Reviewer 1:

Comments:

The author presents results of applying Fourier and other reconstruction and fitting methods suggested as suitable for time domain data with non-stationary signals. The author emphasizes use of their previously-published "JCFit" approach to fit time domain data. Only a limited amount of simulated data is evaluated, using modest numbers of points (~2028) and a small number of signals.

There are useful ideas in this work, and also enthusiastic scholarship, and the manuscript deserves refinement. That said, in its current form, it seemed hard to find a coherent story or an actionable conclusion. Here is some discussion that could lead to a stronger manuscript:

Response:

I thank the reviewer for the constructive and thoughtful comments. I agree that the current manuscript needs a clearer focus, a more coherent story, and more actionable conclusions. In the revised manuscript, I have sharpened the central goal, clarified the distinction between transform-based analysis and direct nonlinear time-domain fitting, and revised the title, abstract, discussion, and conclusion accordingly. The revisions are highlighted in yellow in the tracked-changes version of the manuscript and the page number is for the track version not the final manuscript.

1. What is the goal? Is it extraction of model parameters, generation of spectrograms, or deciding when reconstruction methods fail and model fitting is needed?

The present study does not attempt to establish a universal threshold at which FFT should be replaced by model fitting. Instead, it examines conditions under which complementary methods may be informative. In particular, direct nonlinear fitting may be considered when a rapidly decaying or strongly broadened signal produces a poorly resolved Fourier spectrum, while the number and approximate frequencies of the components can still be constrained by prior information or preliminary spectral analysis. I have revised the abstract on page 1, discussion, and conclusion to clarify this more limited objective.

> "This simulation study evaluates complementary methods for short or non-stationary FID signals in which linewidth, dephasing, or decay behavior changes during acquisition." And "These methods therefore serve complementary roles in analyzing stationary and non-stationary NMR signals."

On page 3: "In this study, simulated FID signals are analyzed using FFT, STFT, wavelet analysis, and direct nonlinear time-domain fitting to evaluate their complementary roles and limitations. This study does not propose a new Fourier, wavelet, or nonlinear-fitting theory. Instead, it provides a simulation-based evaluation of the complementary roles of these methods for stationary, very short, and non-stationary NMR FIDs. The analysis focuses on dynamically broadened signals, the approximation of finite collection-window measurements by point sampling, and the potential and limitations of JCFit for estimating parameters from very short, relatively simple FID segments."

2. There are decades of work on spectral fitting, but most work addresses stationary NMR signals. Non-stationary signals are less common and more in need of analysis methods.

I agree with the reviewer. This is an important point, and I have revised the abstract, and title as the reviewer has suggested in Comment 5, to emphasize non-stationary or dynamically broadened FID signals more clearly. The revised discussion distinguishes stationary FID signals, for which conventional Fourier-domain methods are highly effective, from non-stationary signals, where frequency, linewidth, or decay behavior may evolve during acquisition and where STFT, wavelet analysis, or direct time-domain fitting may become useful. I have also revised the discussion section on page 13 to reflect this important point the reviewer has brought up.

> "When the acquisition duration is short and comparable to or shorter than one wave cycle, the resulting Fourier spectrum becomes intrinsically broad and poorly resolved because of the finite observation window."
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> "For such short signals, direct nonlinear time-domain fitting of parametrized damped sinusoidal models may provide complementary estimates of frequencies, amplitudes, phases, and decay constants."

3. The title "Comparing Fourier Transform and Direct Wave Fitting" could be improved because these methods do not do the same thing.

I agree. Fourier transformation and direct time-domain fitting serve different purposes. FFT deterministically transforms sampled time-domain data into a frequency-domain representation, while nonlinear time-domain fitting estimates model parameters and requires choices about the functional form, number of components, initial guesses, and fitting strategy. In the revised manuscript, I avoid presenting these as equivalent alternatives. I agree that the title should better reflect the manuscript's scope, and I have revised the title to: **"Simulating the Extraction of Signal Parameters and Spectrograms for Non-Stationary NMR Signals."** This title more accurately describes the simulation-based nature and intended focus of the work.

4. There is little discussion of how the number of signals is determined, which is often one of the most difficult aspects of NMR fitting.

This is an excellent point. I agree that determining the number of signal components is a major challenge in time-domain NMR fitting and should be discussed explicitly. In the current implementation, initial frequency guesses can be obtained from FFT peak detection, but this does not solve the general problem of model selection. In the revised manuscript, I have clarified that the present implementation requires an assumed or estimated number of components. FFT peak detection and prior chemical knowledge can provide initial estimates, but they do not solve the general model-selection problem. Residual analysis and statistical criteria such as AIC or BIC may be useful in future implementations, although their performance has not been evaluated in the present study.

> Page 13: "Direct fitting becomes increasingly difficult for complex molecules with many overlapping and unknown components; in such cases, Fourier-domain analysis and prior chemical information can provide initial frequency estimates for subsequent model fitting to estimate parameters associated with non-stationary decay behavior. However, fully automatic and reliable determination of the number of signal components remains challenging."

I also clarify in the discussion that FFT-derived initial guesses are used mainly to reduce fitting time and improve convergence, even when the FFT spectrum itself is too broad or low-resolution for confident interpretation.

> Page 9: "For a well-posed problem that converges to a unique global optimum, the initial guesses would not affect the final solution; in practice, they strongly influence convergence and the solution obtained for non-convex or poorly conditioned problems."

5. A possible title is "Extraction of Signal Parameters and Spectrograms for Non-Stationary NMR Signals."

I thank the reviewer for this helpful suggestion. I agree that this title better reflects the manuscript's direction. To emphasize that the present manuscript is simulation-based rather than a complete experimental study, I revised the title to:

> **"Simulating the Extraction of Signal Parameters and Spectrograms for Non-Stationary NMR Signals."**

6. Fourier-domain and time-domain data contain the same information, although some tasks are easier in one domain than the other.

I agree with the reviewer. For a given discrete dataset, the Fourier-domain representation and the original time-domain data contain equivalent information in principle. The practical difference is not information content, but the ease, stability, and conditioning of extracting particular parameters. Frequency-domain analysis is usually faster, cleaner, and better suited for treating localized spectral regions independently. Time-domain fitting, however, may be useful when one is interested in decay rates, phases, or short-time behavior, although it becomes much more computationally demanding and model-dependent.

I have add a statement in the introduction before introducing the direct wave fitting method:

> Page 2: "For a given discrete dataset, the time-domain and Fourier-domain representations contain equivalent information in principle; the practical distinction concerns how easily and robustly particular parameters can be extracted. An alternative analysis approach is direct nonlinear time-domain fitting of parametrized damped sinusoidal models using least-squares or related objective functions.(Castellano and Bothner-By, 1964; Montigny et al., 1990; Nishiyama and Mita, 1989; Vanhuffel et al., 1994)"

I have revised the conclusion to state this distinction more clearly.

> Page 13: "For such short signals, direct nonlinear time-domain fitting of parametrized damped sinusoidal models may provide complementary estimates of frequencies, amplitudes, phases, and decay constants."

7. The manuscript states that frequency-domain peak fitting and time-domain FID fitting are significantly different, but signal parameters in one domain correspond to parameters in the other.

I agree that the original statement was too strong. I have deleted the sentence "There are significant differences between wave and peak data fitting" and revised the introduction to emphasize that the two domains contain corresponding information, while the numerical conditioning and practical procedures for parameter extraction differ.

8. The discussion of collection windows is useful and could be mentioned in the abstract.

I thank the reviewer for this positive comment. I have added a sentence in the abstract on page 1:

> "Simulations of finite collection windows show that point-sampled FIDs can approximate window-integrated signals after amplitude and phase correction when the collection window is sufficiently sharp and reproducible."

9. The statement about zero filling needs correction.

I agree and have revised the manuscript. The original wording incorrectly implied that zero filling adds enough data points for FFT "to work." I have revised this to state that zero filling is commonly used to interpolate the discrete spectrum, improve visual appearance, and facilitate certain peak-picking or processing algorithms, but it does not add new experimental information or recover missing frequency resolution beyond what is contained in the measured time-domain data.

In the abstract on page 1:

> "Zero filling improves spectral interpolation but does not recover frequency resolution lost through short acquisition."

In the conclusion on page 13-14:

> "Zero filling can interpolate the discrete spectrum and improve its visual appearance or facilitate peak picking, but it does not add experimental information or recover the missing frequency resolution."

10. Are there guidelines regarding data size and signal count for choosing the best approach?

I thank the reviewer for raising this important question. The present study does not establish universal quantitative thresholds based on data size or signal count. Such thresholds are unlikely to depend on these variables alone because performance also depends on signal-to-noise ratio, spectral overlap, decay rate, model complexity, sampling interval, and prior constraints.

In the revised abstract, I have clarified that these approaches are complementary rather than interchangeable:

> “These methods therefore serve complementary roles in analyzing stationary and non-stationary NMR signals.”

I have also restricted the proposed application of direct model fitting to very short signals with durations comparable to one oscillation period and explicitly stated its limitations on page 13:

> “For such short signals, direct nonlinear time-domain fitting of parametrized damped sinusoidal models may provide complementary estimates of frequencies, amplitudes, phases, and decay constants. However, this approach is challenging because the objective-function landscape can contain many local minima, and the fitted results depend strongly on the assumed number of signal components, the model form, the initial parameter estimates, and the signal-to-noise ratio.”

In current NMR practice, dynamic information is often obtained through specialized pulse sequences and multidimensional experiments. Direct model fitting may provide an additional approach for extracting dynamic parameters from selected short or non-stationary FID segments, but its practical range of applicability requires further systematic evaluation.

Reviewer 2:

Comments:

This reviewer is unable to discern any novel results in this manuscript. Furthermore, the manuscript is highly misleading and fails to grasp the important distinctions among methods. The discussion of "least squares" methods implies they are a separate class from the discrete Fourier transform or the discrete wavelet transform. The discrete Fourier transform is a "least squares" solution to the problem of expanding a time series in a discrete Fourier basis. There happens to be a particularly efficient method for computing the coefficients, and there is a unique solution, because the Fourier basis is orthogonal. The same holds for the discrete wavelet transform -- it is a least-squares solution to an expansion in an orthogonal wavelet basis. Least squares solutions using other models -- for example exponentially decaying sinusoids -- don't have such efficient methods for computing the coefficients, nor do they typically have a unique solution without some additional regularization criterion (as exponentially decaying sinusoids form an "over complete" basis). It's noteworthy that the orthogonality of wavelet bases precludes perfectly symmetric wavelets (although nearly-symmetric symlets have been developed) which makes them less than ideal for fitting symmetric NMR lineshapes. All of these observations are reflected in more than five decades of results on signal processing of time series.

Response:

I thank the reviewer for this candid and technically important critique. I agree that the original manuscript used the term “least-squares fitting” too broadly and did not adequately distinguish orthogonal transform methods from nonlinear fitting of parametrized time-domain models. To clarify the scope and contribution of the study, I have added the following paragraph at the end of the Introduction on page 3 of the track-change version:

> “In this study, simulated FID signals are analyzed using FFT, STFT, wavelet analysis, and direct nonlinear time-domain fitting to evaluate their complementary roles and limitations. This study does not propose a new Fourier, wavelet, or nonlinear-fitting theory. Instead, it provides a simulation-based evaluation of the complementary roles of these methods for stationary, very short, and non-stationary NMR FIDs. The analysis focuses on dynamically broadened signals, the approximation of finite collection-window measurements by point sampling, and the potential and limitations of JCFit for estimating parameters from very short, relatively simple FID segments.”

I thank the reviewer for this candid and technically important critique. I agree that the original manuscript used the term "least-squares fitting" too broadly and did not adequately distinguish orthogonal transform methods from nonlinear fitting of parametrized time-domain models.

I have revised the manuscript to clarify that the discrete Fourier transform can be interpreted mathematically as an orthogonal least-squares projection of a discrete signal onto the Fourier basis, while the FFT is an efficient computational algorithm for obtaining the DFT coefficients. Similarly, an orthogonal discrete wavelet transform can be interpreted as a projection onto an orthogonal wavelet basis. I have replaced broad or potentially ambiguous references to "least-squares fitting" with "direct nonlinear time-domain fitting" where the distinction between the analysis methods is important.

I also agree that fitting a sum of exponentially damped sinusoids is fundamentally different from projection onto an orthogonal basis. The damped sinusoidal components are generally non-orthogonal, and an overparameterized model may produce non-unique or poorly conditioned solutions. The fitted results can depend strongly on the assumed number of components, parameter constraints, initial estimates, signal-to-noise ratio, and the fitting algorithm. Regularization, statistical model selection, or prior chemical information may therefore be required for complex signals. I have added this limitation to the revised discussion and summarized the associated model-dependence in the conclusion. The revised manuscript (track-change version) now states on page 10:

> "Direct nonlinear time-domain fitting of parametrized damped sinusoidal models may provide complementary estimates of frequencies, amplitudes, phases, and decay constants for very short and relatively simple FID signals. However, the fitted solution may be non-unique or poorly conditioned when the model contains many overlapping or weakly constrained components. Its reliability therefore depends strongly on the assumed number of signal components, the model form, parameter constraints, initial estimates, signal-to-noise ratio, and, where appropriate, regularization or model-selection criteria."

I appreciate the reviewer's additional point regarding wavelet symmetry. Orthogonal wavelet bases generally cannot be exactly symmetric, and this can limit their suitability for directly reproducing symmetric NMR line shapes. In the present manuscript, the wavelet transform is not intended as a line-shape-fitting method. It is used only to generate a time–frequency representation of non-stationary FID behavior. I have revised the manuscript to make this purpose and limitation explicit on page 11:

> “The Gaussian-windowed wavelet analysis used here provides a time–frequency representation of the FID and is not intended as an orthogonal basis expansion or as a method for fitting symmetric NMR line shapes. The resulting spectrogram should therefore not be interpreted as an exact reconstruction of the underlying spectral line shape.”

I have also moderated the discussion of short signals. A finite observation window intrinsically broadens Fourier-domain features, and zero filling interpolates the discrete spectrum but does not add experimental information or restore the missing resolution. Direct parametric fitting does not recover additional information beyond that contained in the measured FID. Rather, it imposes a model and may sometimes estimate signal parameters from a sub-cycle segment when the model form and number of components are sufficiently constrained. I have revised the text accordingly on page 10:

> “When the observation window contains only a partial oscillation cycle, the corresponding Fourier-domain feature becomes broad and frequency estimation becomes poorly conditioned. Zero filling improves spectral interpolation but does not recover the resolution lost through short acquisition. Parametric time-domain fitting may nevertheless estimate a frequency under sufficiently accurate model assumptions and prior constraints, although such estimates are strongly model-dependent. For this simple, model-matched simulation, direct fitting produced more accurate parameter estimates than Fourier-domain peak estimation for most isolated components.”
>
> And
>
> “This result does not establish general superiority because the fitting performance depends strongly on model accuracy, signal complexity, spectral overlap, noise, and prior constraints. Direct nonlinear time-domain fitting of parametrized damped sinusoidal models may provide complementary estimates of frequencies, amplitudes, phases, and decay constants for very short and relatively simple FID signals. However, the fitted solution may be non-unique or poorly conditioned when the model contains many overlapping or weakly constrained components. Its reliability therefore depends strongly on the assumed number of signal components, the model form, parameter constraints, initial estimates, signal-to-noise ratio, and, where appropriate, regularization or model-selection criteria.”

Finally, I have revised the title, abstract, discussion, and conclusion to avoid presenting Fourier transformation, wavelet analysis, and nonlinear fitting as mathematically equivalent alternatives. The manuscript does not claim novelty in Fourier theory, wavelet theory, or damped-sinusoid fitting. Its more limited contribution is a simulation-based evaluation of their complementary practical roles for stationary, very short, and non-stationary NMR FIDs. Specifically, the revised manuscript examines dynamically broadened FID simulations, evaluates the approximation of finite collection-window signals by point sampling after amplitude and phase correction, and illustrates both the potential and limitations of the previously developed JCFit algorithm for fitting very short, relatively simple FID segments. To reflect this narrower scope, the title has been revised to:

> **"Simulating the Extraction of Signal Parameters and Spectrograms for Non-Stationary NMR Signals."**