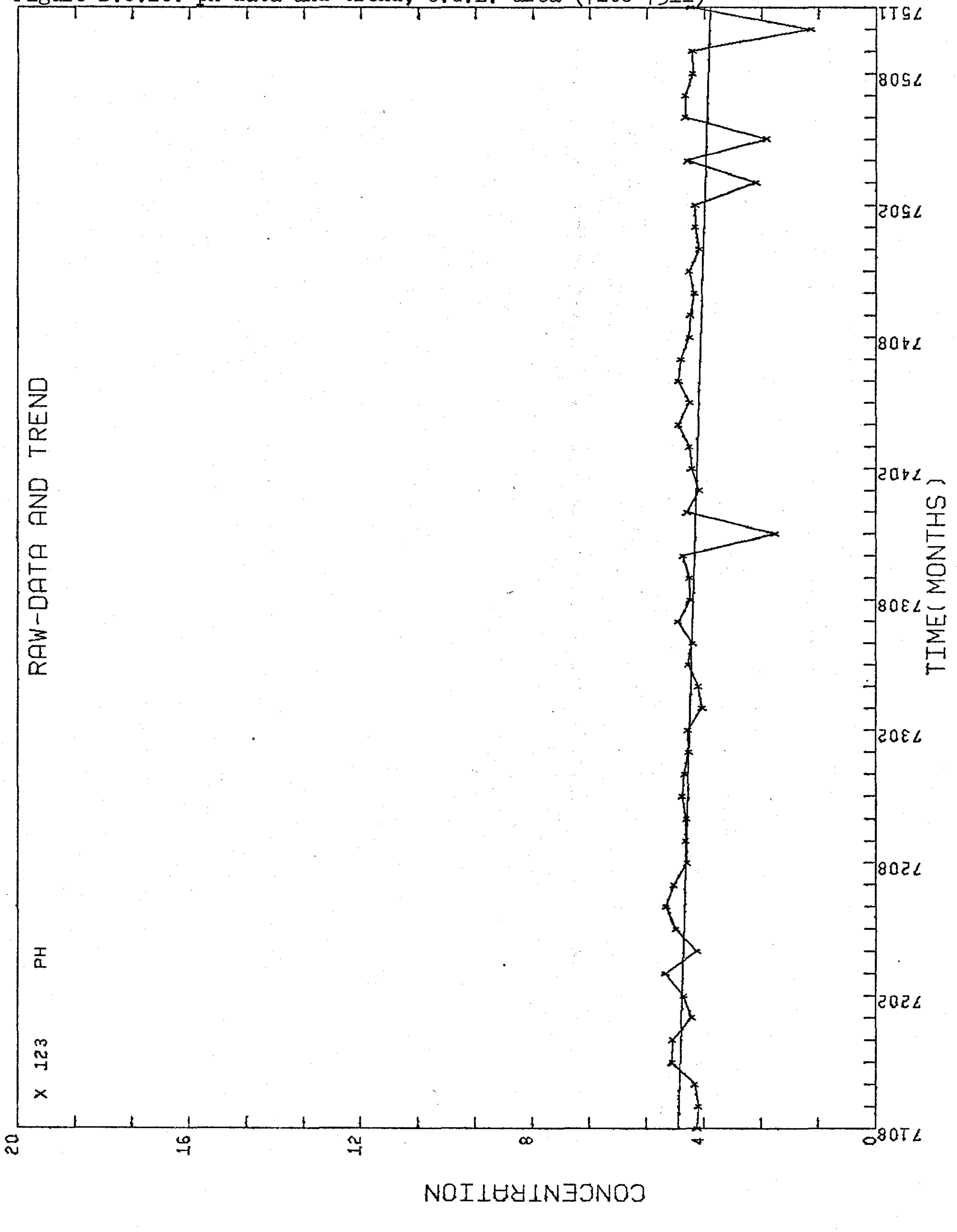


Figure B.6.11: SO_4 data and trend, U.G.L. area (7108-7511)

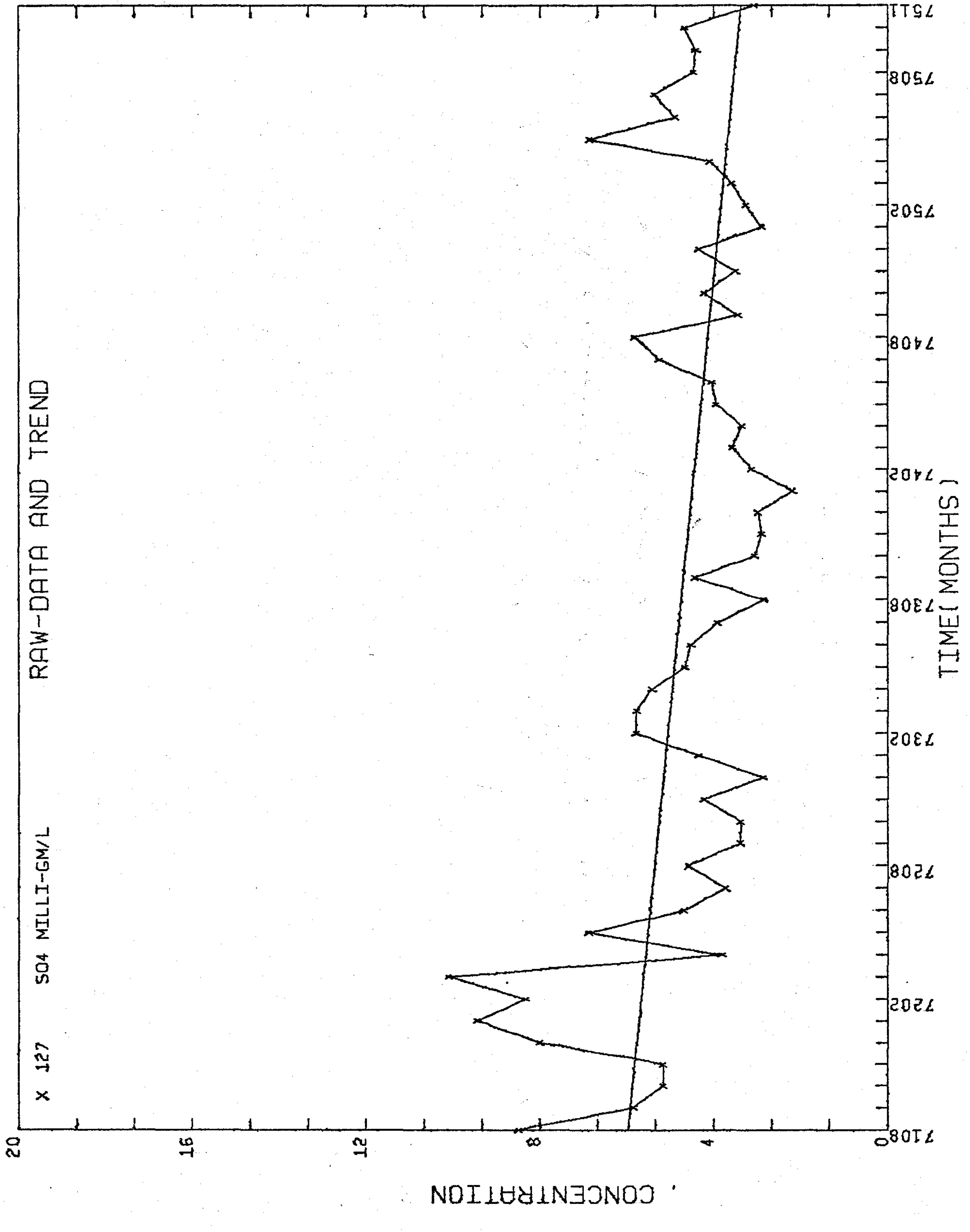


Figure B.6.12: T PART data and trend, U.G.L. area (7204-7511)

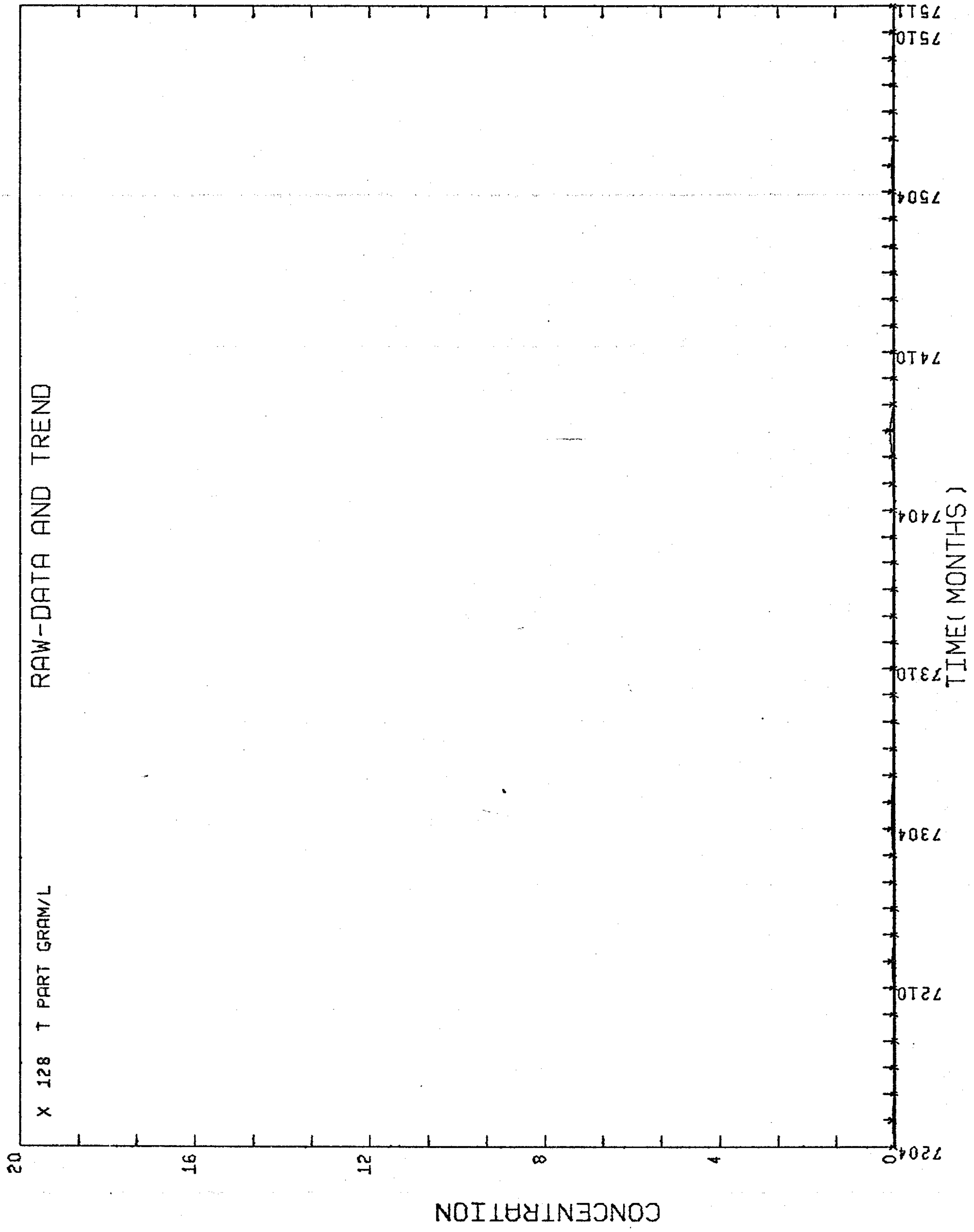
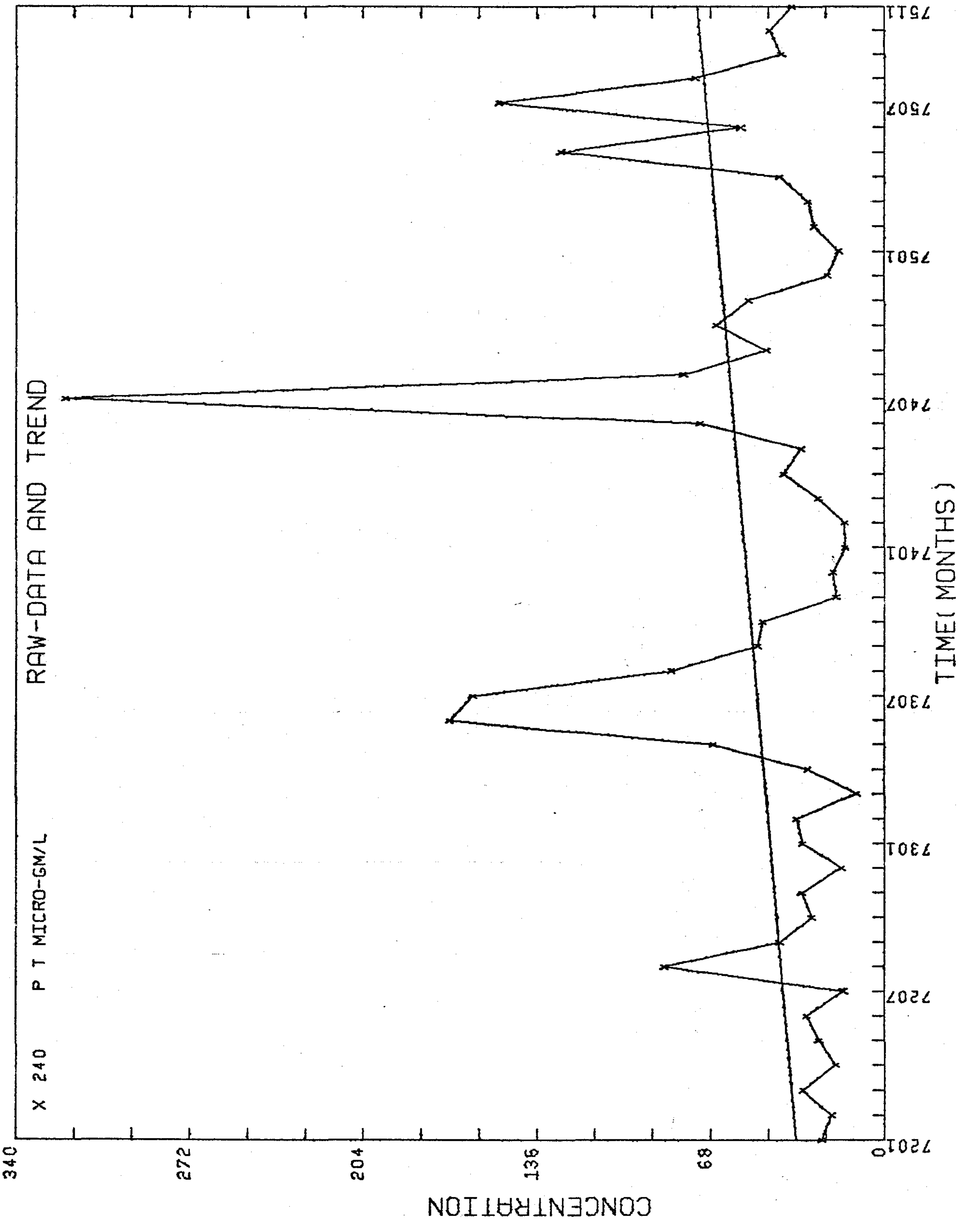


Figure B.6.13: P T data and trend, U.G.L area (7201-7511)



B.7 Spectra

The following Figures give the spectra of pairs of parameters which are given in Table B.7.1.

TABLE B.7.1: Pairs of spectral parameters considered

Parameters	Area	Time Interval
pH & SO_4	Station 12	7206-7511
pH & SO_4	Station 2	7206-7510
pH & SO_4	Station 3	7206-7511
pH & SO_4	Station 4	7203-7509
pH & SO_4	Sudbury	7108-7511
pH & SO_4	U.G.L.	7108-7511
pH & SP CON	Sudbury	7108-7511
pH & SP CON	U.G.L.	7108-7511
Fe T & Zn T	Station 12	7206-7511
Fe T & Zn T	Station 2	7206-7510
Fe T & Zn T	Station 3	7206-7511
Fe T & Zn T	Station 4	7204-7509
Fe T & Pb T	Sudbury	7204-7511
Fe T & Pb T	U.G.L.	7201-7511
Zn T & Pb T	Sudbury	7204-7511
pH & P T	Station 12	7210-7511
pH & P T	Station 2	7211-7510
pH & P T	U.G.L.	7201-7511

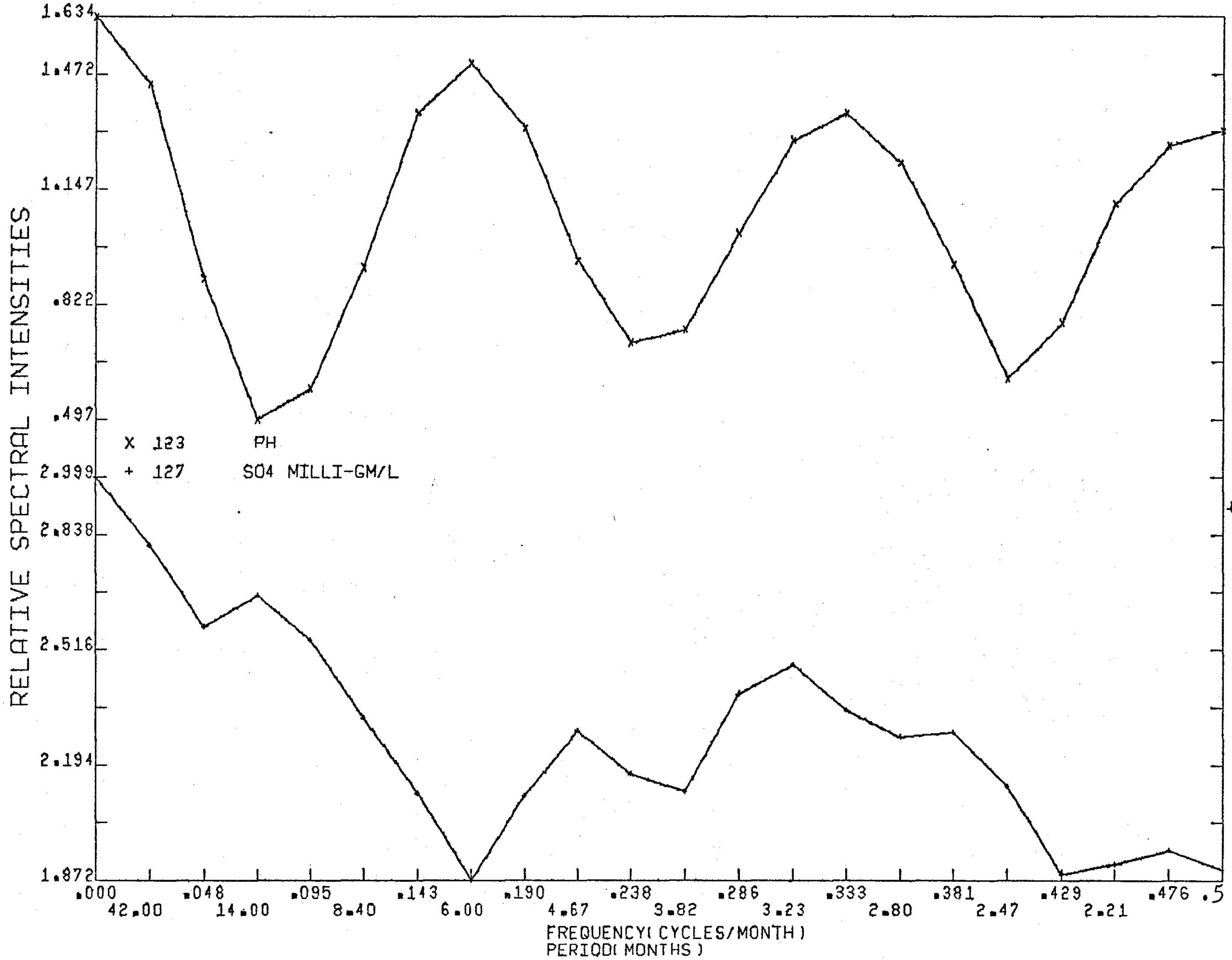


Figure B.7.1: Power spectrum for pH & SO_4, station 12 (7206-7511)

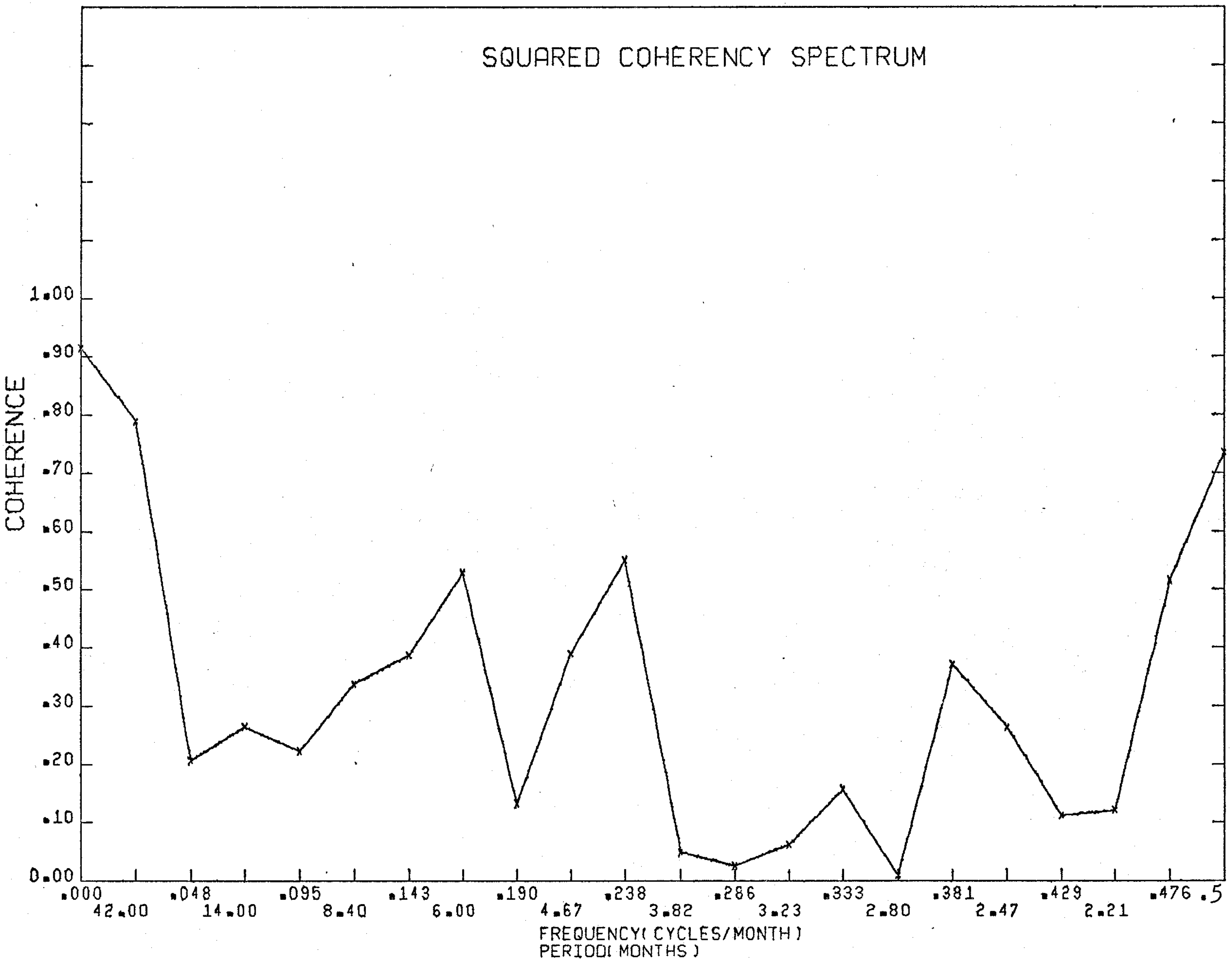


Figure B.7.2: Coherency spectrum for pH & SO_4, station 12 (7206-7511)

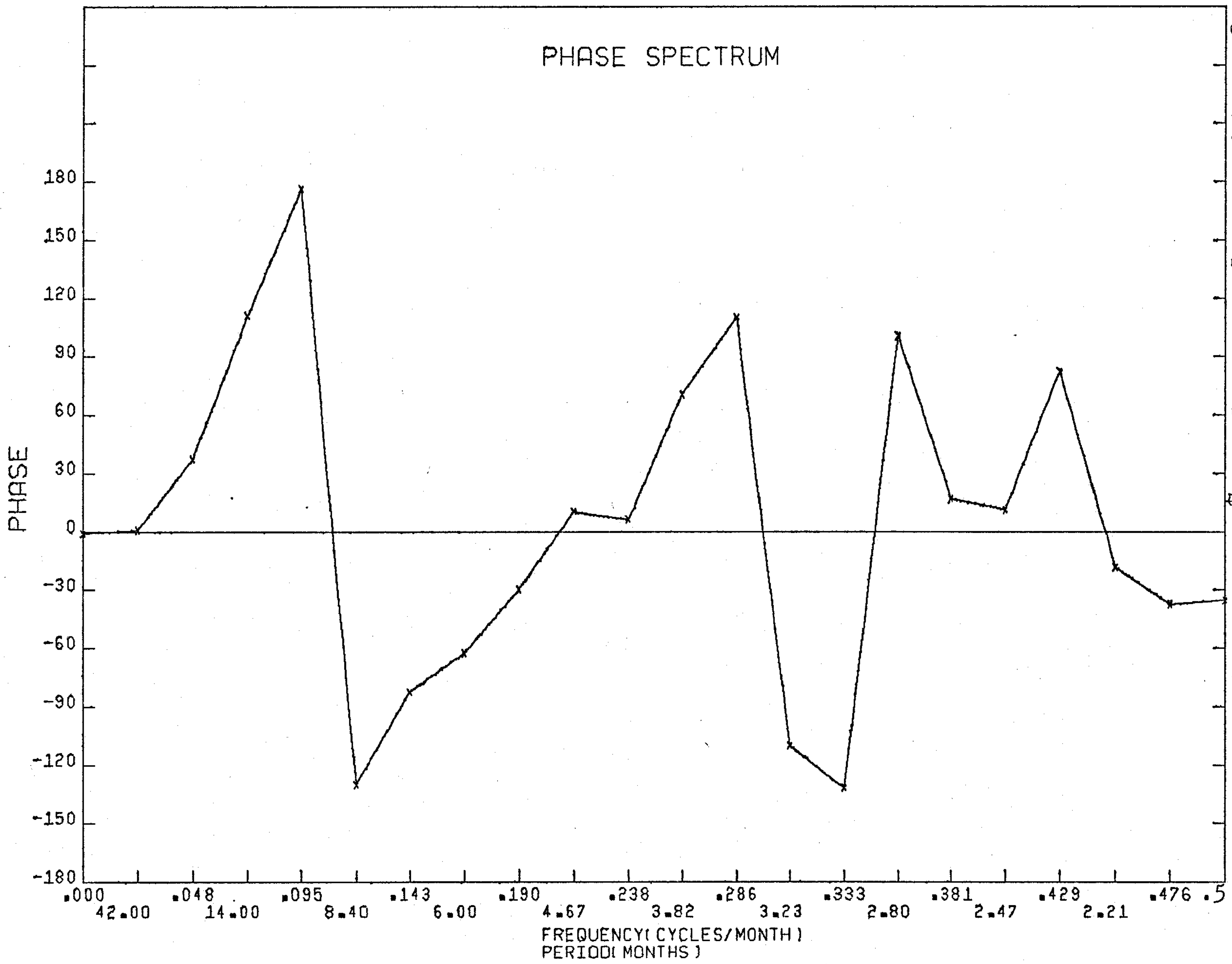


Figure B.7.3: Phase spectrum for pH & SO_4, station 12 (7206-7511)

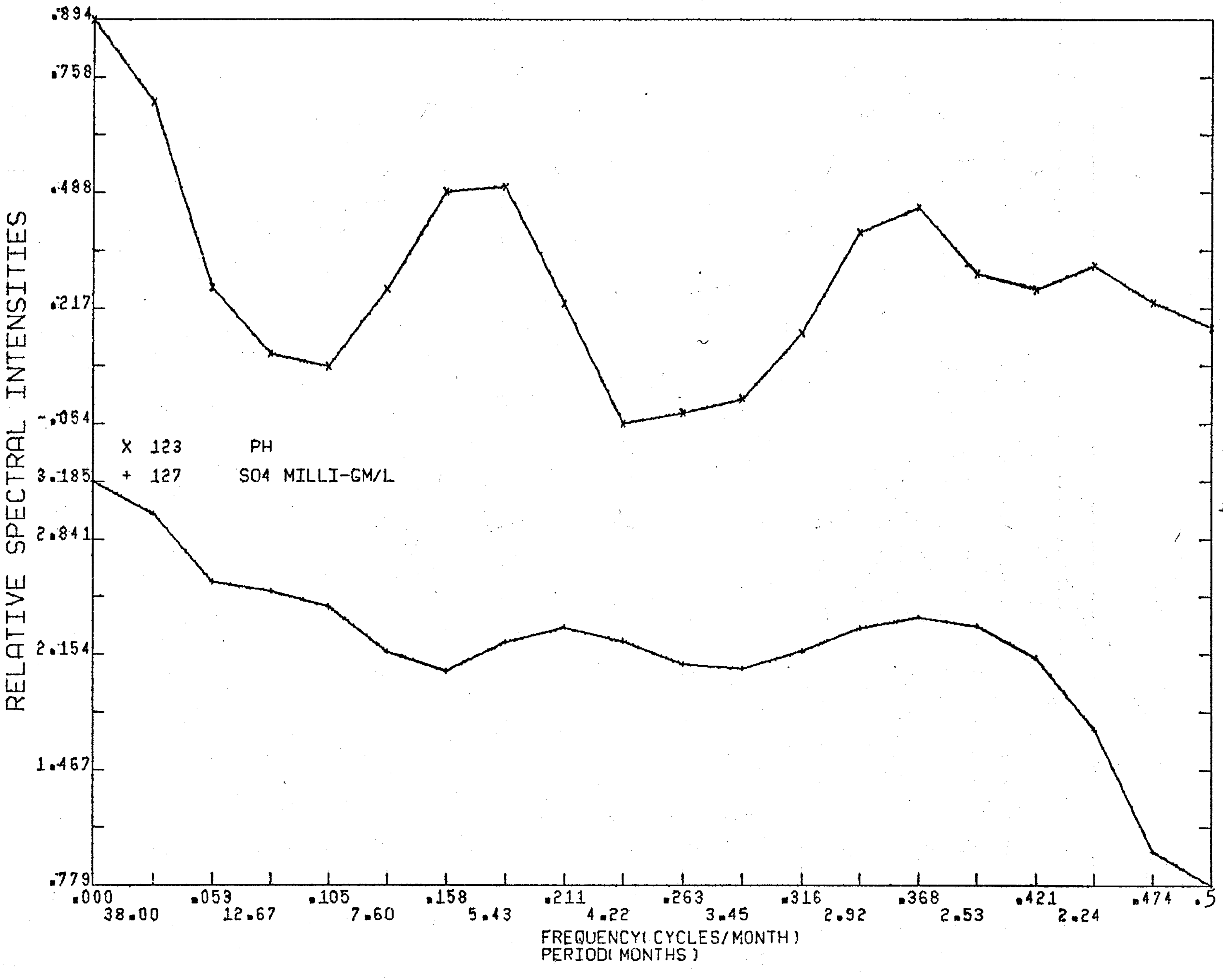


Figure B.7.4: Power spectrum for pH & SO_4, station 2 (7206-7510)

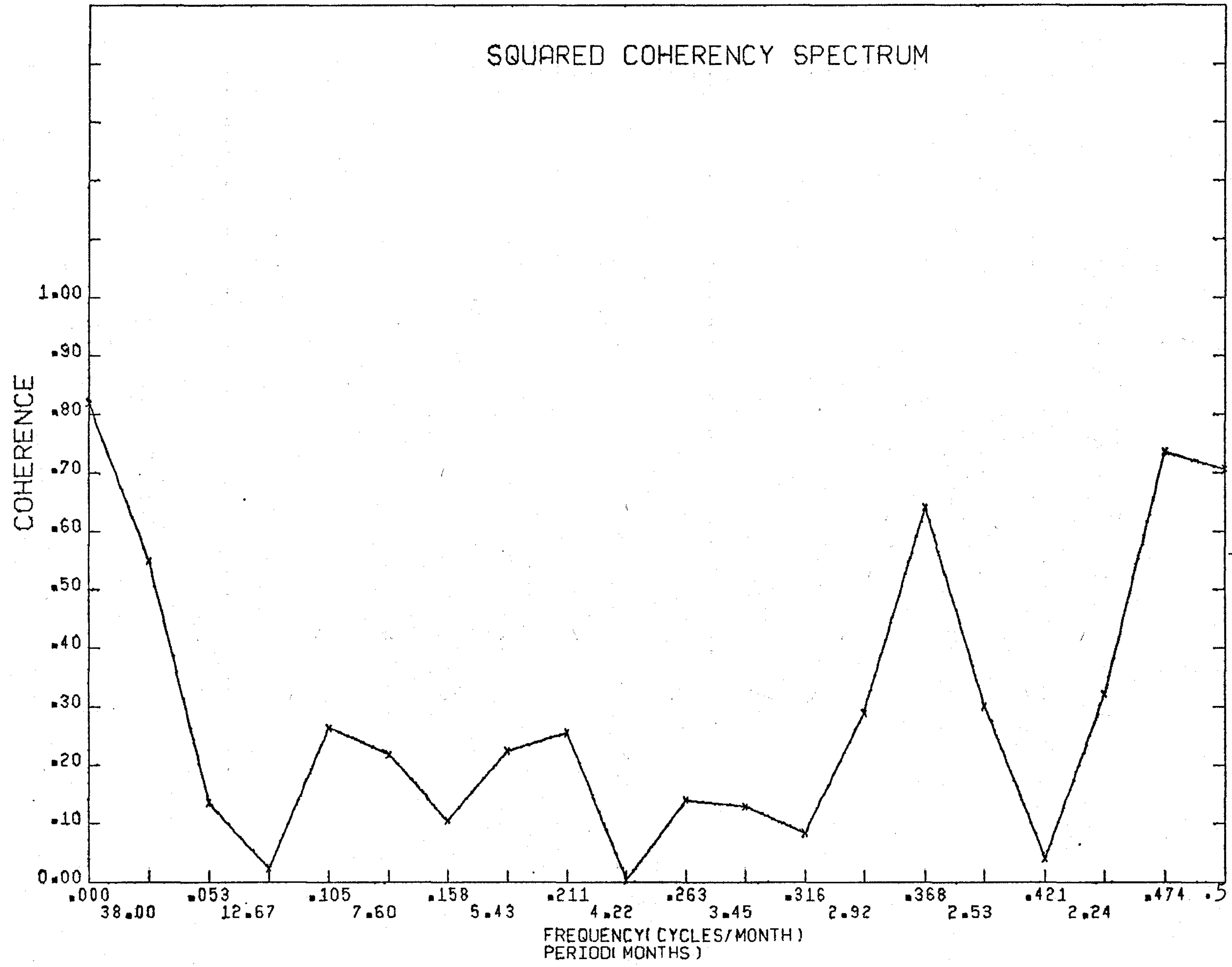


Figure B.7.5: Coherency spectrum for pH & SO_4, station 2 (7206-7510)

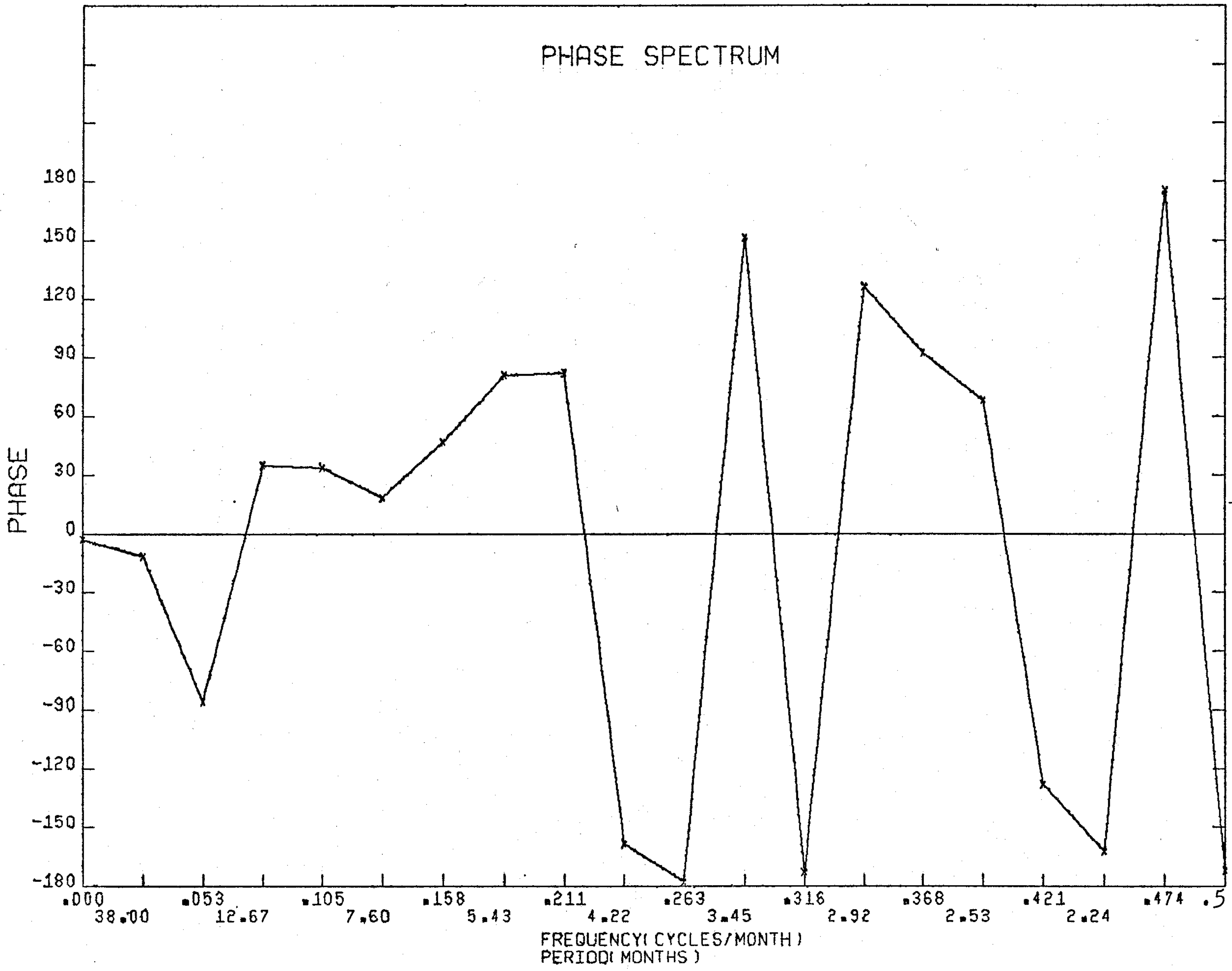


Figure B.7.6: Phase spectrum for pH & SO$_4$, station 2 (7206-7510)

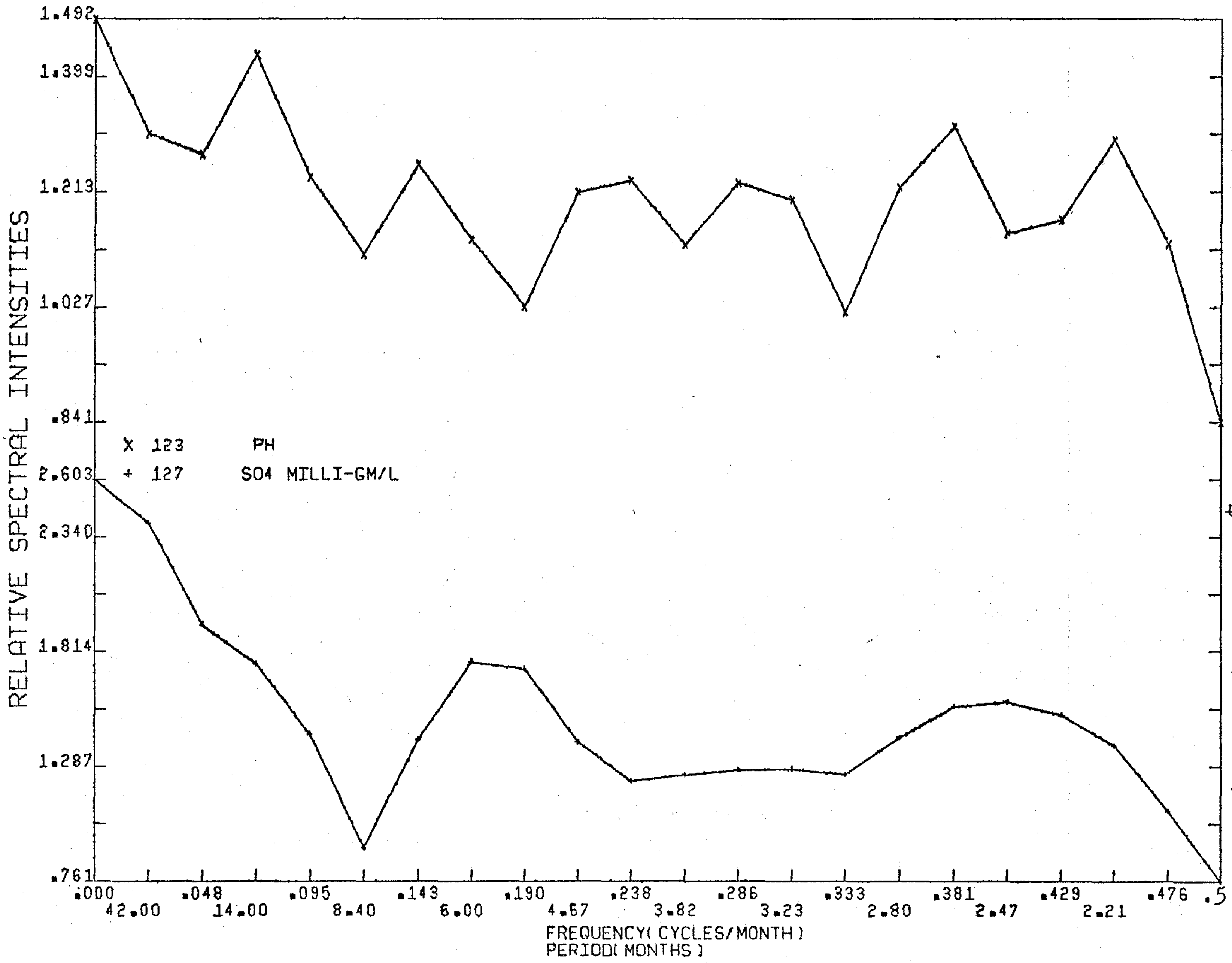


Figure B.7.7: Power spectrum for pH & SO_4, station 3 (7206-7511)

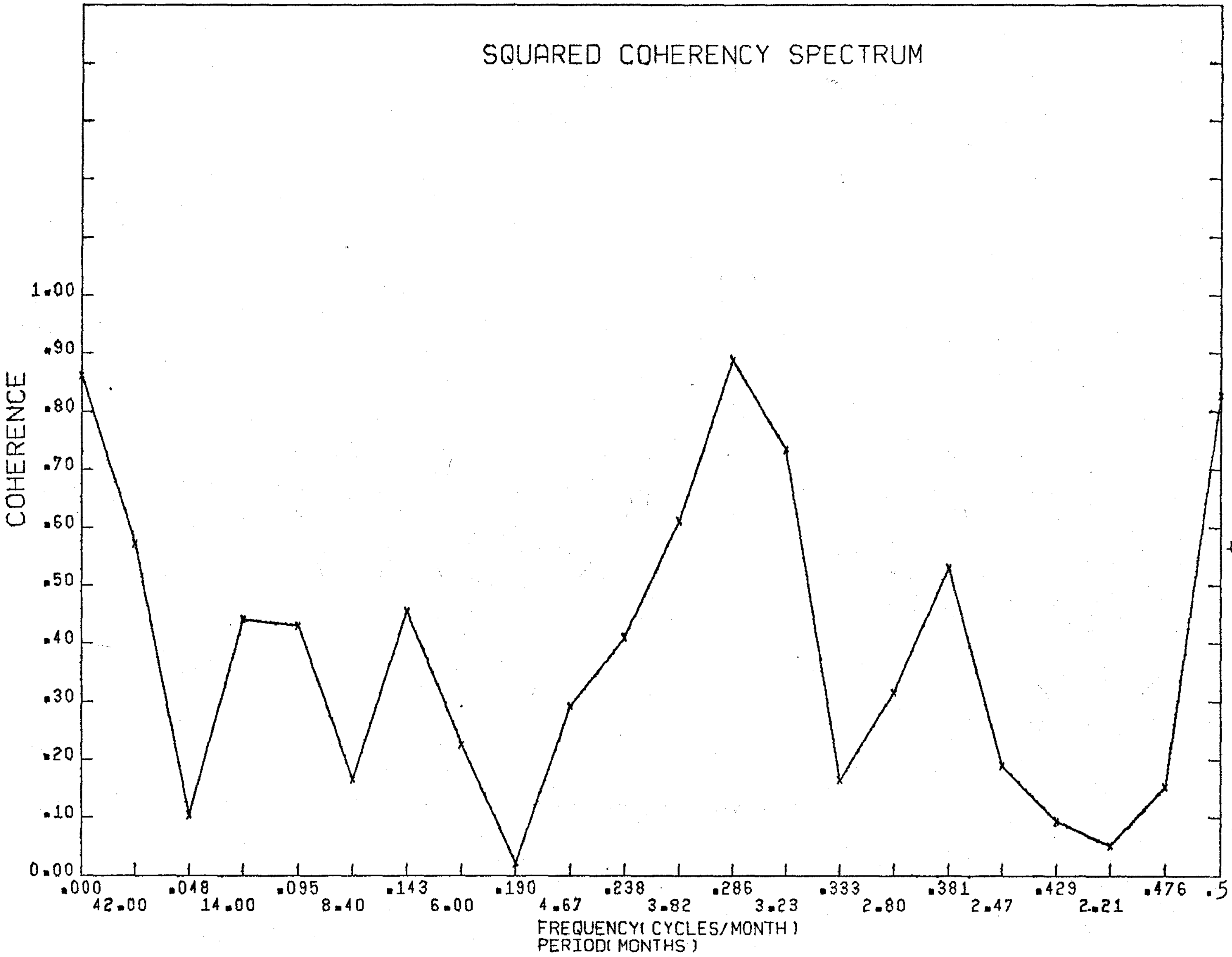


Figure B.7.8: Coherency spectrum for pH & SO_4, station 3 (7206-7511)

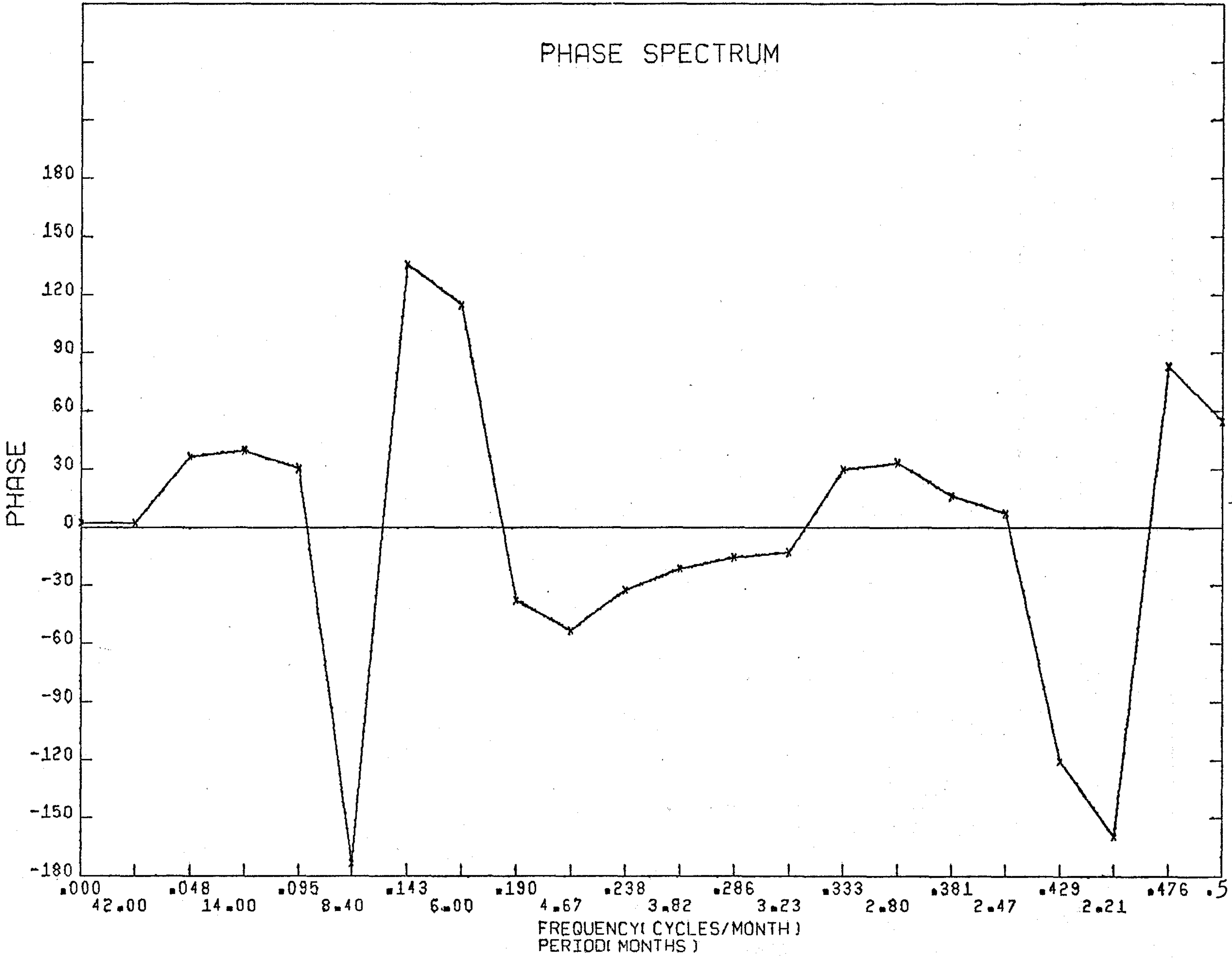


Figure B.7.9: Phase spectrum for pH & SO_4, station 3 (7206-7511)

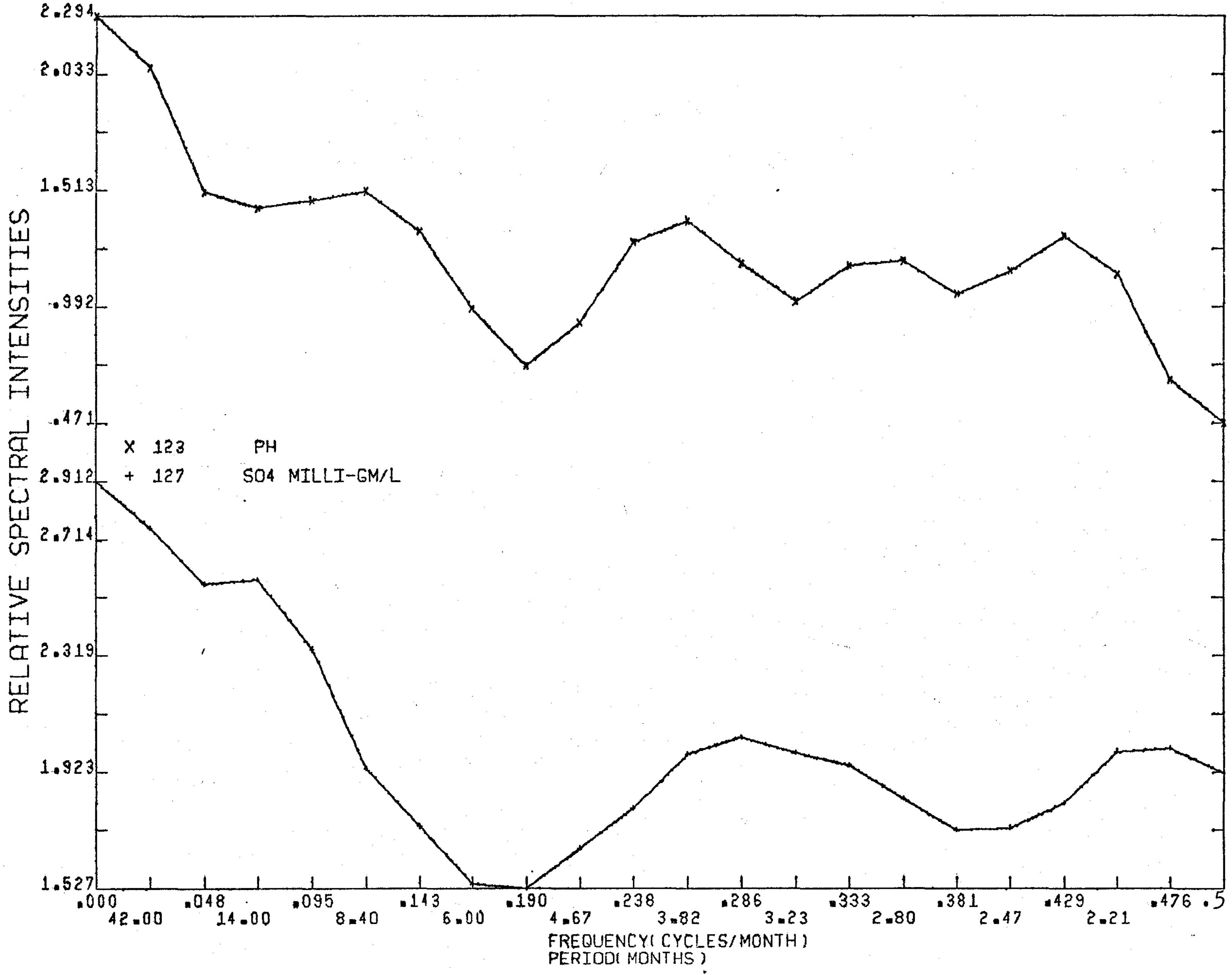


Figure B.7.10: Power spectrum for pH & SO_4, station 4 (7203-7509)

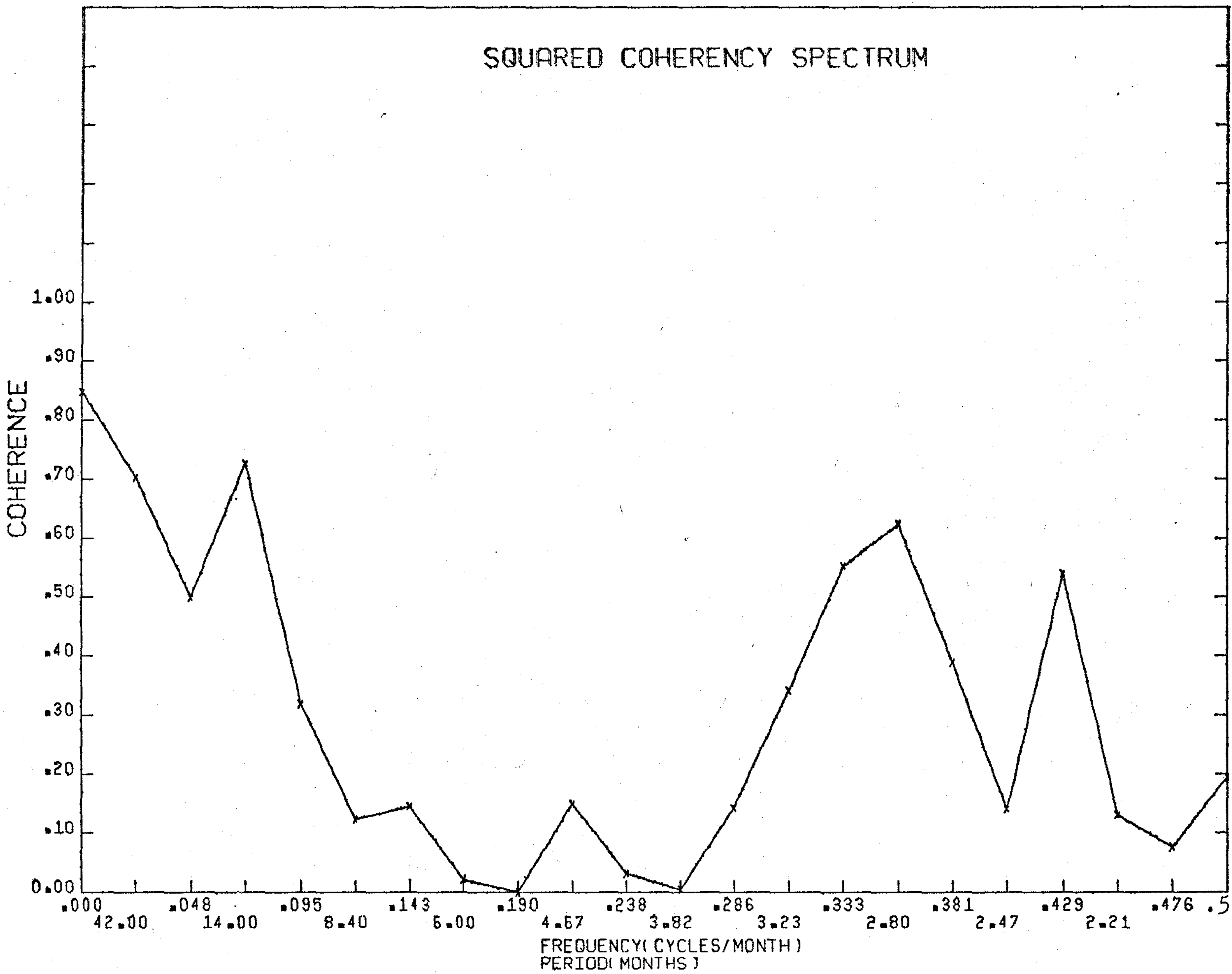


Figure B.7.11: Coherency spectrum for pH & SO_4, station 4 (7203-7509)

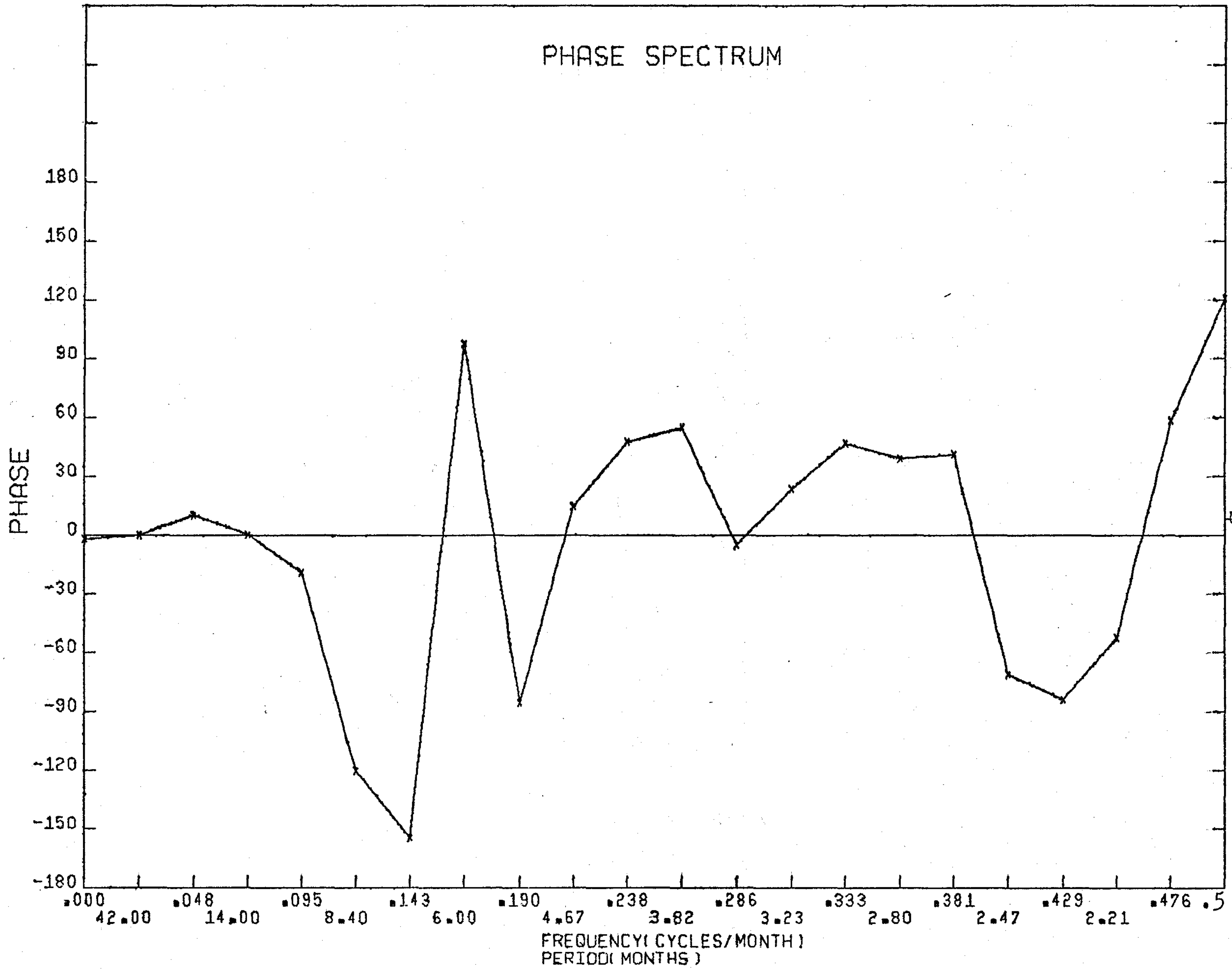


Figure B.7.12: Phase spectrum for pH & SO_4, station 4 (7203-7509)

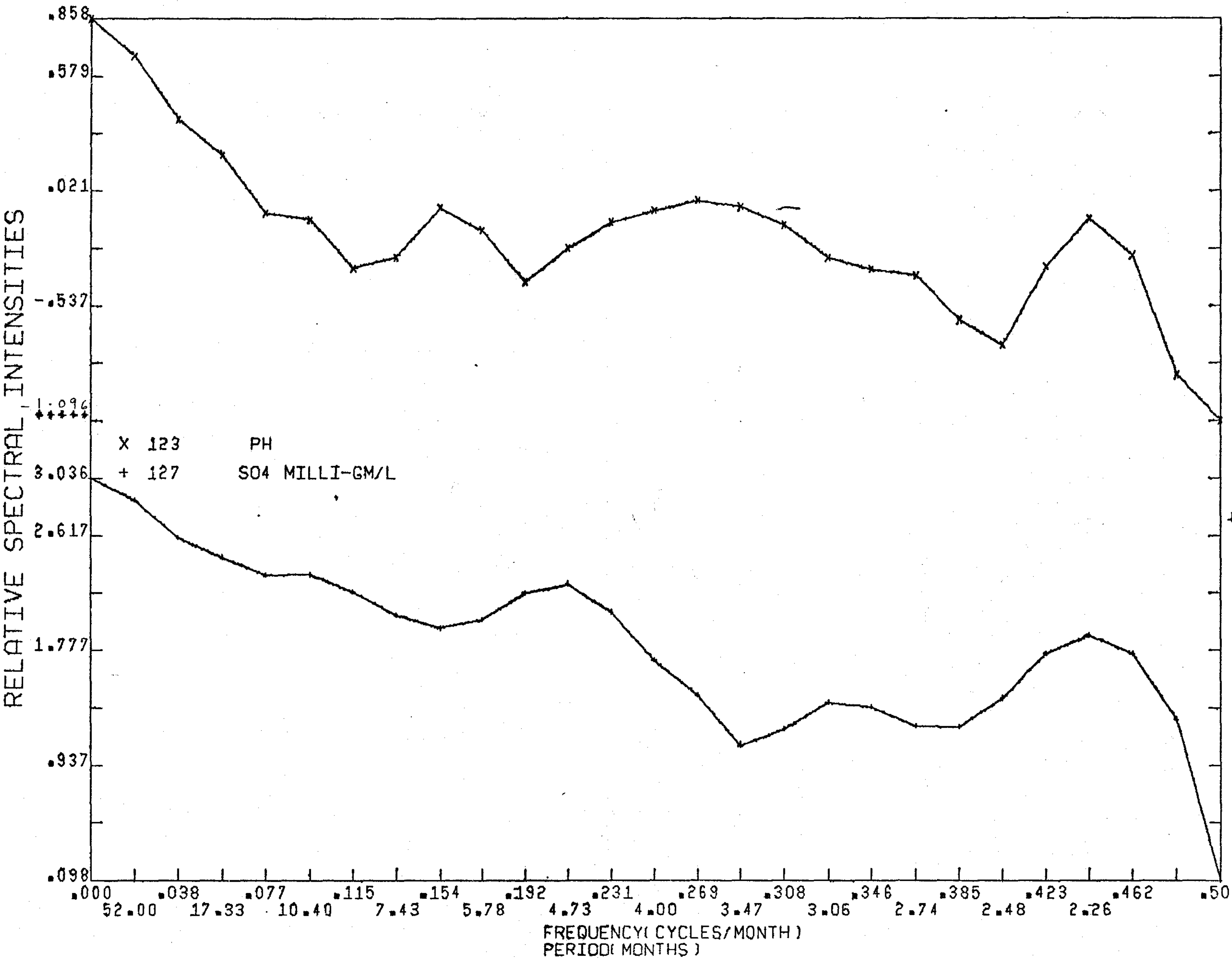


Figure B.7.13: Power spectrum for pH & SO_4, Sudbury area (7108-7511)

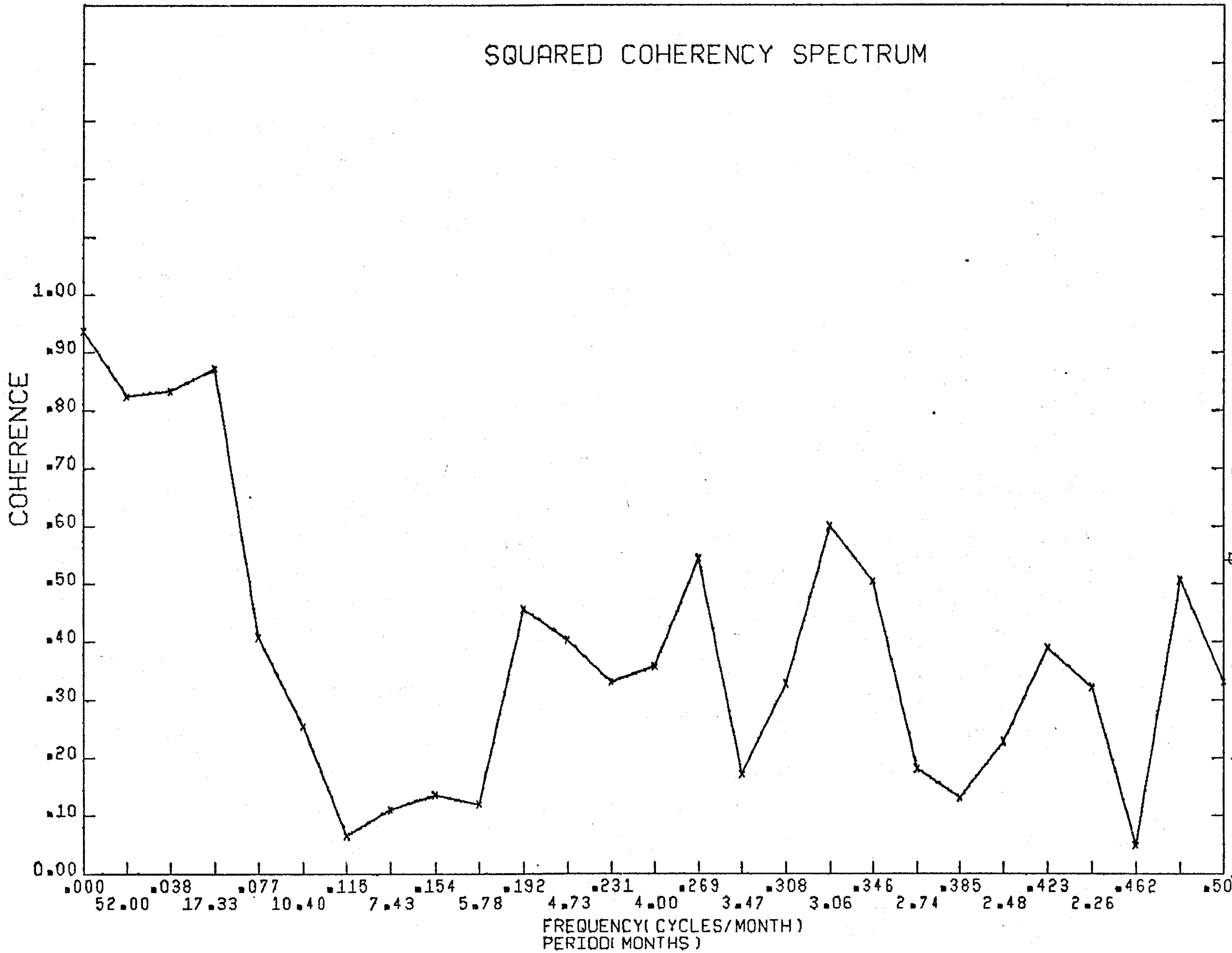


Figure B.7.14: Coherency spectrum for pH & SO_4, Sudbury area (7108-7511)

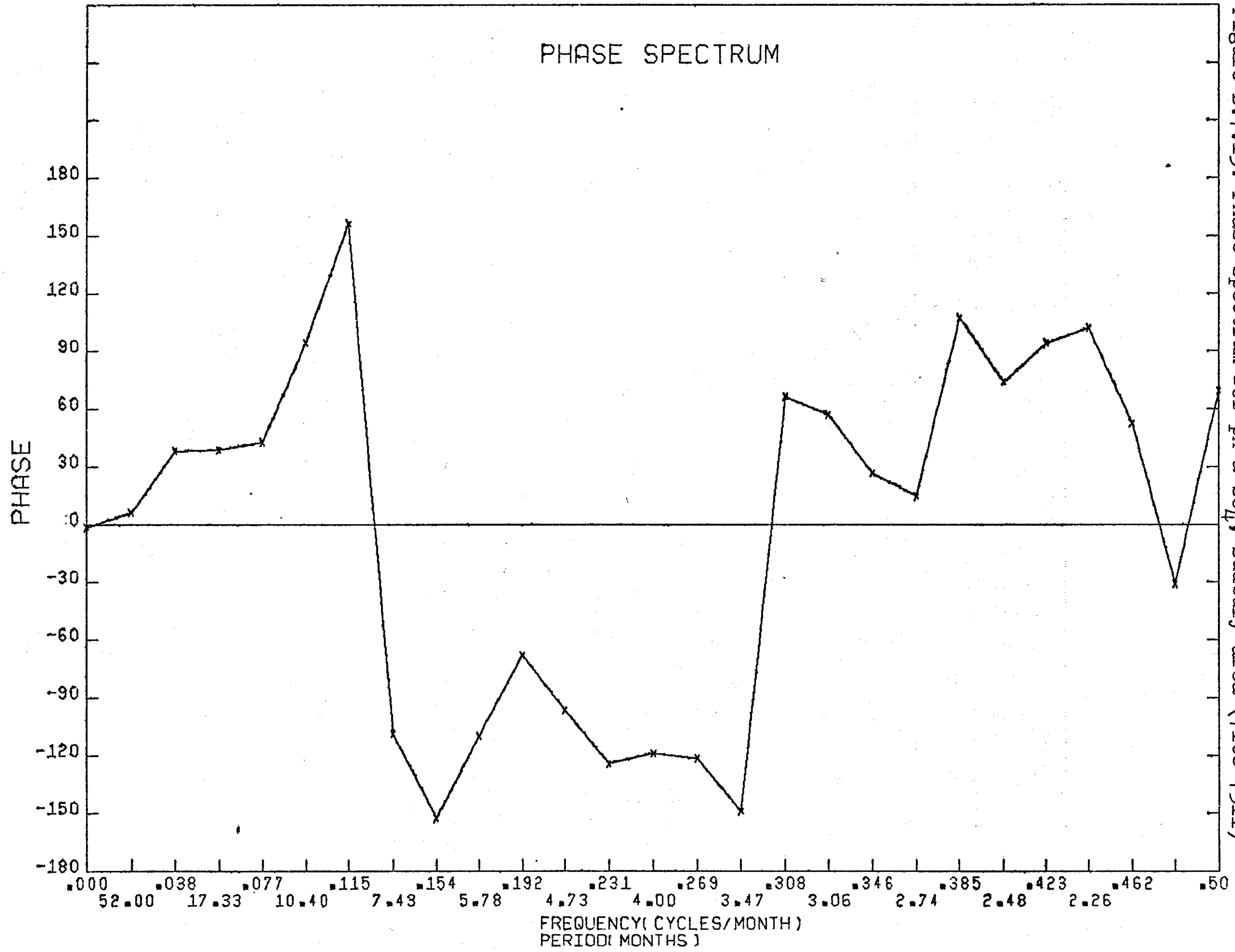


Figure B.7.15: Phase spectrum for pH & SO_4, Sudbury area (7108-7511)

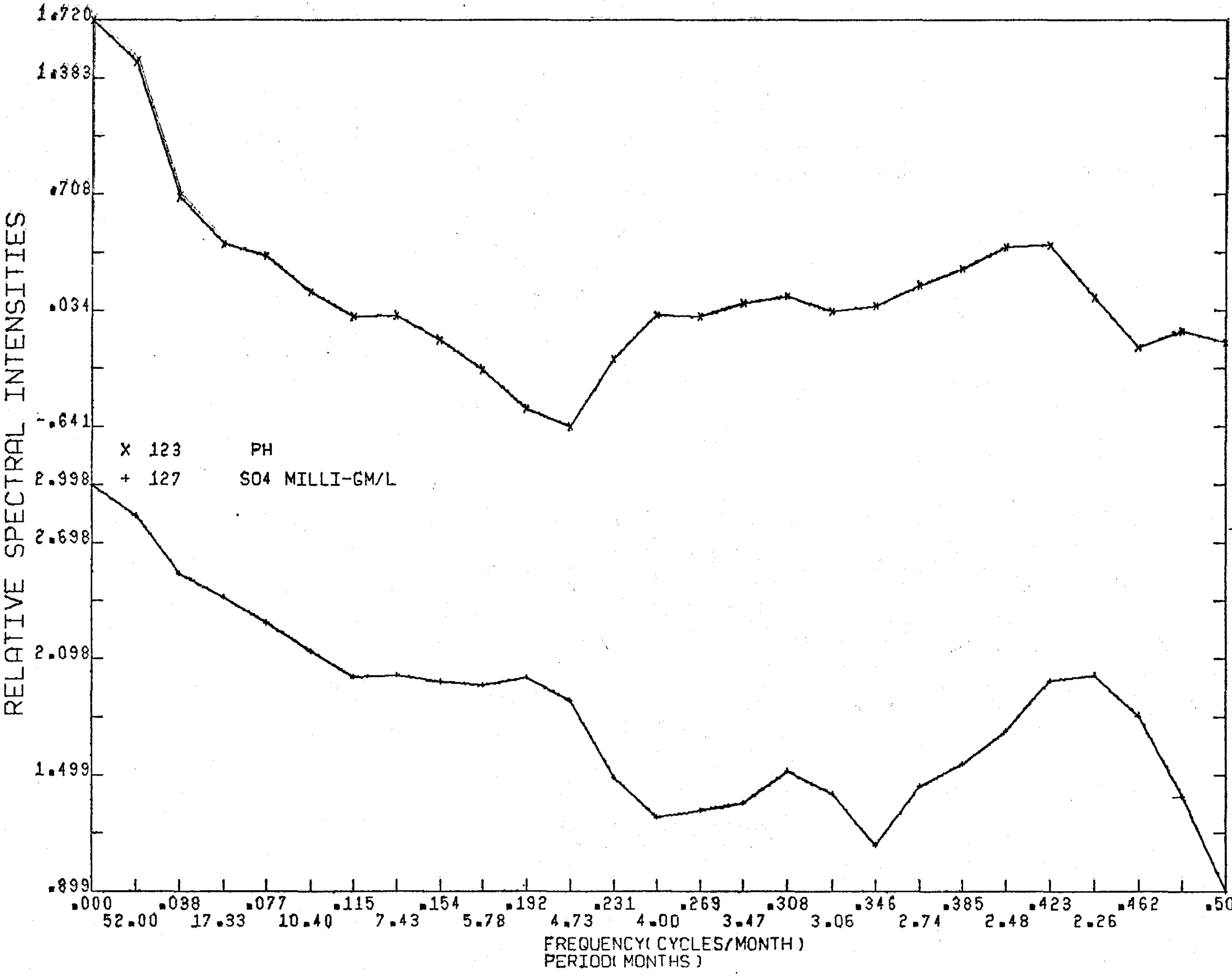


Figure B.7.16: Power spectrum for pH & SO_4, U.G.L. area (7208-7511)

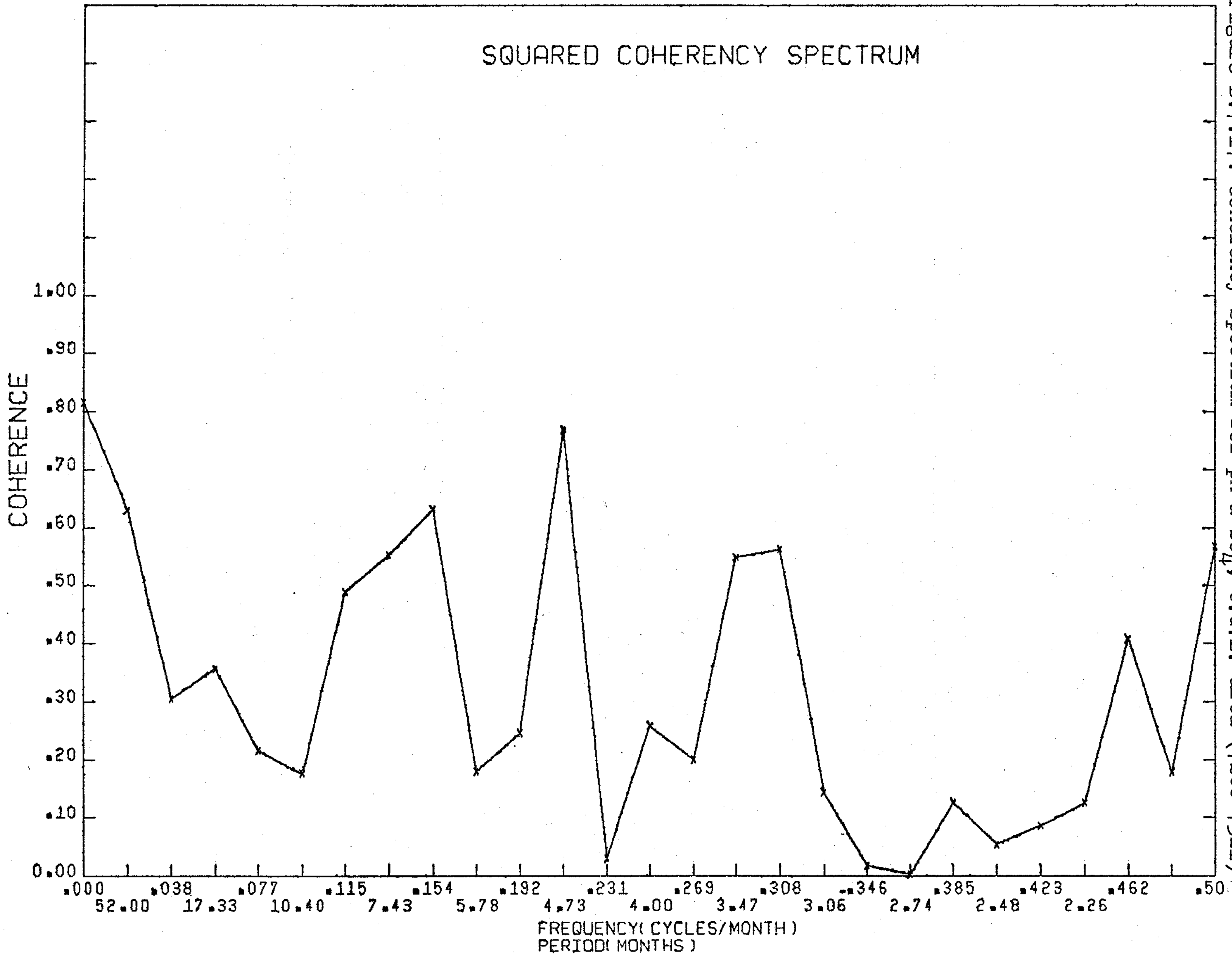


Figure B.7.17: Coherency spectrum for pH & SO_4, U.G.L. area (7208-7511)

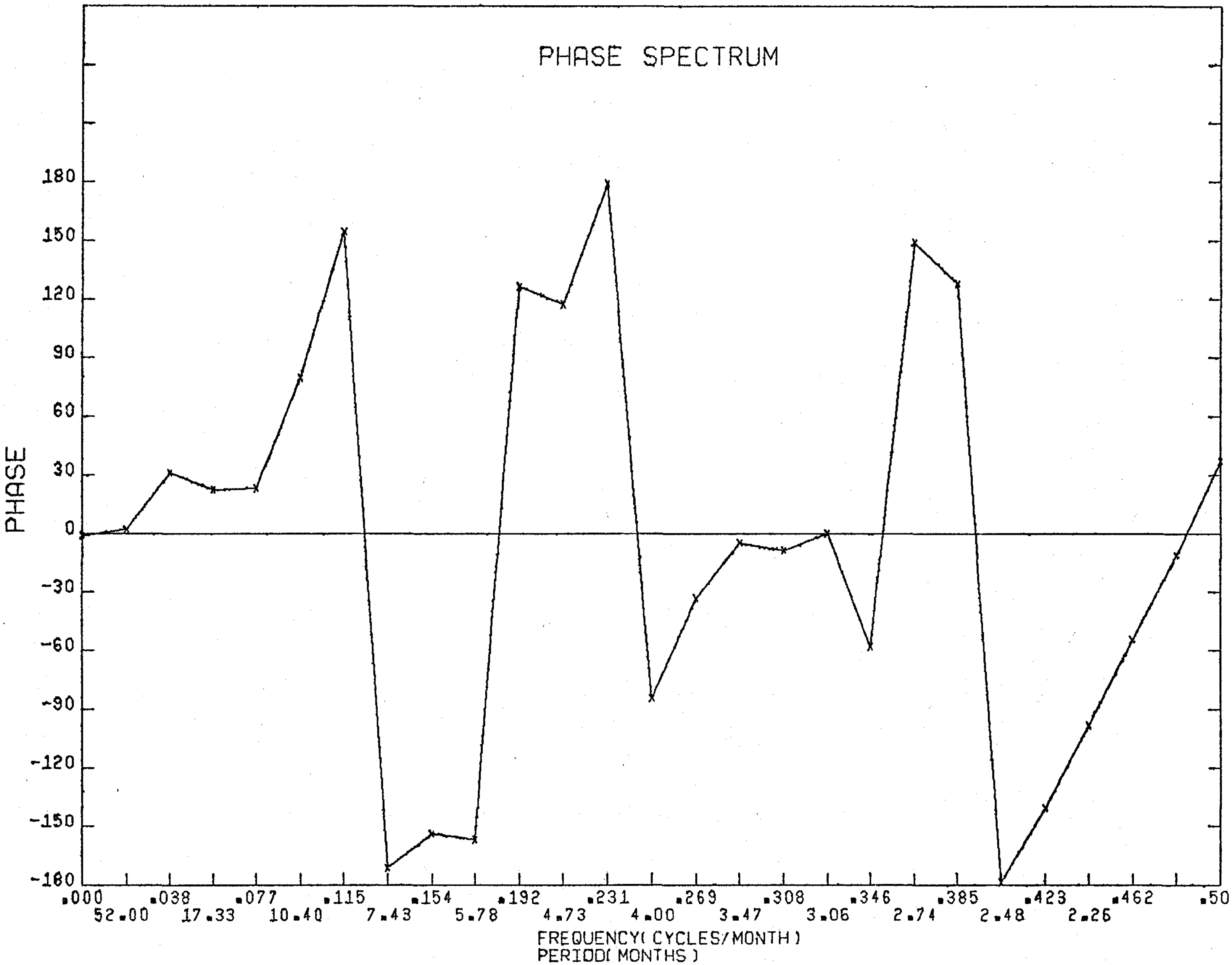


Figure B.7.18: Phase spectrum for pH & SO_4, U.G.L. area (7208-7511)

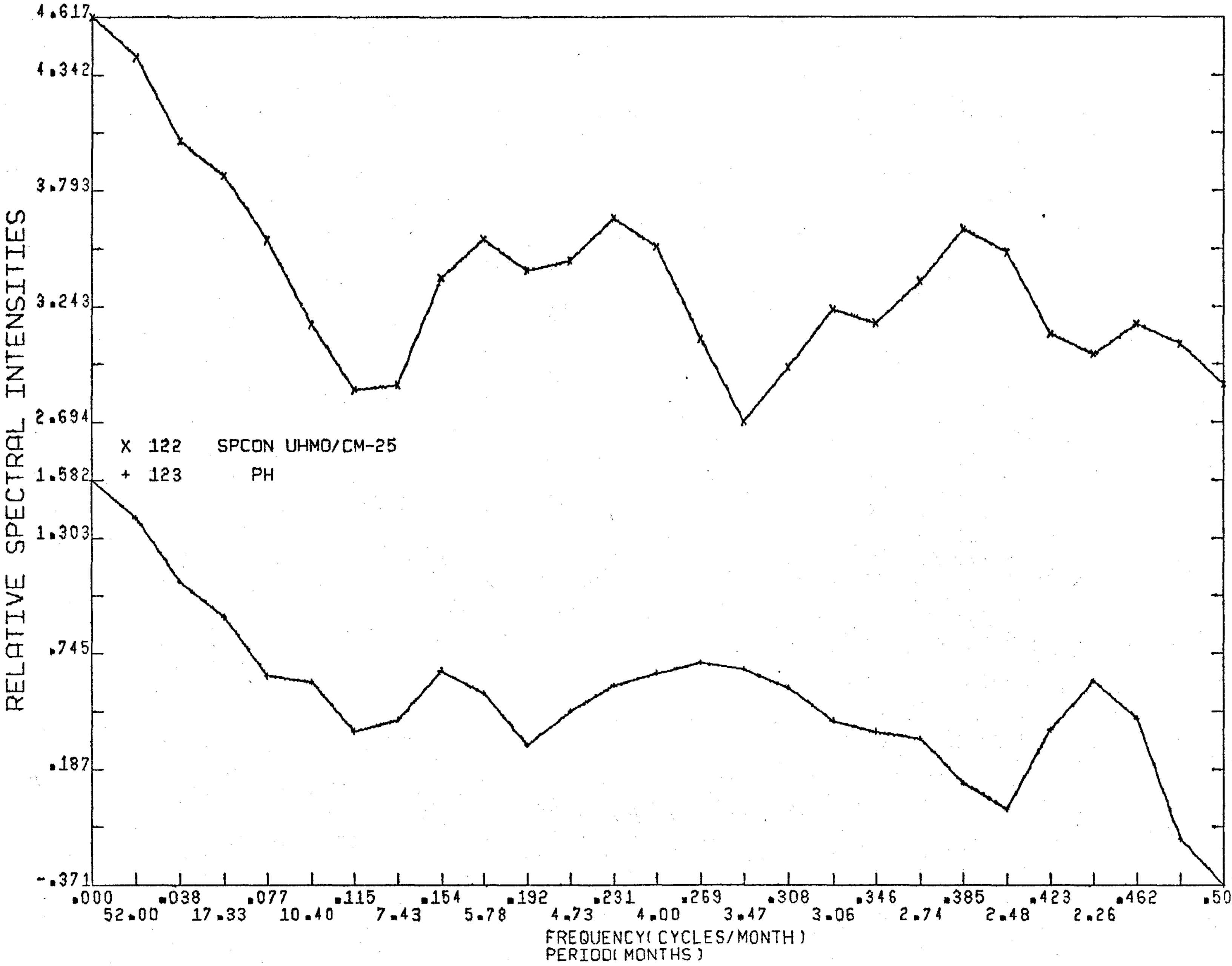


Figure B.7.19: Power spectrum for pH & SP CON, Sudbury area (7108-7511)

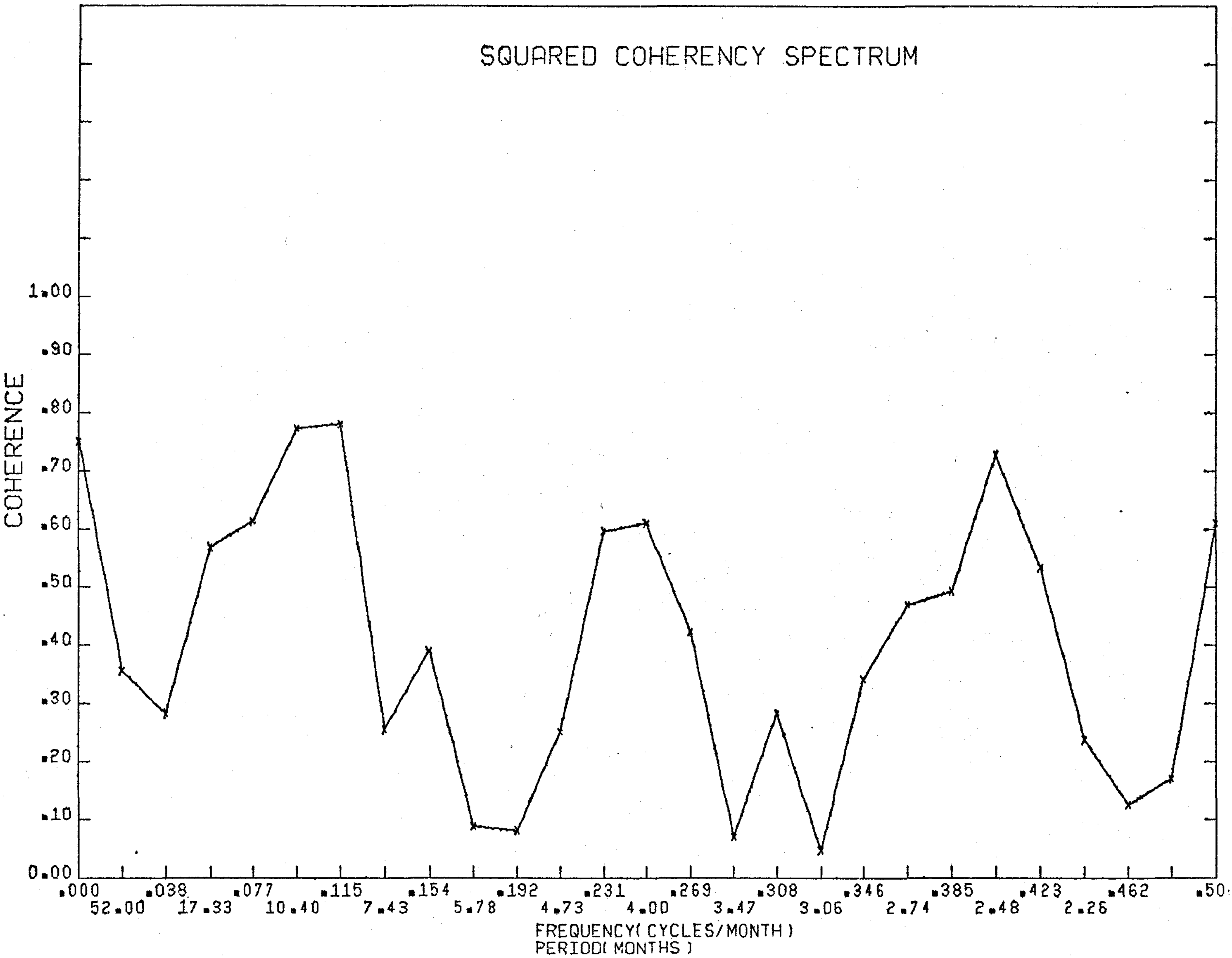


Figure B.7.20: Coherency spectrum for pH & SP CON, Sudbury area (7108-7511)

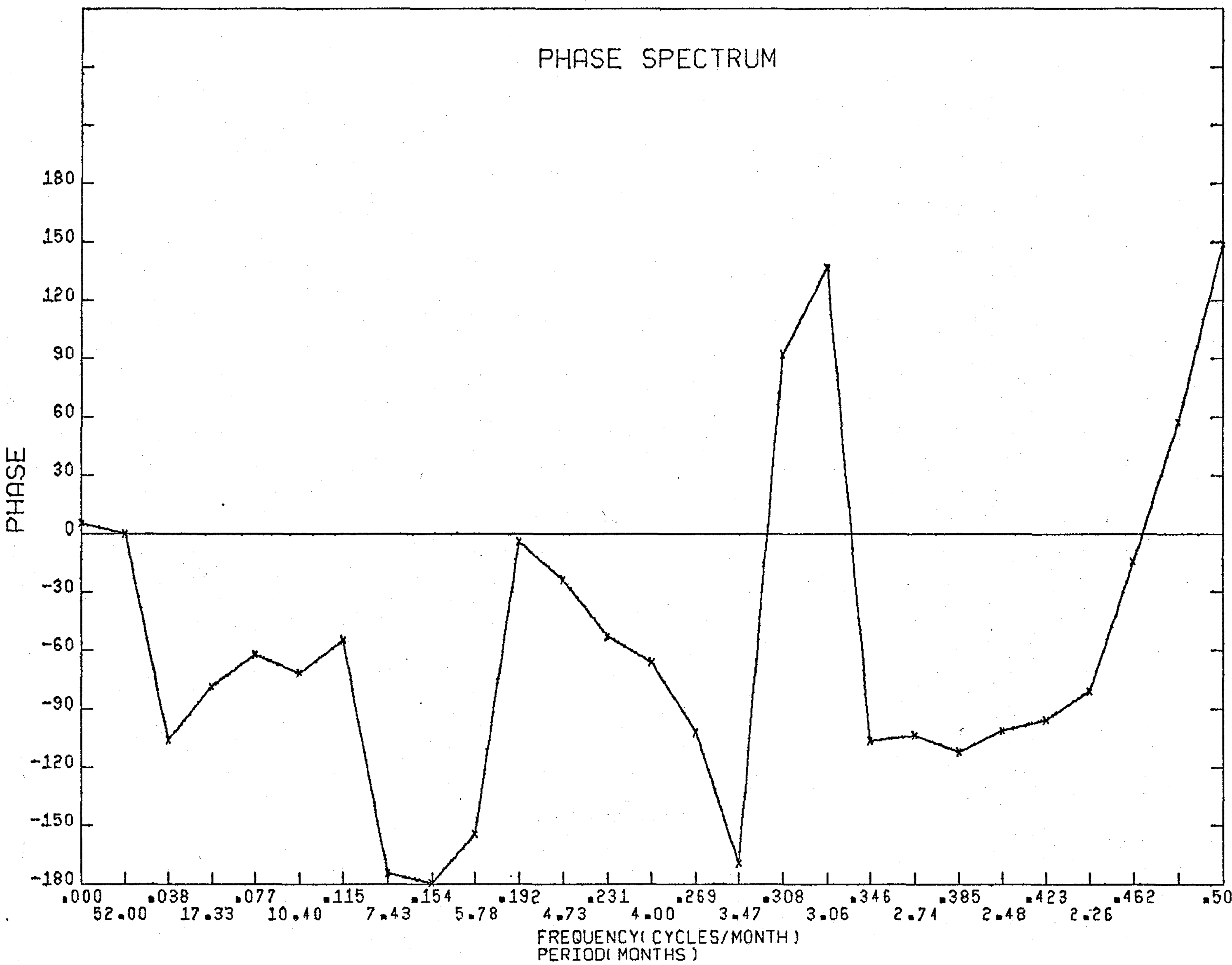


Figure B.7.21: Phase spectrum for pH & SP CON, Sudbury area (7108-7511)

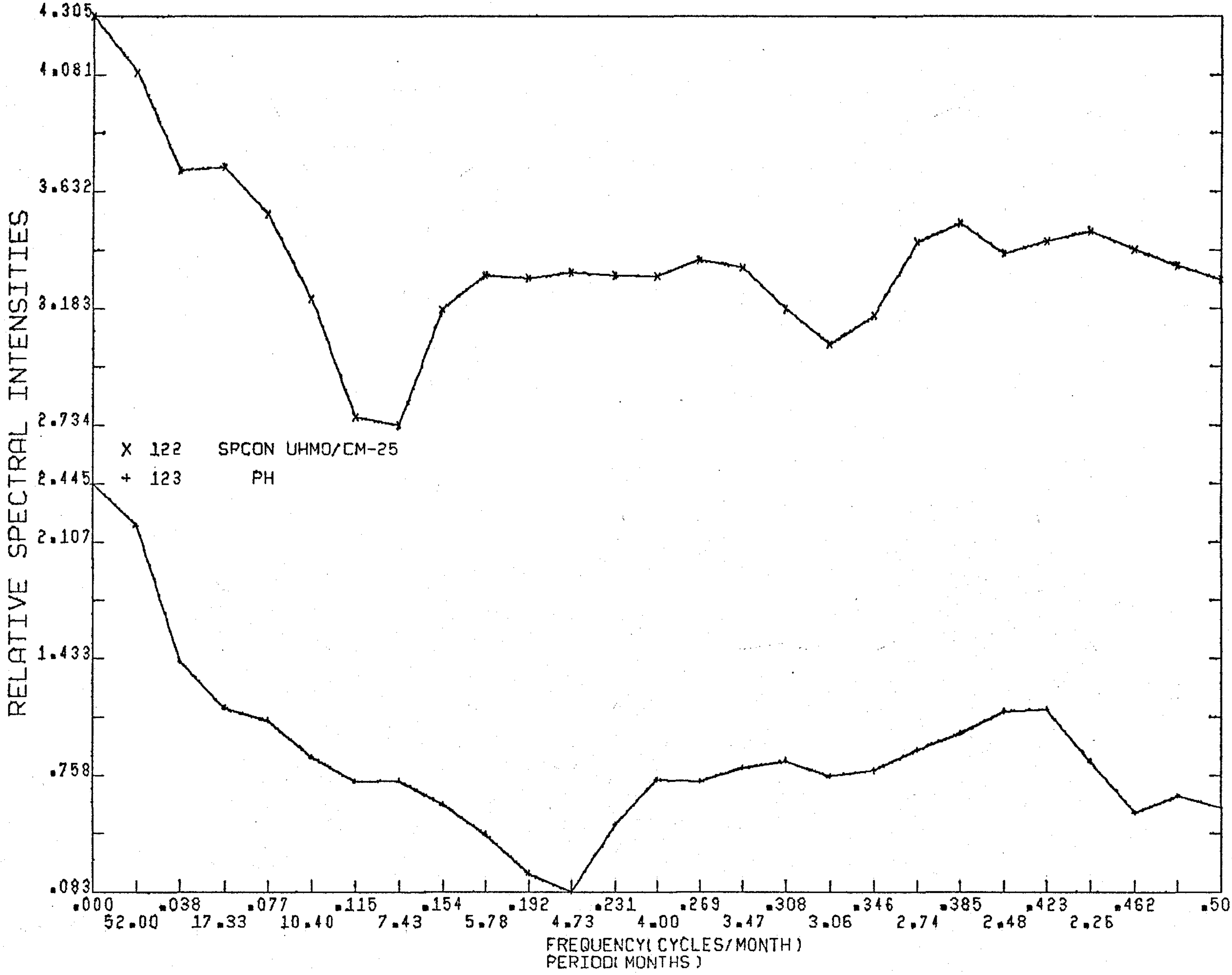


Figure B.7.22: Power spectrum for pH & SP CON, U.G.I. area (7108-7511)

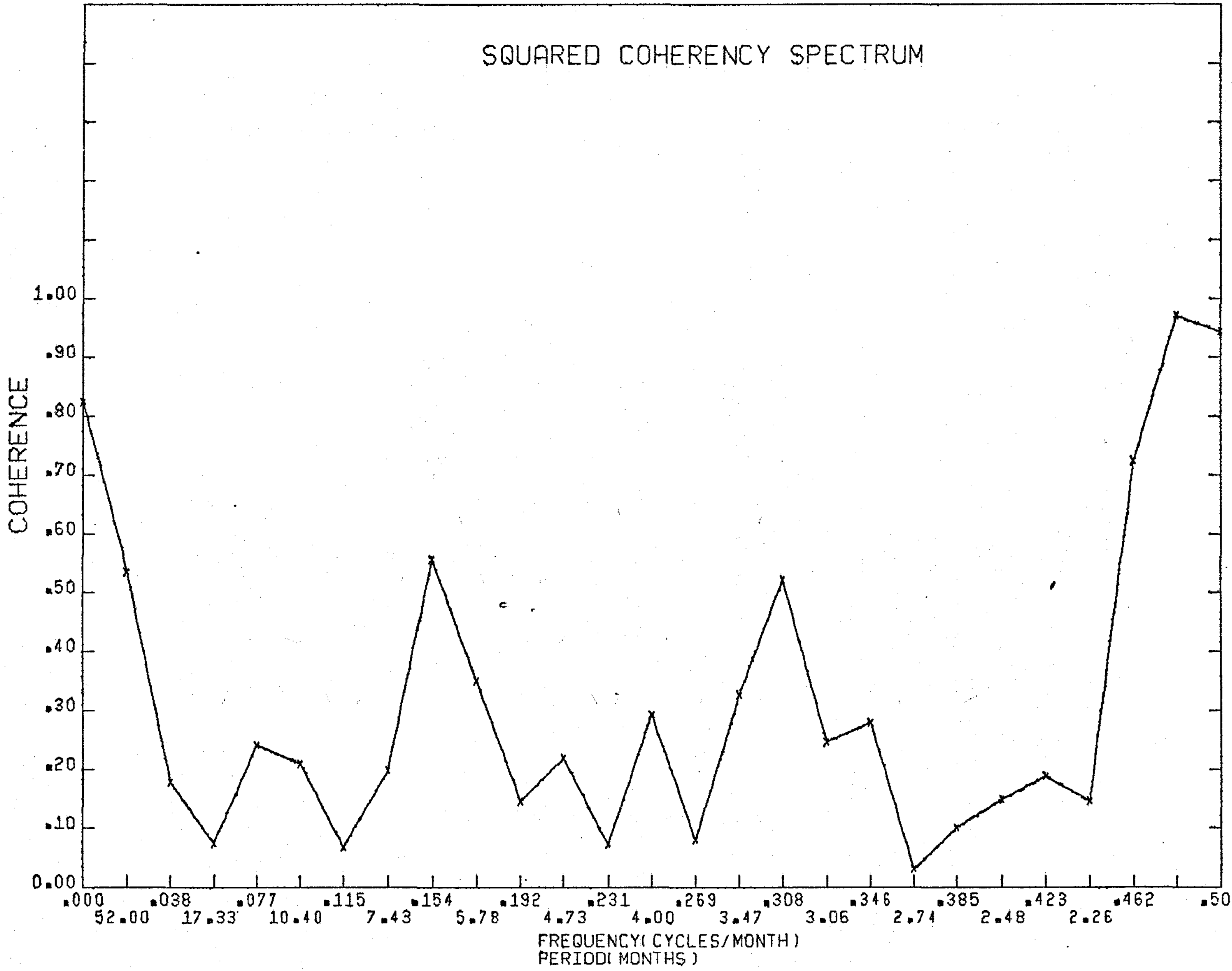


Figure B.7.23: Coherency spectrum for pH & SP CON, U.G.L. area (7108-7511)

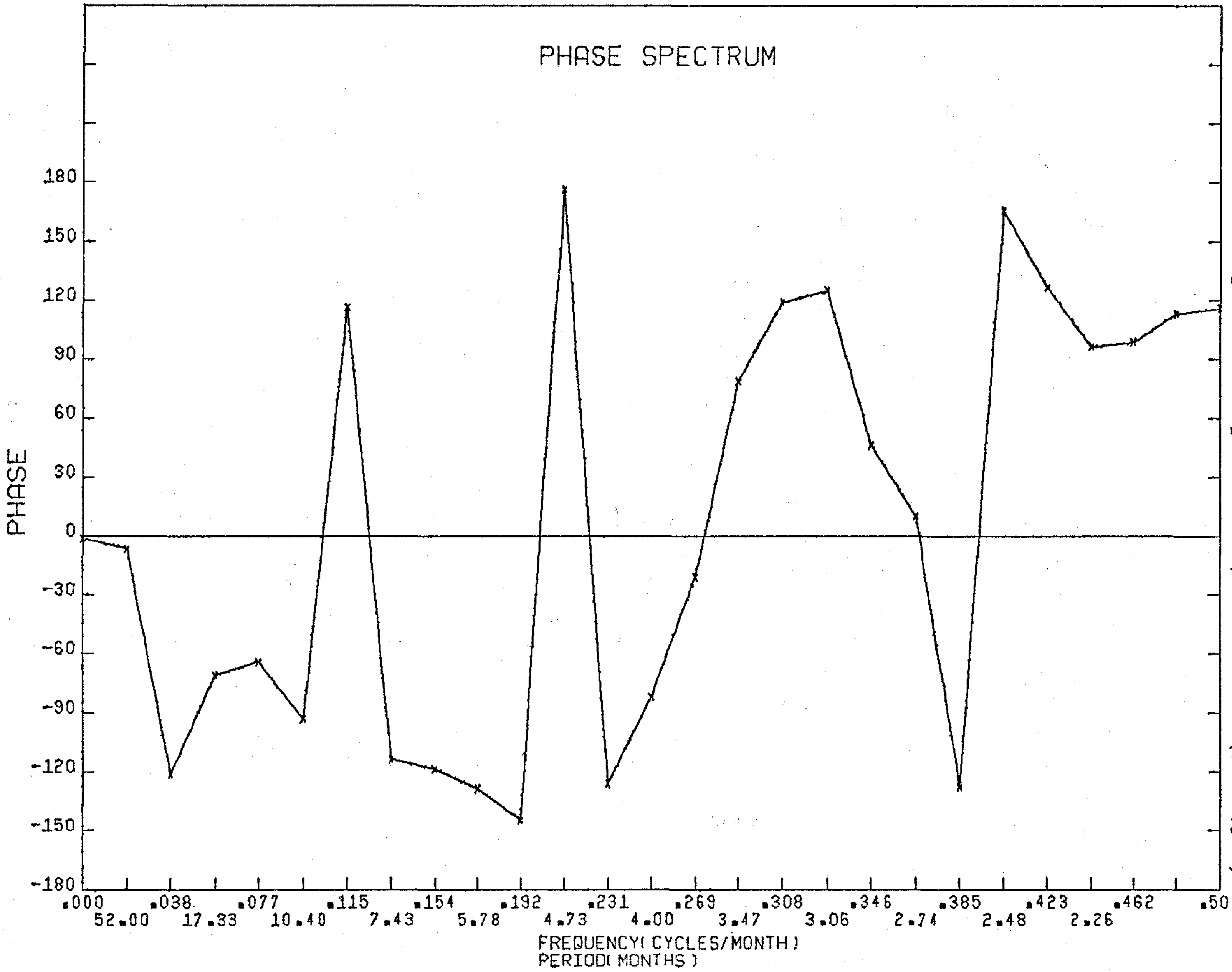


Figure B.7.24: Phase spectrum for pH & SP CON, U.G.L. area (7108-7511)

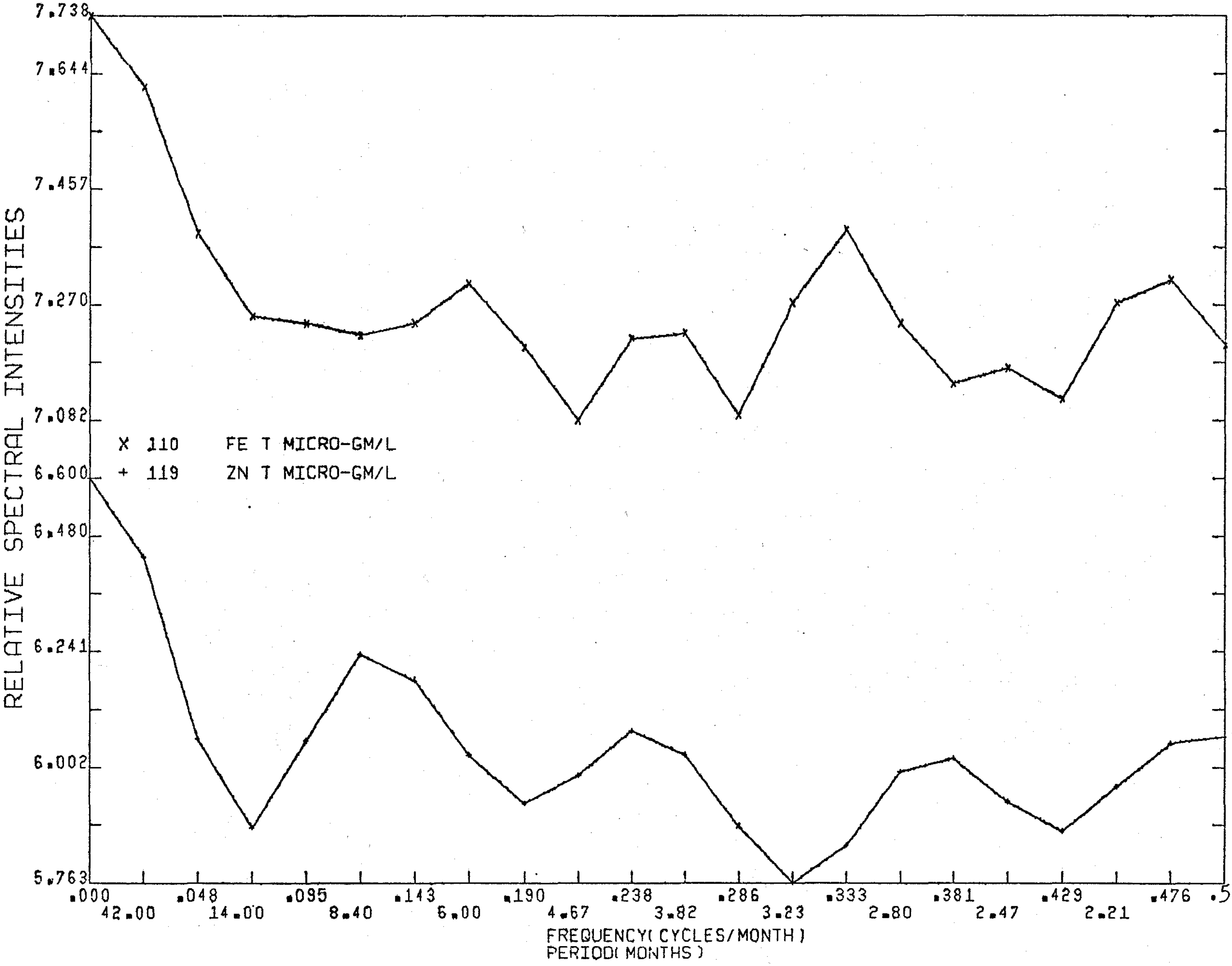


Figure B.7.25: Power spectrum for Fe T & Zn T, station 12 (7206-7511)

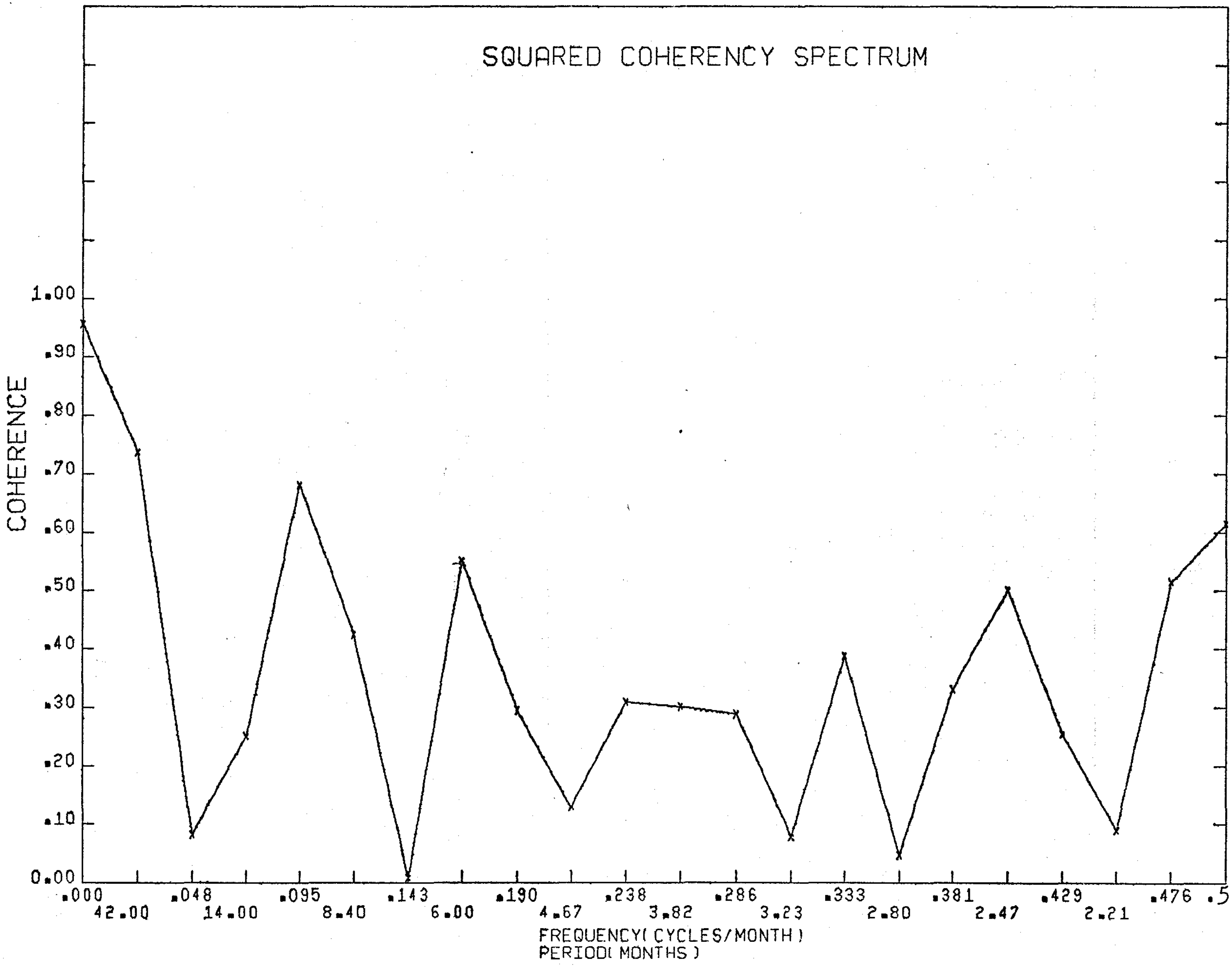
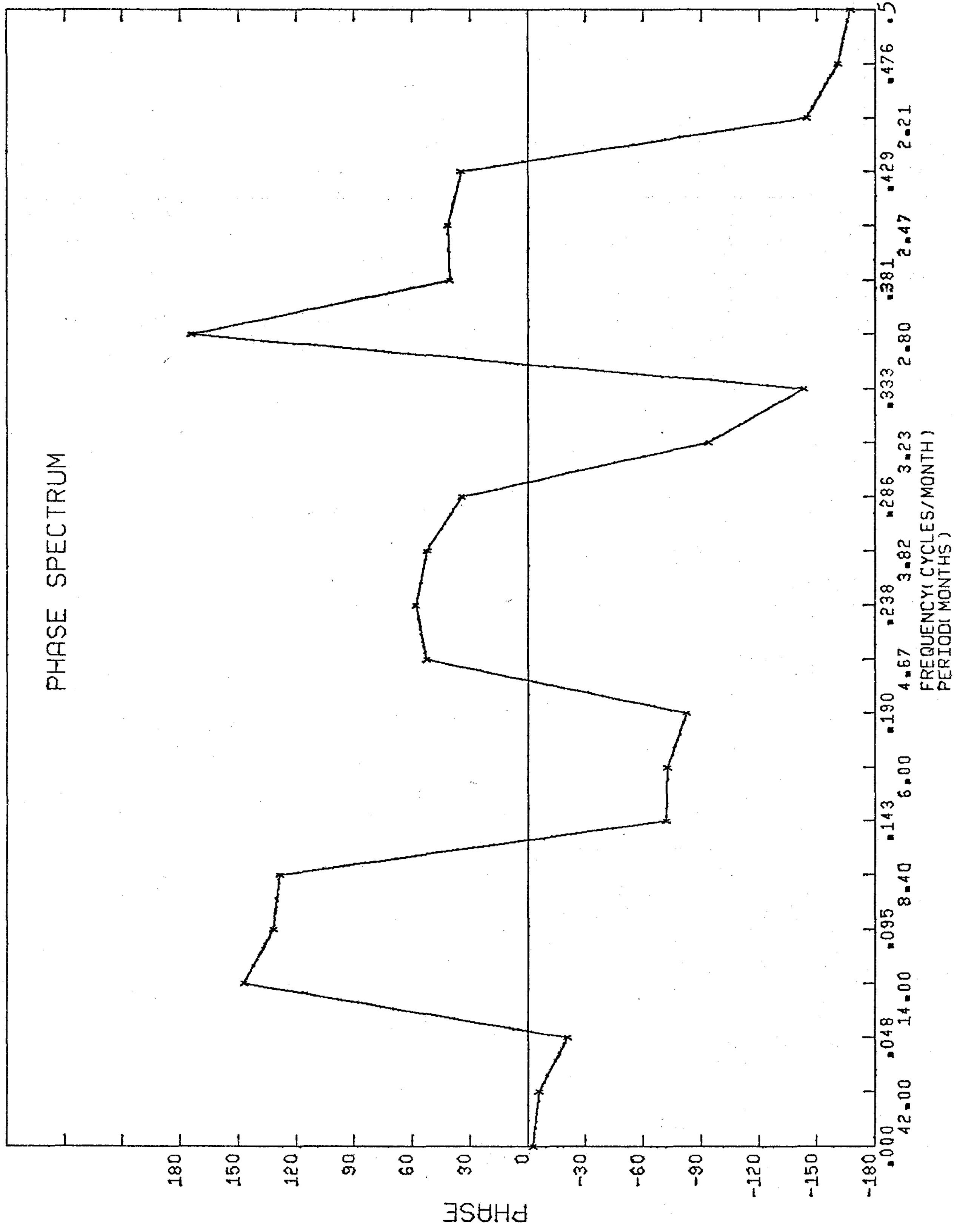


Figure B.7.26: Coherency spectrum for Fe T & Zn T, station 12 (7206-7511)

Figure B.7.27: Phase spectrum for Fe T & Zn T, station 12 (7206-7511)



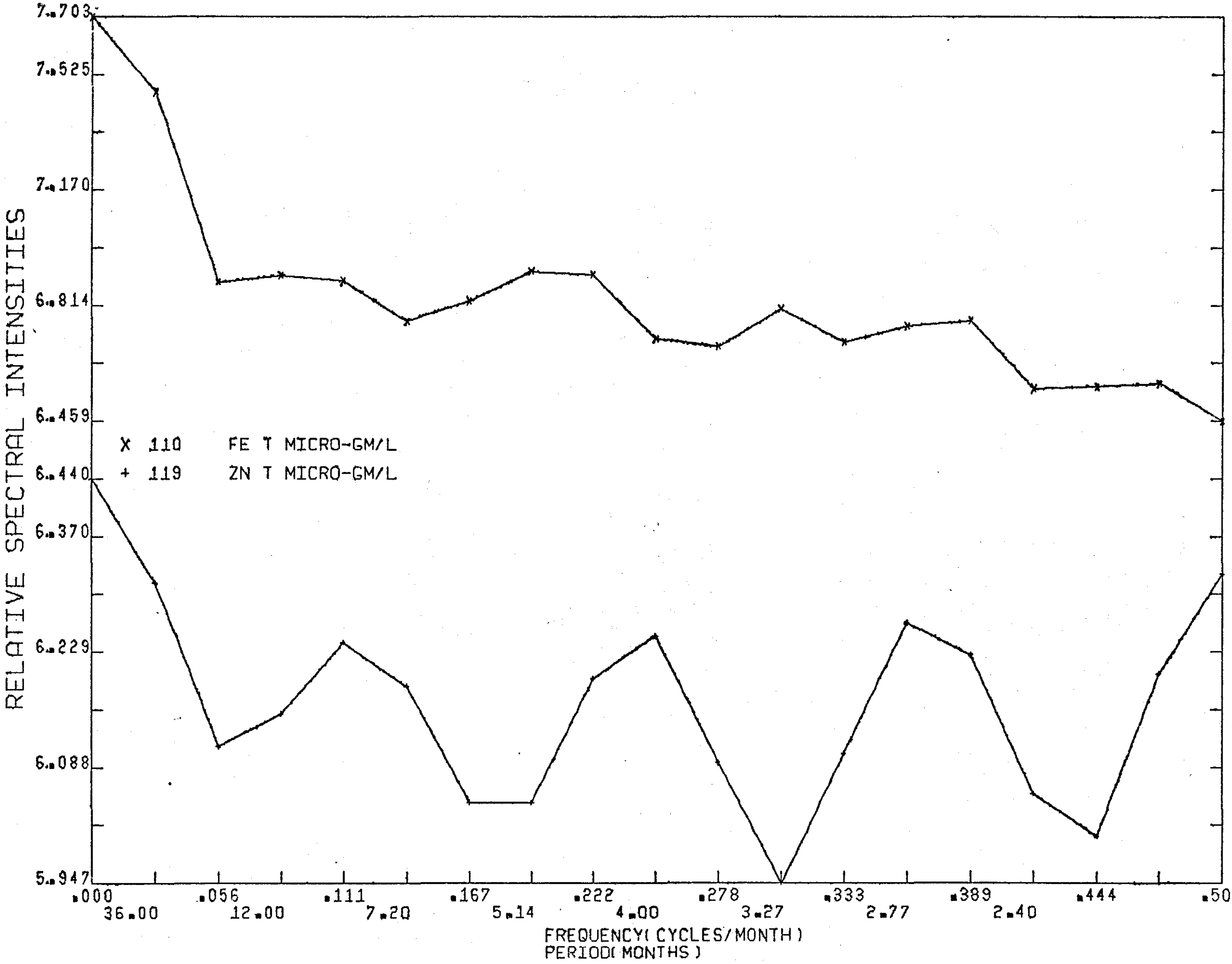


Figure B.7.28: Power spectrum for Fe T & Zn T, station 2 (7206-7510)

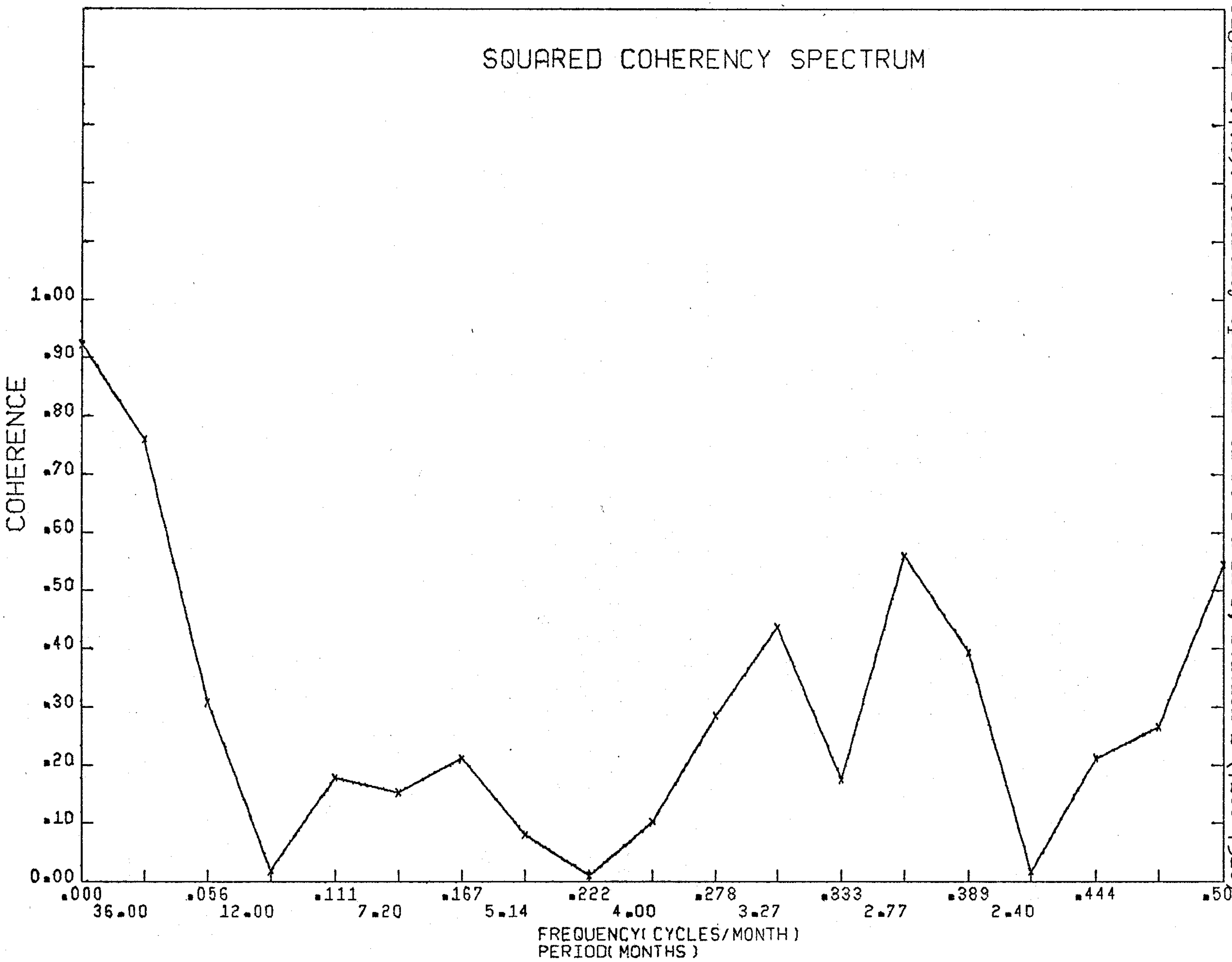


Figure B.7.29: Coherency spectrum for Fe T & Zn T, station 2 (7206-7510)

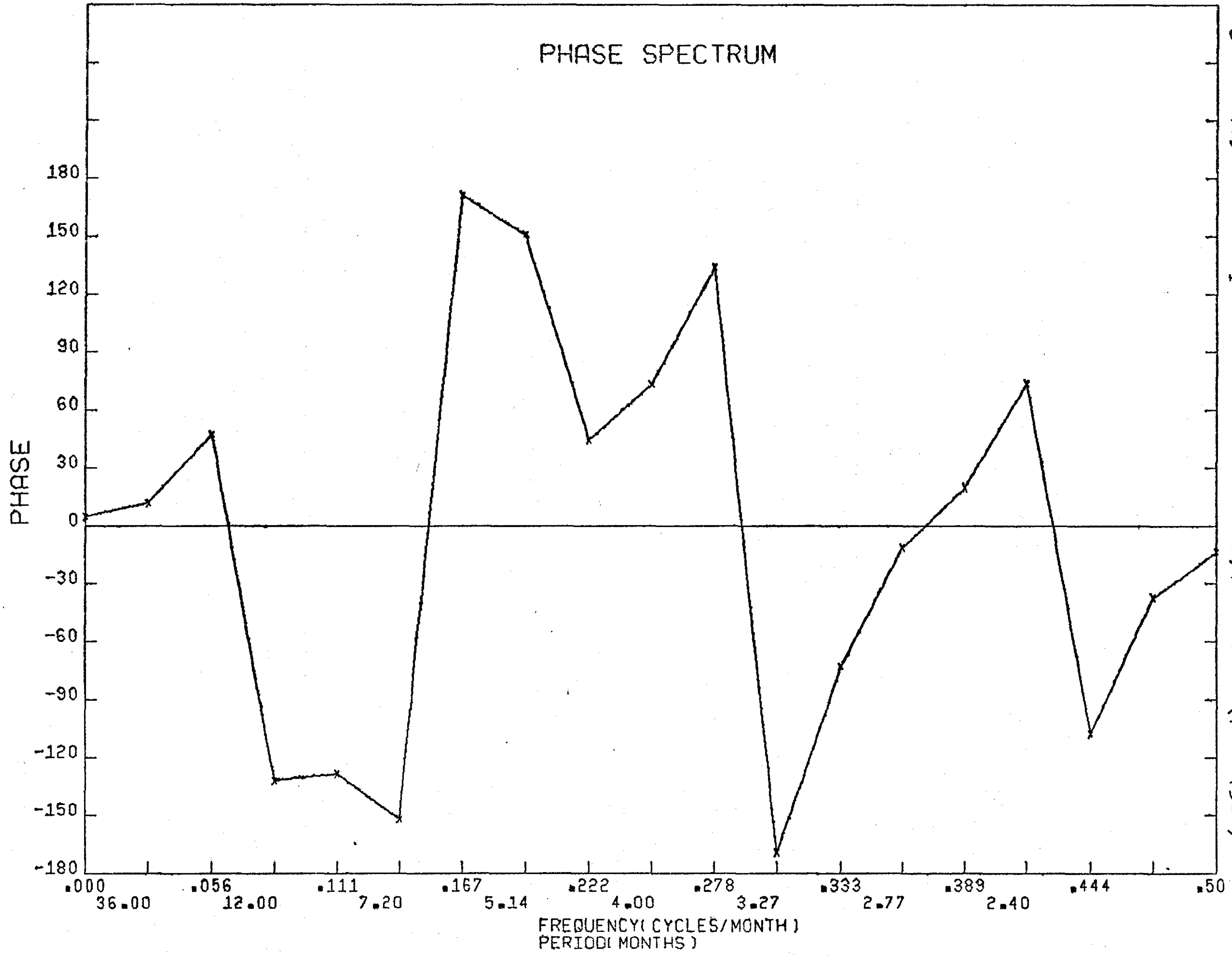


Figure B.7.30: Phase spectrum for Fe T & Zn T, station 2 (7206-7510)

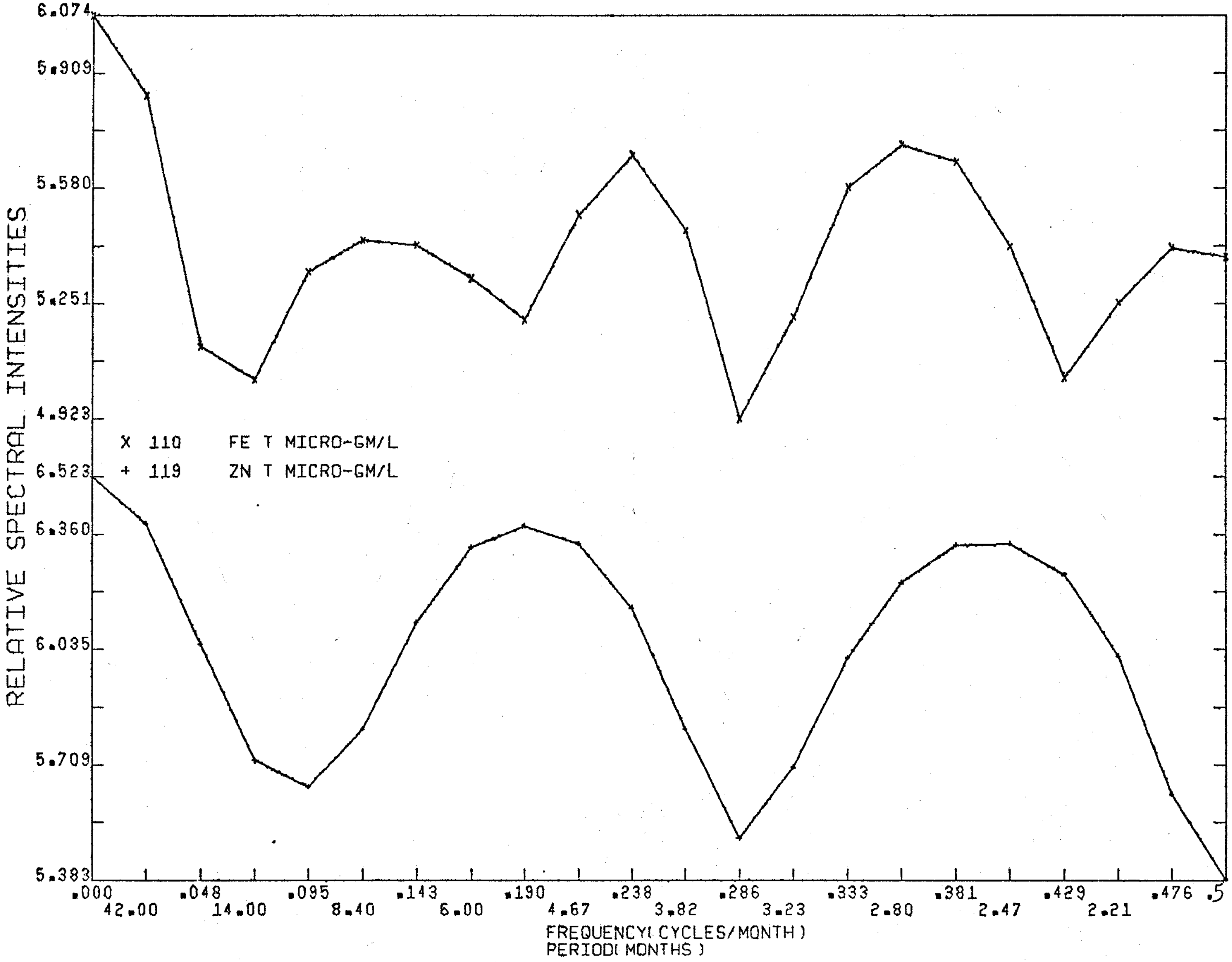


Figure B.7.31: Power spectrum for Fe T & Zn T, station 3 (7206-7511)

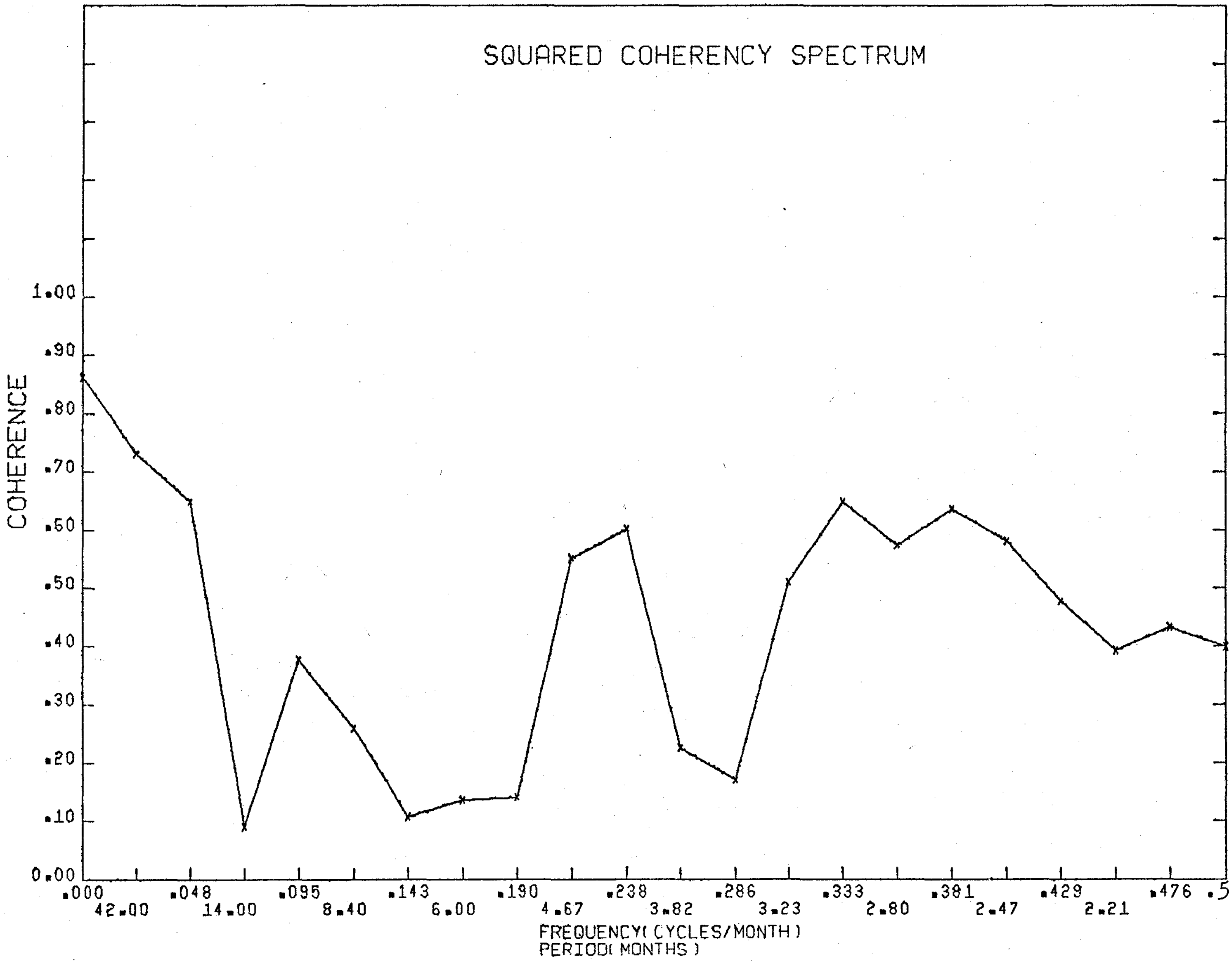


Figure B.7.32: Coherency spectrum for Fe T & Zn T, station 3 (7206-7511)

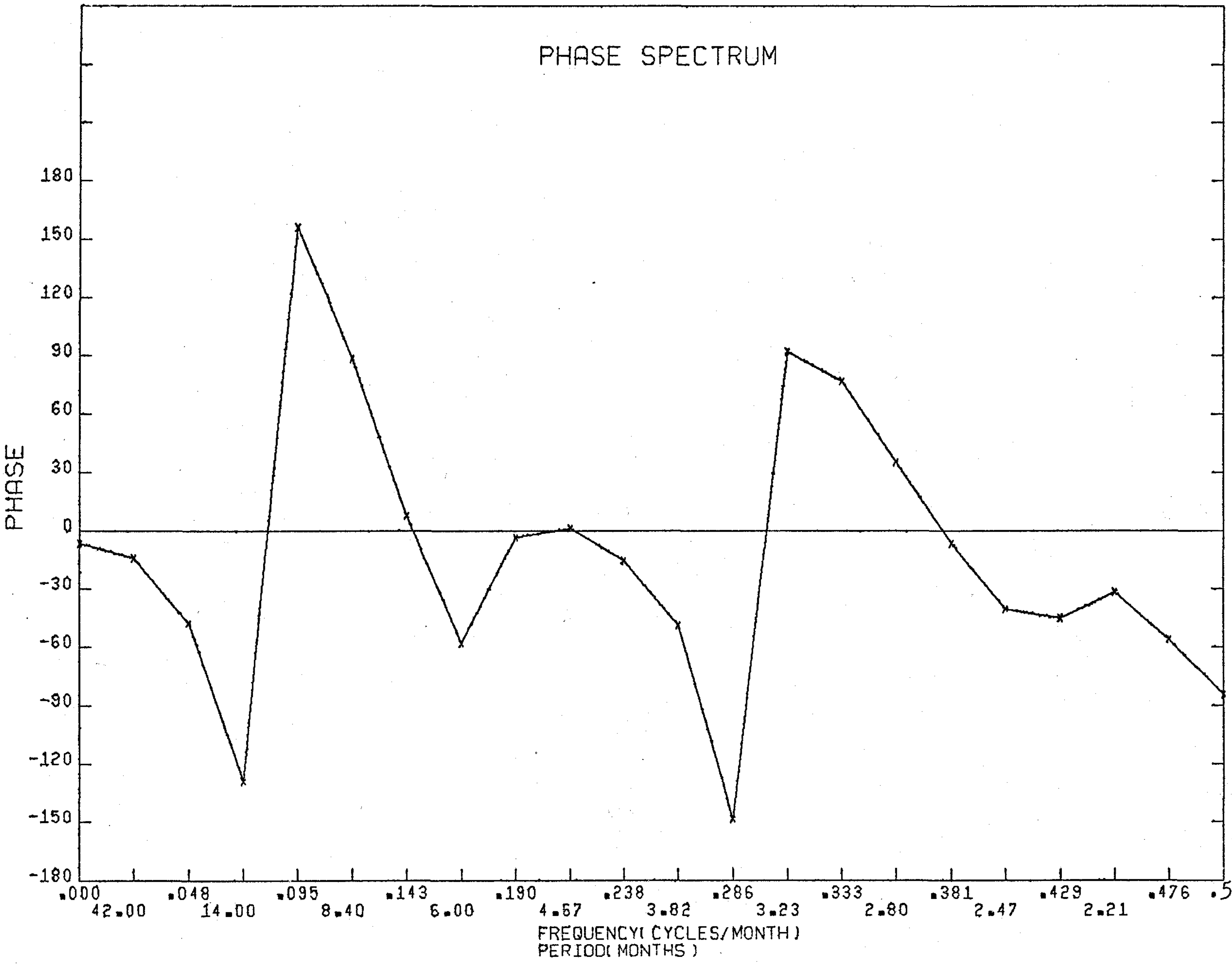


Figure B.7.33: Phase spectrum for Fe T & Zn T, station 3 (7206-7511)

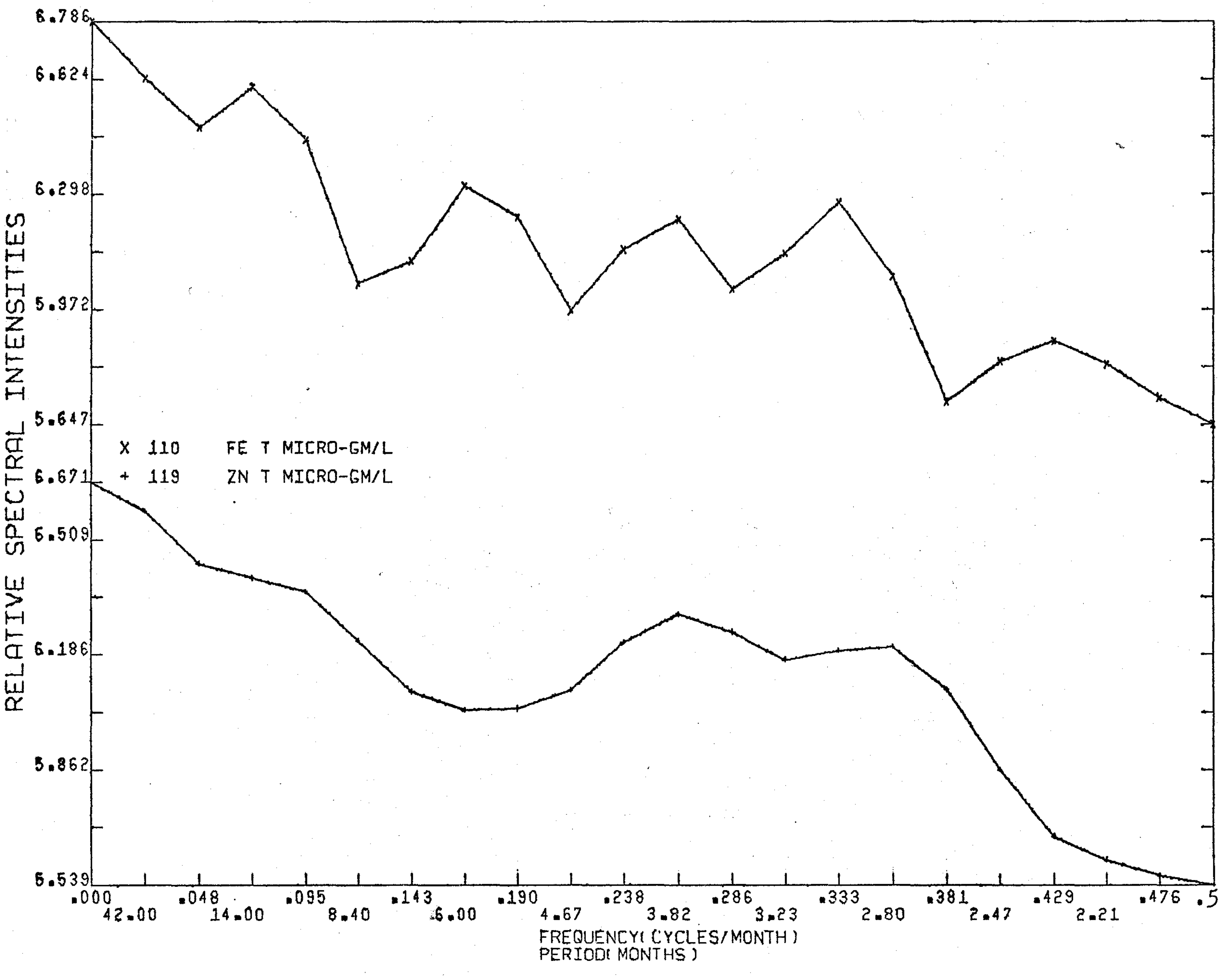


Figure B.7.34: Power spectrum for Fe T & Zn T, station 4 (7204-7509)

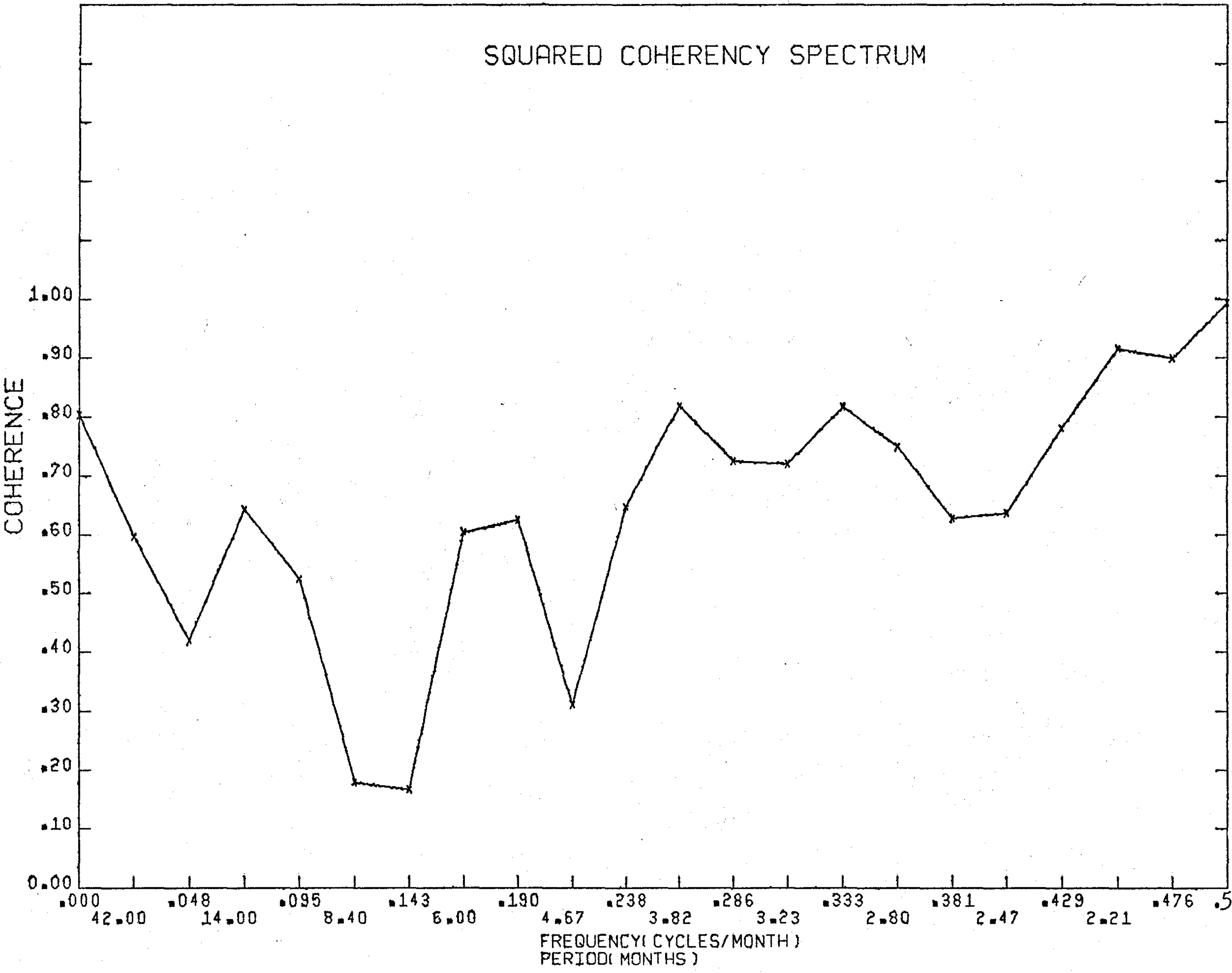


Figure B.7.35: Coherency spectrum for Fe T & Zn T, station 4 (7204-7509)

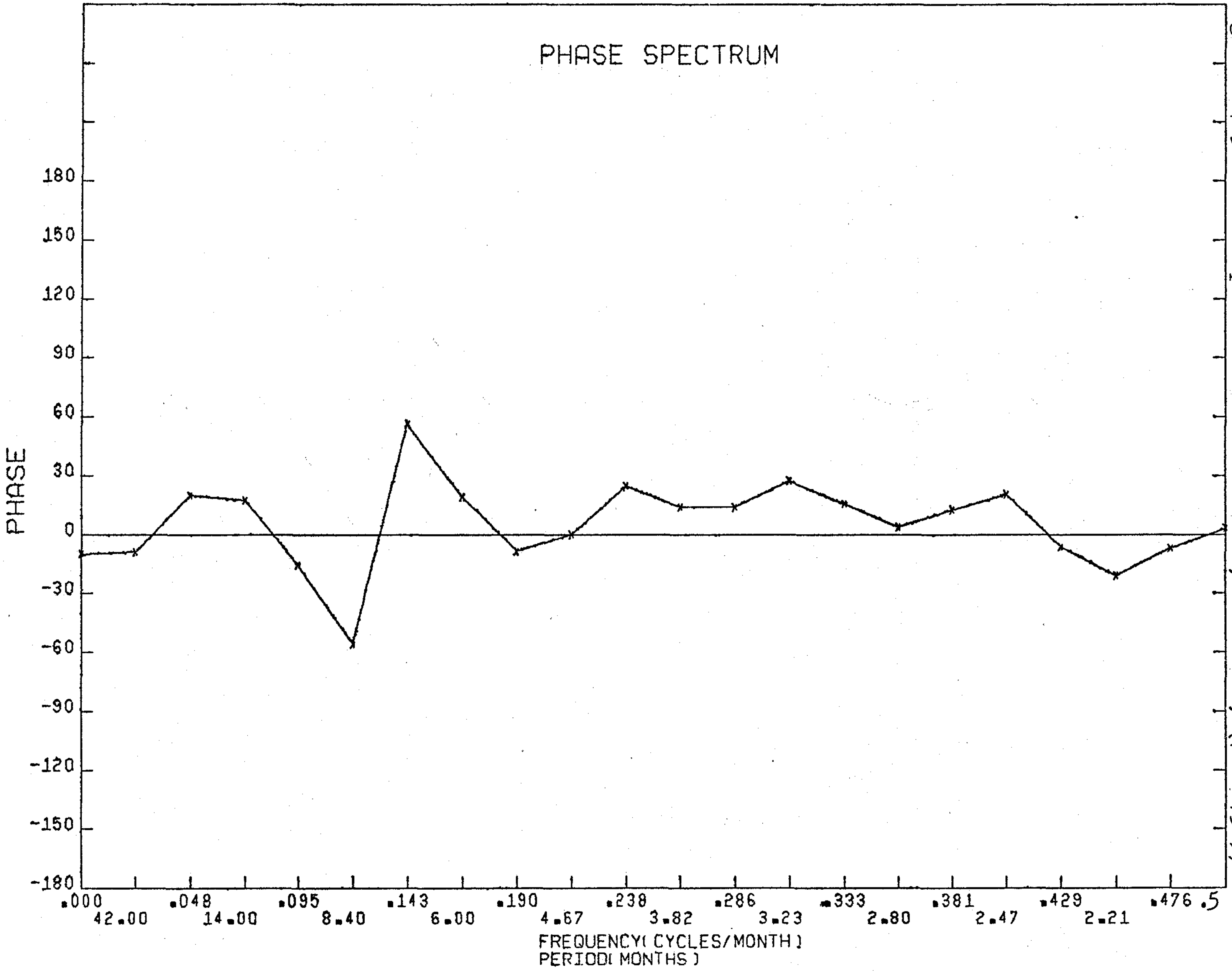


Figure B.7.36: Phase spectrum for Fe T & Zn T, station 4, (7204-7509)

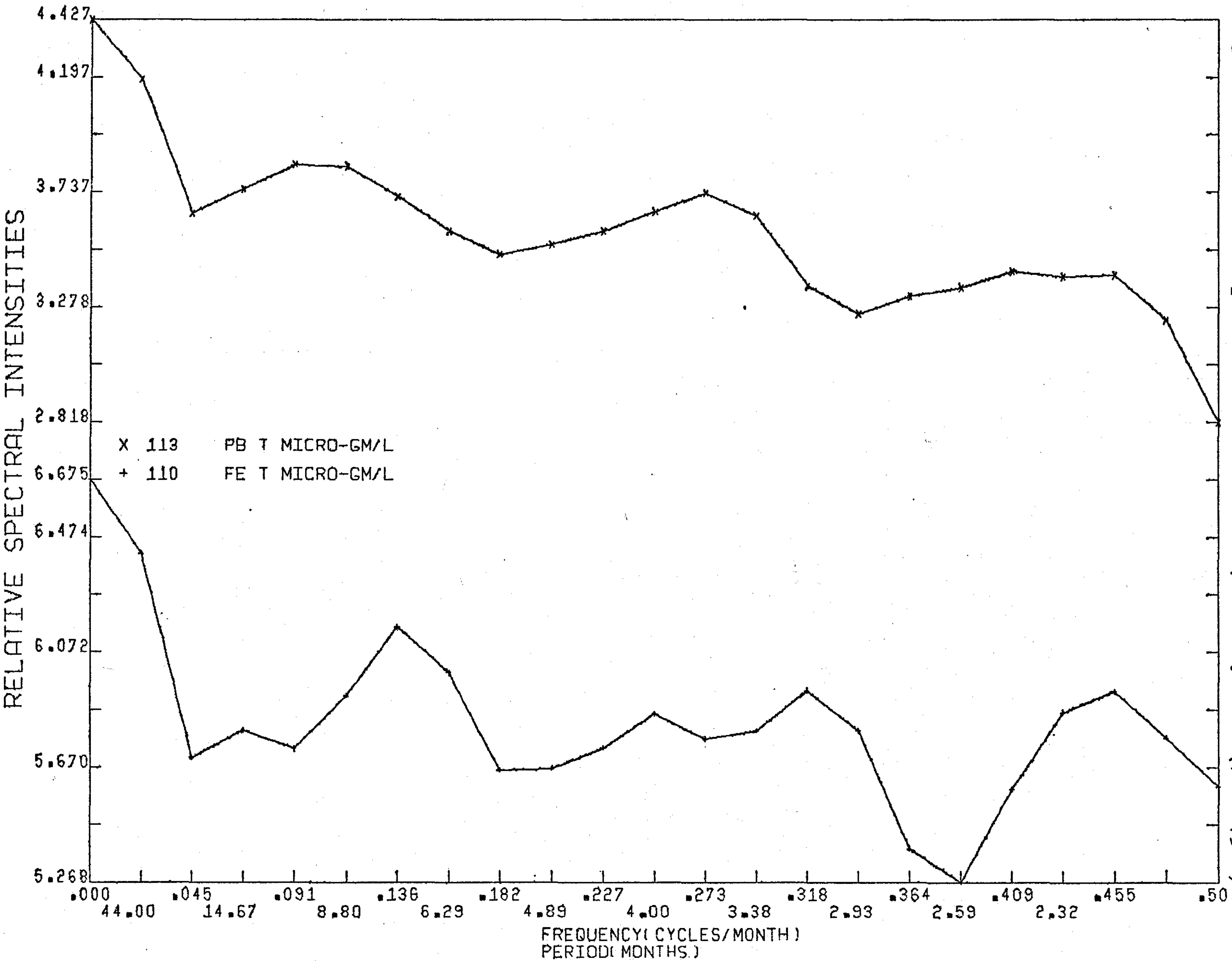


Figure B.7.37: Power spectrum for Fe T & Pb T, Sudbury area (7204-7511)

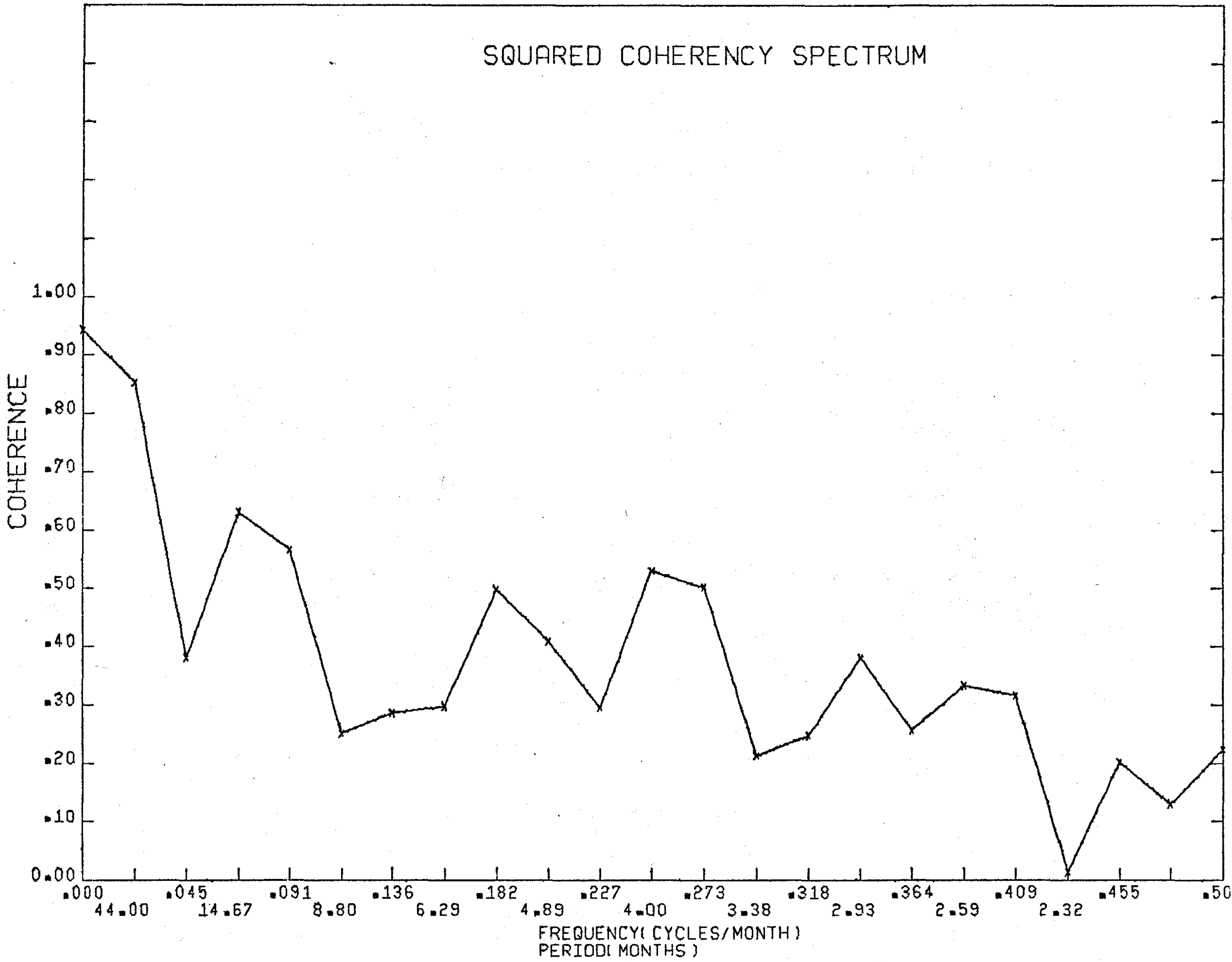


Figure B.7.38: Coherency spectrum for Fe T & Pb T, Sudbury area (7204-7511)

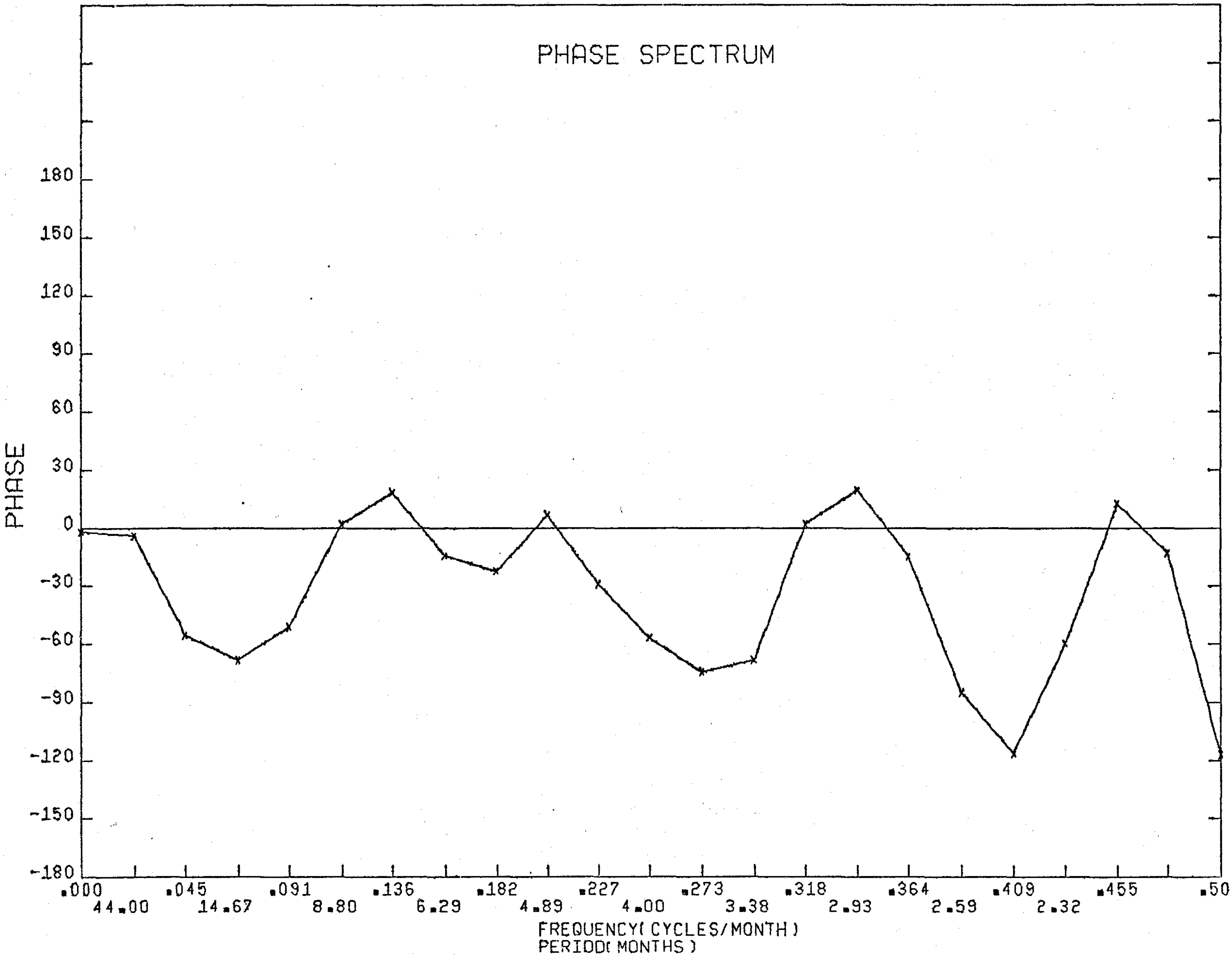


Figure B.7.39: Phase spectrum for Fe T & Pb T, Sudbury area (7204-7511)

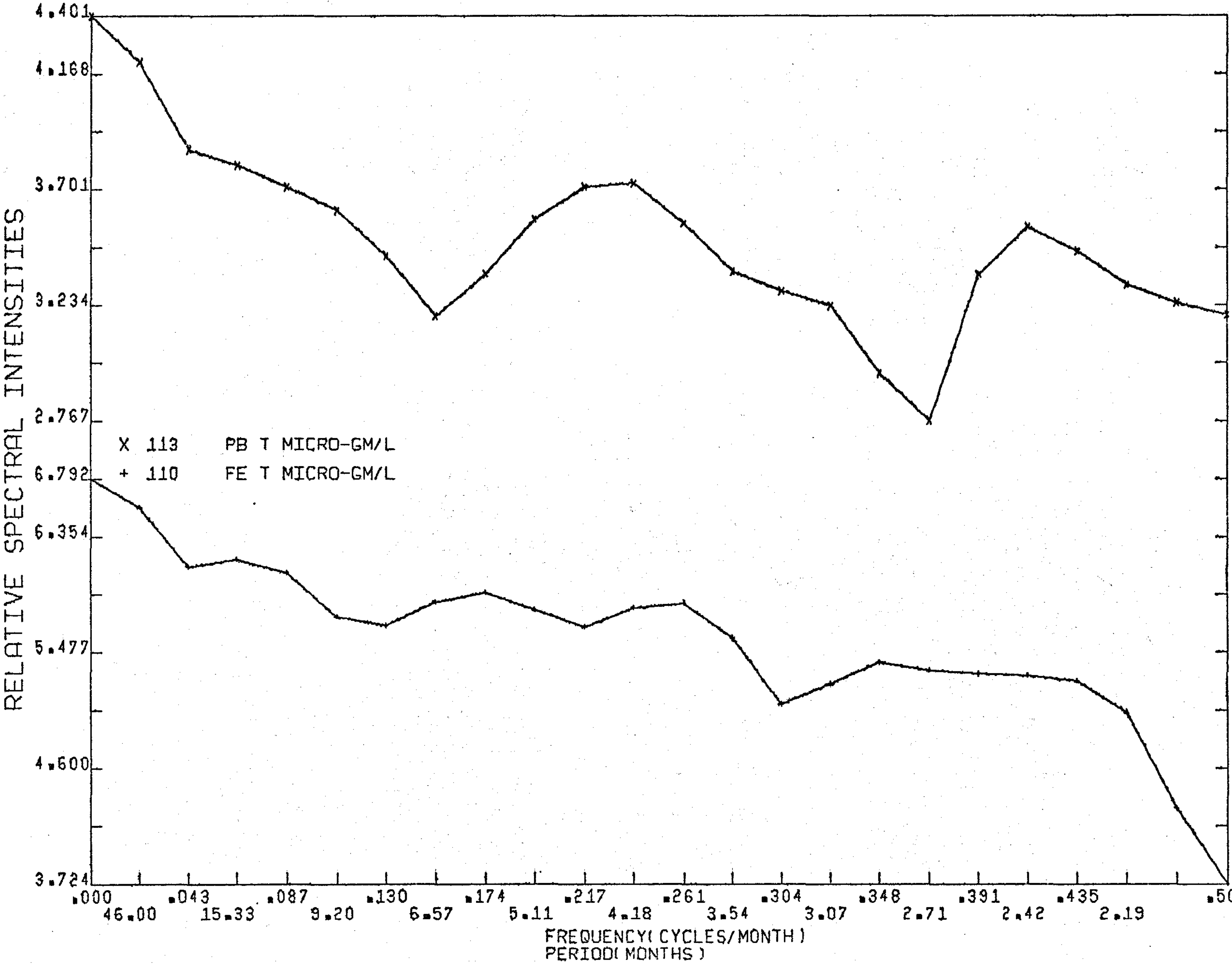


Figure B.7.40: Power spectrum for Fe T & Pb T, U.G.L. area (7201-7511)

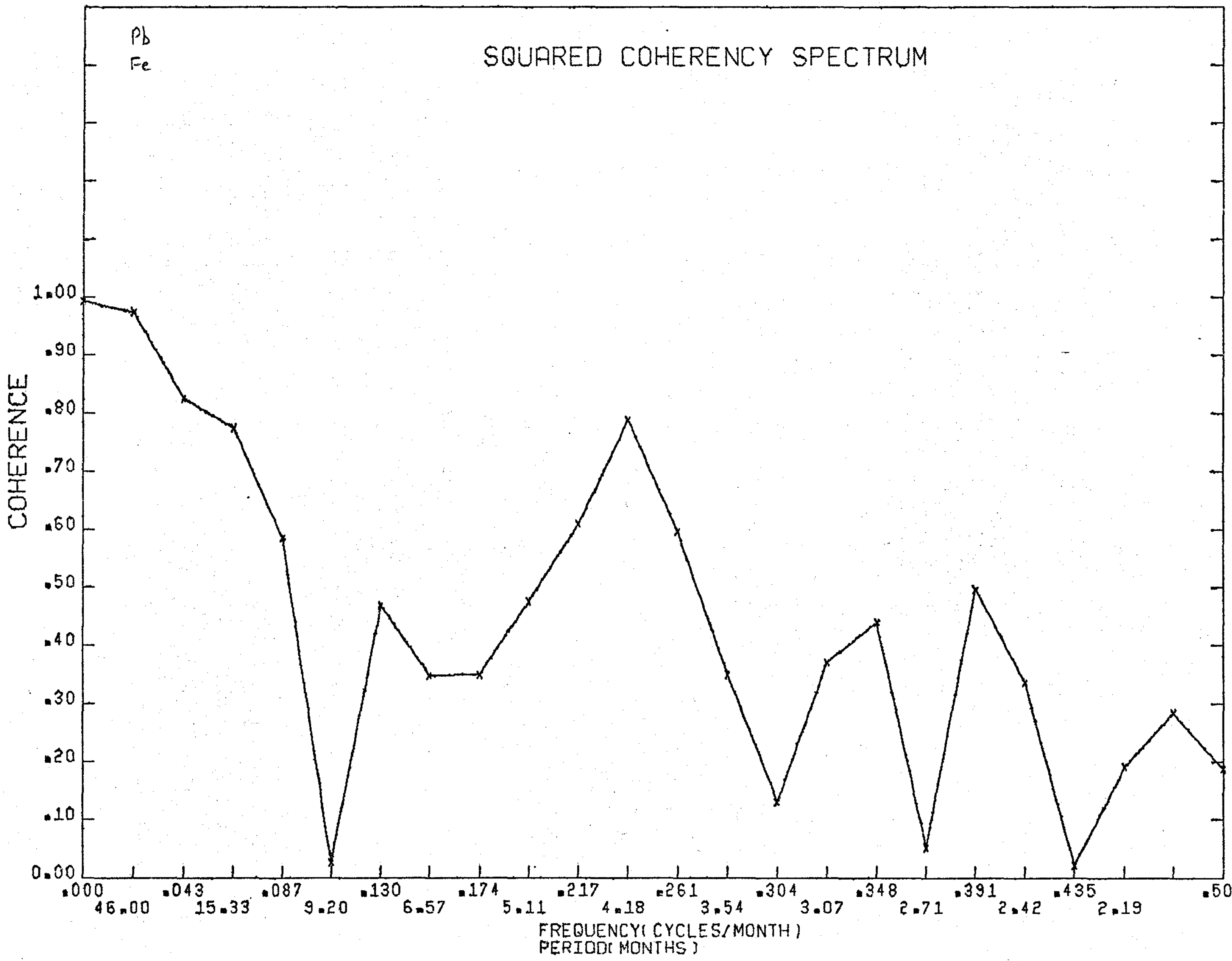


Figure B.7.41: Coherency spectrum for Fe T & Pb T, U.G.L. area (7201-7511)

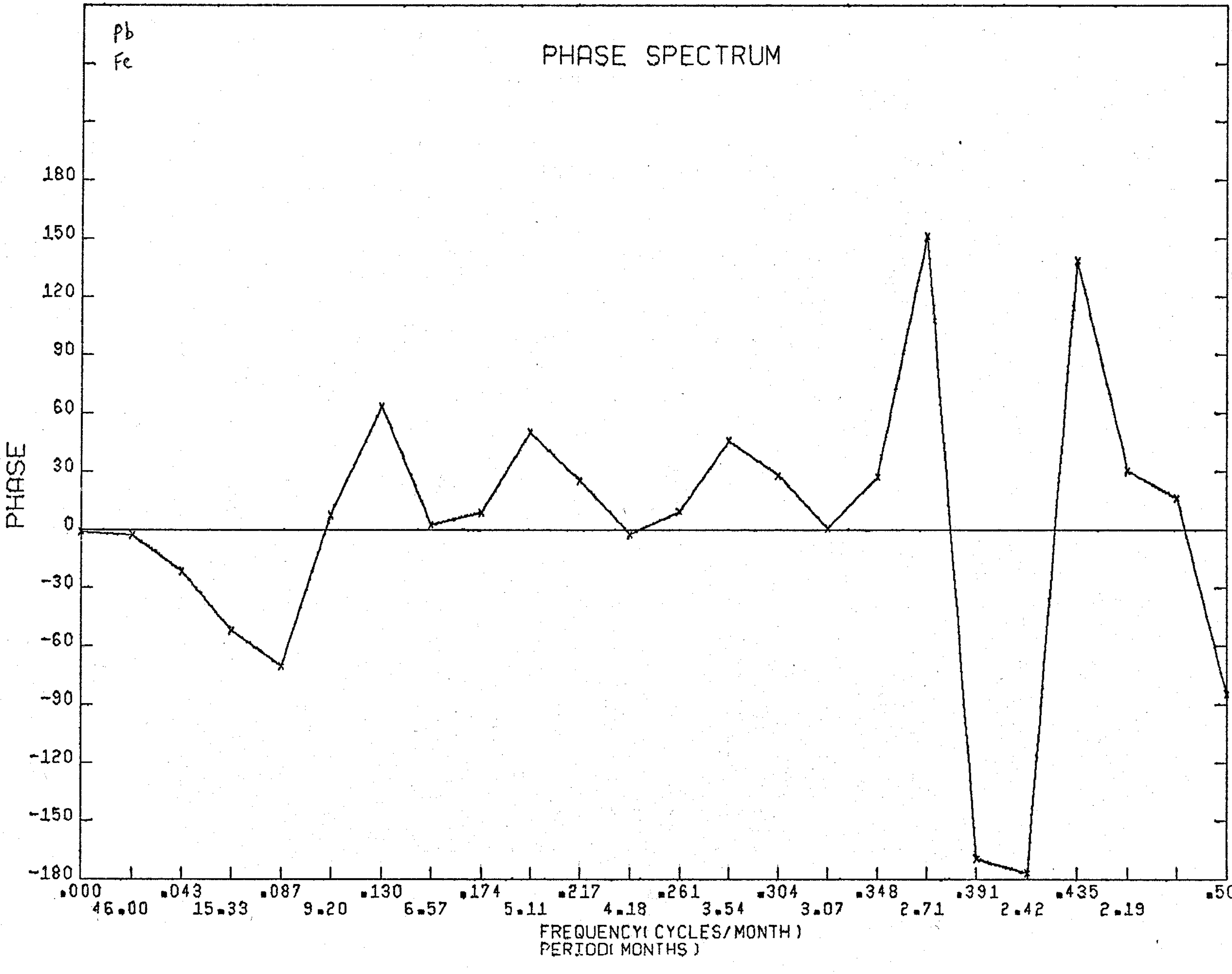


Figure B.7.42: Phase spectrum for Fe T & Pb T, U.G.L. area (7201-7511)

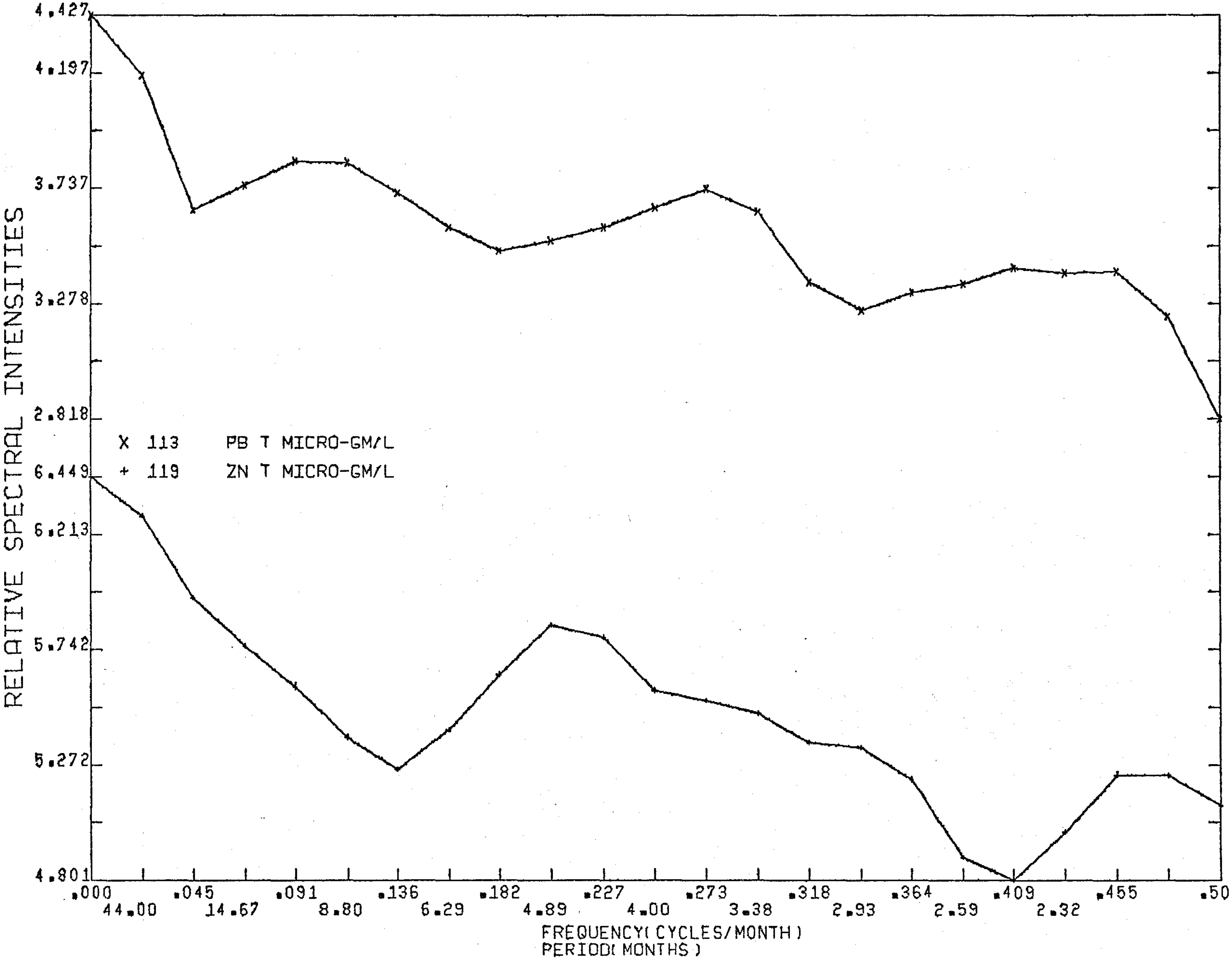


Figure B.7.43: Power spectrum for Pb T & Zn T, Sudbury area (7204-7511)

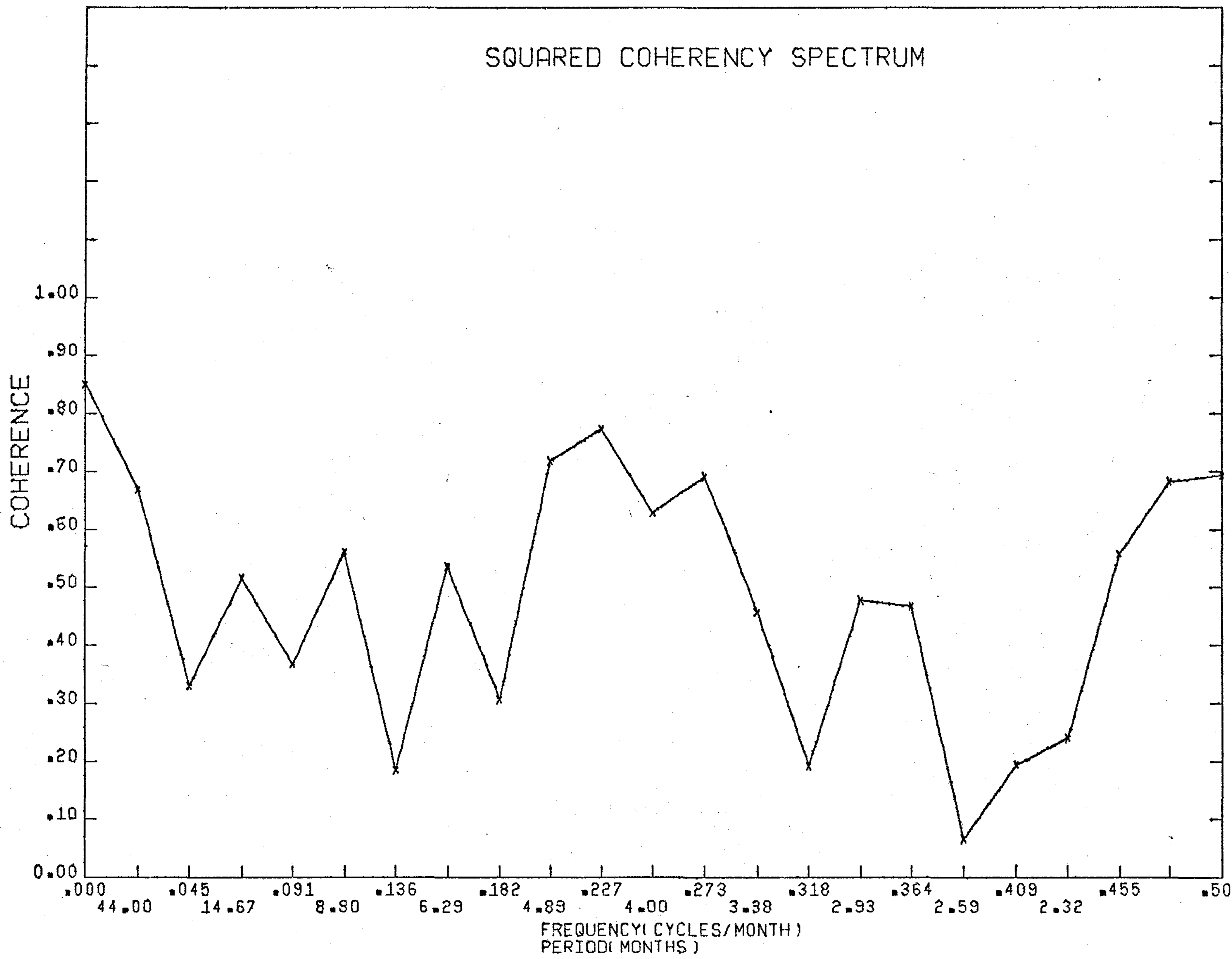


Figure B.7.44: Coherency spectrum for Pb T & Zn T, Sudbury area (7204-7511)

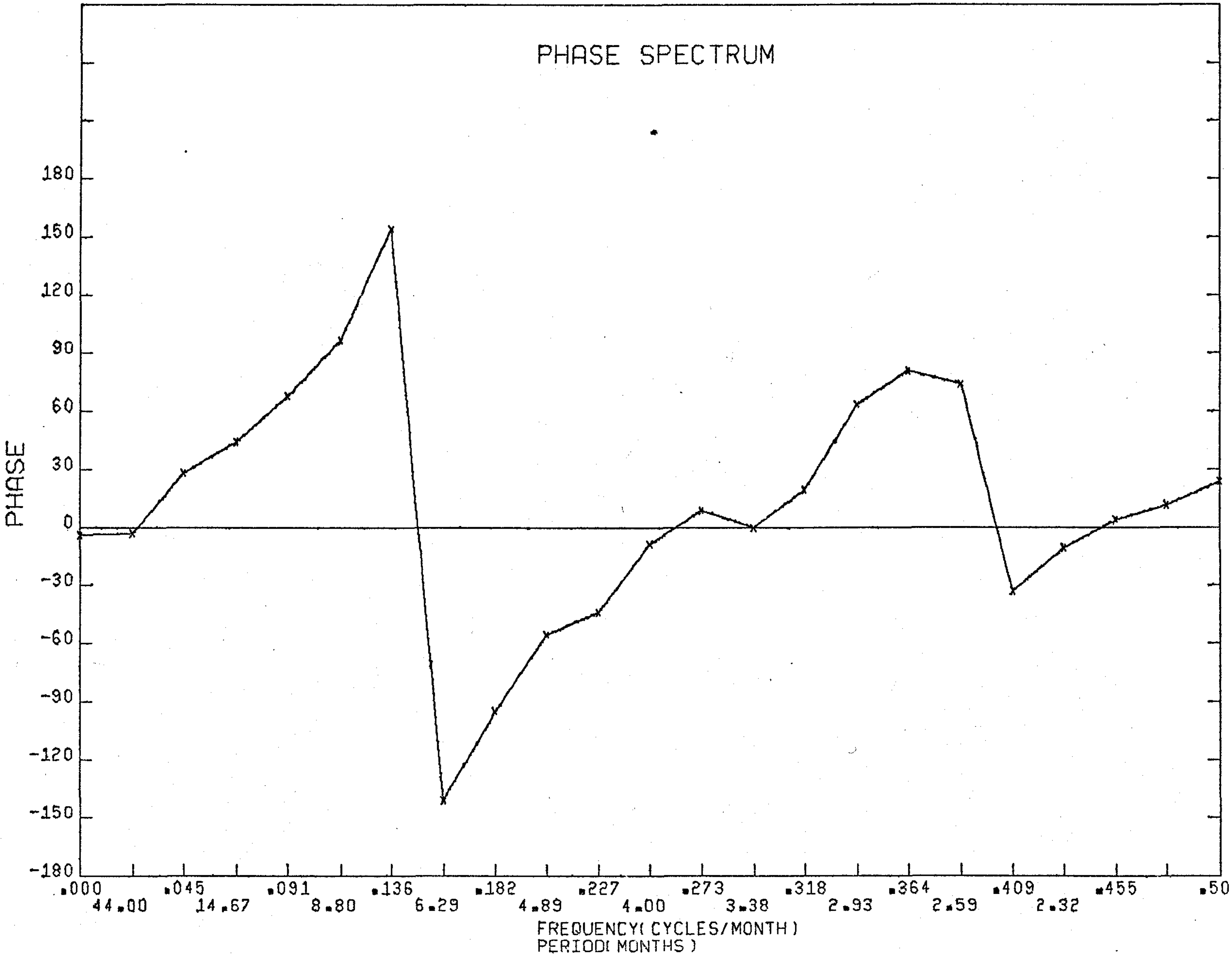


Figure B.7.45: Phase spectrum for Pb T & Zn T, Sudbury area (7204-7511)

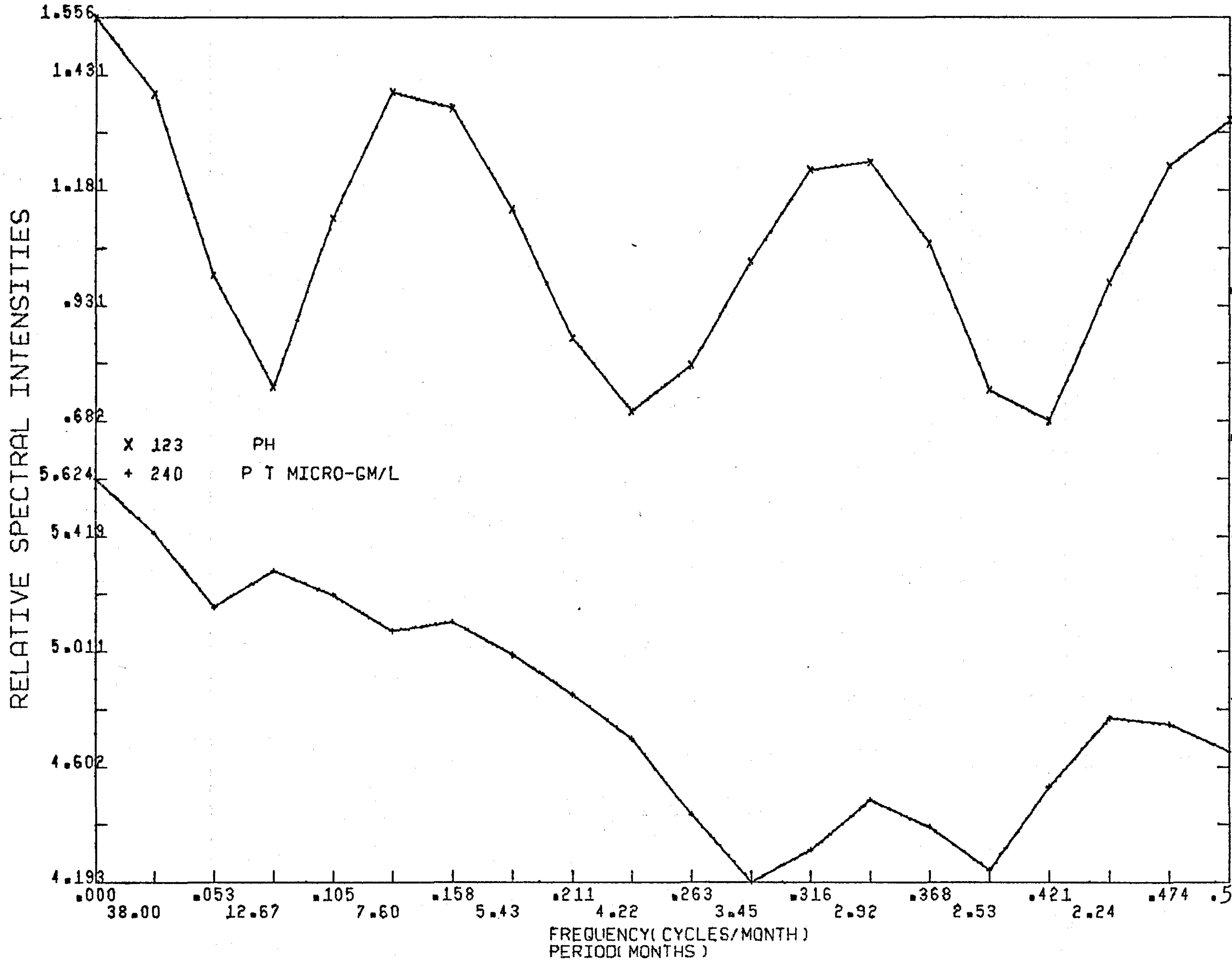


Figure B.7.46: Power spectrum for pH & P T, station 12 (7210-7511)

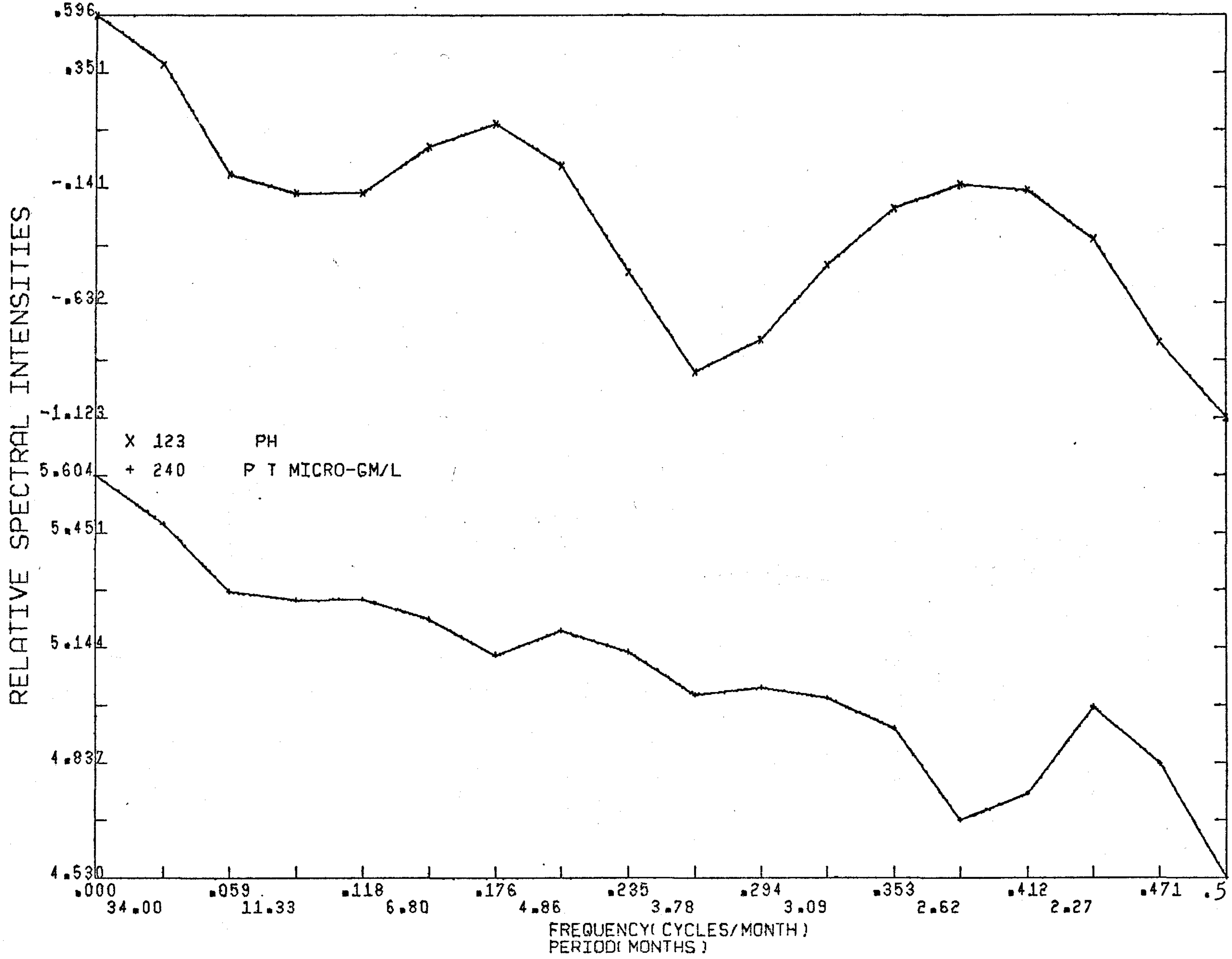


Figure B.7.47: Power spectrum for pH & P T, station 2 (7211-7510)

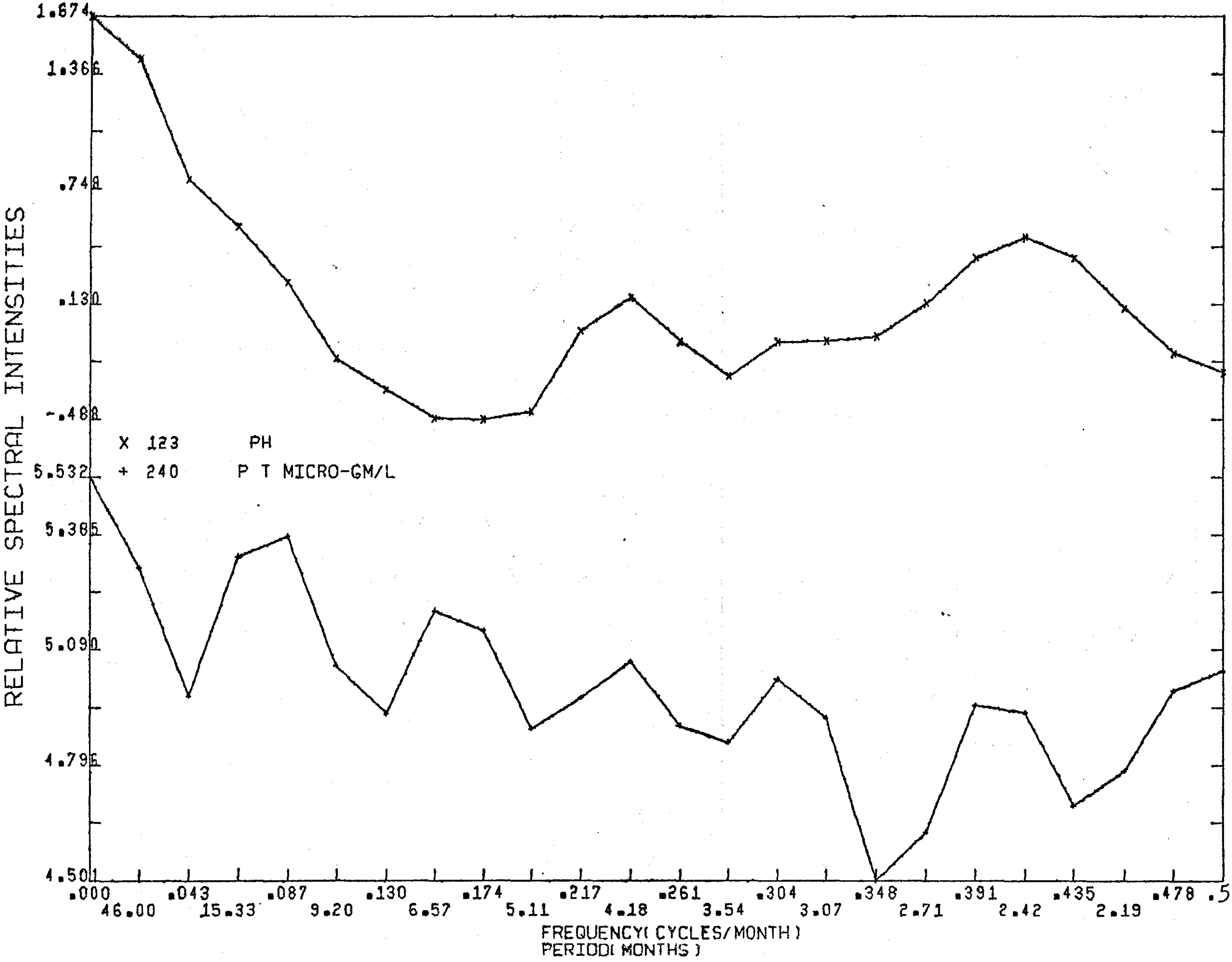


Figure B.7.48: Power spectrum for pH & P T, U.G.L. area (7201-7511)