# Deconvolution of Noisy Tracer Response Data by a Linear Filtering Method

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Deconvolution is used in a variety of areas to extract the impulse response $e(t)$ of a linear system from experimental measurements of the system output $y(t)$ and input excitation function $x(t)$. Some specific examples where deconvolution is important include: analysis of fluorescence decay data (Jakeman et al., 1978a); evaluation of transport functionals in the human circulatory system (Jakeman et al., 1978b; Jordinson et al., 1976); determination of transport properties of the phloem in plants (Minchin, 1978); interpretation of hydrological response data (Young, 1978); refinement and reconstruction of absorption spectra (Blass and Halsey, 1981); process control (Young et al., 1973); and impulse response estimation from chemical reactor tracer response data (Wen and Fan, 1975). A knowledge of the impulse response $e(t)$ is a necessary first step for these and other related applications where model discrimination and parameter estimation are key objectives.

For linear systems, the impulse response $e(t)$ is related to the system input excitation function $x(t)$ and system output $y(t)$ by the convolution integral

$$y(t) = \int_0^t e(\tau) \times (t - \tau) d\tau + n(t) \tag{1}$$

Random noise and measurement errors that are present in the experimental response data are represented by the unknown noise function $n(t)$. As applied to tracer testing, $x(t)$ and $y(t)$ may represent the responses of two sensors located in a flow process using the configuration shown in Figure 1 of Van Zee et al. (1987). Also, $x(t)$ could be the response of the combined tracer injection-sampling system when the test section of a flow process is removed, while $y(t)$ would be the response obtained when the test section is inserted in place (cf., Mills and Duduković, 1981; Wakao et al., 1980).

The common approach for obtaining the impulse response from Eq. 1 is to use Fourier transforms. Applications of this method to Eq. 1 yields the following estimate of the Fourier transformed impulse response

$$\hat{E}(j\omega) = Y(j\omega)/X(j\omega) = E(j\omega) + N(j\omega)/X(j\omega) \tag{2}$$

where $j = \sqrt{-1}$ and $\omega$ denotes the frequency. Applying inverse Fourier transformation to Eq. 2, or so-called inverse filtering (Blass and Halsey, 1981), yields the following estimate of the time-domain impulse response where $F^{-1}$ denotes the inverse Fourier transform operator

$$\hat{e}(t) = F^{-1}\{\hat{E}(j\omega)\} \tag{3}$$

Expressions for the Fourier transform and its inverse operator are given in standard texts (cf., Churchill, 1972). In practice, evaluation of the Fourier transform and the inverse Fourier transform is often performed by numerical quadrature (Davis and Rabinowitz, 1975).

When random noise and measurement errors in the data are sufficiently small, the quotient $Y(j\omega)/X(j\omega)$ is well-behaved so that a reliable estimate of the time-domain impulse response $\hat{e}(t)$ can be obtained. However, for many experimental systems, the errors in $\hat{E}(j\omega)$ defined by $N(j\omega)/X(j\omega)$ either rapidly increase with increasing values of the frequency $\omega$, or randomly vary in such a way that the estimate $\hat{e}(t)$ is obscured by noise. One objective of this paper is to briefly illustrate this observation using actual data collected in our laboratory from trickle-bed reactor response measurements. A second objective is to demonstrate the use of an efficient method which filters the noise so that smooth solutions for $\hat{E}(j\omega)$ and $\hat{e}(t)$ can be obtained. While the current application corresponds to system identification, the method given here can also be applied to the input identification problem for linear systems also.

## Inverse Linear Filter Method

A variety of methods have been proposed for the solution of Eq. 1 for the impulse response $e(t)$ when the function $x(t)$ and $y(t)$ are described by experimental measurements, or are presented as closed-form functions. Some of these methods are described in several excellent monographs (Baker, 1977; Blass and Halsey, 1981; Jansson, 1984) to which the reader is referred for details. The one adopted here is based upon a particular method of regularization first developed by Phillips (1962) and

Twomey (1965) which was later cast into a form more suitable for rapid computations using the fast Fourier transform by Hunt (1970, 1971). It belongs to the class of inverse linear filter methods that are described by Jansson (1984). According to this method, the approximate impulse response in the Fourier transform domain at a discrete sample point $n$ is given by

$$\hat{E}(n/NT) = \frac{Y(n/NT)X^*(n/NT)}{X(n/NT)X^*(n/NT) + \gamma C(n/NT)C^*(n/NT)}; \quad n = 0,1,\ldots,N-1 \qquad (4)$$

In the above equation, $X(n/NT)$ denotes the discrete Fourier transform (DFT) of the samples of $x(t) = x(kT)$ where $k = 0,1,2,\ldots,N-1$, and $T = N\Delta t$ denotes the time interval over which samples of $x(t)$ are available. The DFT of $x(kT)$ is defined by (Brigham, 1974)

$$X(n/NT) = \sum_{k=0}^{N-1} x(kT)\exp\left(-2\pi j\frac{nk}{N}\right); \quad n = 0,1,\ldots,N-1$$
$$= \text{DFT}\,[x(kT)] \qquad (5)$$

An equivalent expression is used for $Y(n/NT)$. The asterisks in Eq. 4 denote the complex conjugate. The term $\gamma C(n/NT)C^*(n/NT)$ in Eq. 4 is the DFT of a digital filter that removes noise from the Fourier-transformed experimental response data. It is based upon a second-order finite difference smoothing formula that is used as a constraint in the least-squares regularization solution. Details on specification of $c(kT)$ and its mathematical properties are given by Hunt (1971). Once Eq. 4 is evaluated for $n = 0,1,\ldots,N-1$, the time-domain impulse response is obtained using the inverse discrete Fourier transform (Brigham, 1974).

$$\hat{e}(kT) = \frac{1}{N}\sum_{n=0}^{N-1} \hat{E}(n/NT)\exp\left(2\pi j\frac{nk}{N}\right); \quad k = 0,1,\ldots,N-1$$
$$= \text{IDFT}\,[\hat{E}(n/NT)] \qquad (6)$$

Evaluation of both the DFT and IDFT according to Eqs. 5 and 6 can be performed using the fast Fourier transform (FFT) (Brigham, 1974) with standard computer software, such as that given in the IMSL Mathematical Library. The one-sided DFT and IDFT is used here since it is assumed that $x(kT)$, $y(kT)$ and $\hat{e}(kT)$ are zero for $t < 0$ according to the causality conditions.

Particularly noteworthy is that Eq. 4 reduces to the standard result for the discrete deconvolution $\hat{E}(n/NT) = Y(n/NT)/X(n/NT)$ in the limit as $\gamma \rightarrow 0$. This corresponds to the case of an unconstrained solution for the impulse response where random noise and measurement errors are not removed.

## Selection of the Smoothing Parameter $\gamma$

Proper specification of the smoothing parameter $\gamma$ that appears in the digital filter in Eq. 4 is critical since too large a value will result in an oversmoothed solution, while too small a value will produce a solution that contains the ill effects of random noise. The results given here require searching for $\gamma$ until a smooth solution for the impulse response can be obtained that satisfies the following squared relative error criteria.

$$\epsilon_1 \leq \left(\frac{\hat{\mu}_1 - \mu_1}{\mu_1}\right)^2 \qquad (7a)$$

$$\epsilon_2 \leq \left(\frac{\hat{\sigma}^2 - \sigma^2}{\sigma^2}\right)^2 \qquad (7b)$$

The quantity $\mu_1$ in Eq. 7a denotes the first absolute moment of the system impulse response defined as the difference in the first absolute moments of the output and input responses (Levenspiel, 1972).

$$\mu_1 = \mu_{1y} - \mu_{1x} \qquad (8)$$

where

$$\mu_{1x} = \int_0^\infty t x(t)\,dt \qquad (9)$$

and similarly for the system output response $y(t)$. The variance $\sigma^2$ of the system impulse response is defined in terms of the second absolute moment $\mu_2$ according to

$$\sigma^2 = \sigma_y^2 - \sigma_x^2 \qquad (10)$$

where

$$\sigma_x^2 = \mu_{2x} - (\mu_{1x})^2, \quad \sigma_y^2 = \mu_{2y} - (\mu_{1y})^2 \qquad (11a)$$

and

$$\mu_{2x} = \int_0^\infty t^2 x(t)\,dt \qquad (11b)$$

with an analogous expression for $\mu_{2y}$. Evaluation of Eq. 11 was performed using a combination of numerical quadrature for $0 \leq t \leq t^*$, and closed-form integration for $t^* \leq t \leq \infty$ as described elsewhere (Mills and Duduković, 1981).

The moment quantities $\hat{\mu}_1$ and $\hat{\sigma}^2$ in Eqs. 7a and 7b have the same meaning as those given above in Eqs. 8 and 10 except they are calculated directly by using the system impuse response $\hat{e}(t)$ obtained by deconvolution using the following expressions:

$$\hat{\mu}_1 = \int_0^\infty t\hat{e}(t)\,dt \qquad (12)$$

$$\hat{\sigma}^2 = \hat{\mu}_2 - (\hat{\mu}_1)^2 \qquad (13)$$

where

$$\hat{\mu}_2 = \int_0^\infty t^2\hat{e}(t)\,dt \qquad (14)$$

For a given value of the smoothing parameter $\gamma$, the discrete approximation of $\hat{e}(t)$ is obtained from Eq. 6 as $\hat{e}(kT)$ for $k = 0, 1, \ldots, N-1$. With $\hat{e}(kT)$ available, the moments defined by Eqs. 12 and 14 can be calculated using a selected quadrature method, such as the trapezoidal or Simpson's rule. The moments are restricted to second order due to the larger errors associated with higher order moments. The zeroth moment is excluded here since our experience has shown that it is insensitive to the smoothing parameter $\gamma$ even when $\gamma$ is varied over several orders of magnitude.

## Statistical Properties of the Estimated Responses

The digital filtering method described above produces an estimate of the system impulse response in both the Fourier transform and time domain with certain statistical properties. Both the derivation of key statistical parameters and an explanation of their significance from both a practical and theoretical perspective has been given by Hunt (1971) so that these will not be repeated here. Evaluation of selected parameters, such as the mean square error, the variance, and the bias, can be performed using the expressions given by Hunt (1971). This evaluation would typically not be required for this application and other related ones since the expected impulse response is a smooth function that does not contain any periodic behavior or discontinuities. For this type of response, it is known that digital filtering will produce estimates having a reduced variance except that biasing will be introduced. Fortunately, the amount of bias is negligible as proved by both inspection of the theoretical expression for the bias given by Hunt (1971), and by numerical evaluation of this expression using the data and results given in the next section. In more advanced applications where the general shape of the impulse response cannot be predicted beforehand, evaluation of these and other statistical parameters may be useful in assessing the quality of the estimated responses.

## Results and Discussion

Figure 1 shows a typical set of normalized input-output tracer response data measured in our laboratory using a trickle-bed reactor packed with porous $\gamma$-alumina catalyst support. Helium and hexane were used as the carrier gas and liquid, respectively, while pentane was found in previous liquid-full experiments to be a suitable nonadsorbing tracer. Additional details on the experimental apparatus and methodology have been given in a previous publication (Mills and Duduković, 1981; Ramachandran et al., 1986). The normalized data are very smooth and show no obvious effects of noise or measurement errors. This suggests that the conventional approach for determination of the impulse response according to Eqs. 2 and 3 should yield smooth solutions in both the Fourier transform domain and the time domain.

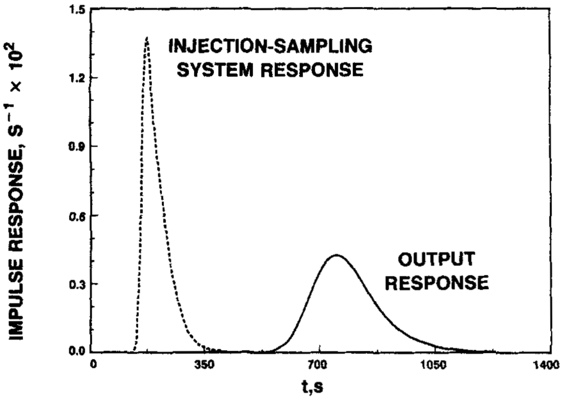


**Figure 1. Input-output nonadsorbing tracer response data for the laboratory-scale trickle-bed reactor system using porous packing.**

Parameters: $d_p = 7.18 \times 10^{-4}$ m, $d_t = 1.35 \times 10^{-2}$ m, $L_p = 0.4$ m, $L_m = 0.15$ kg/m$^2 \cdot$ s, $G_m = 0.12$ kg/m$^2 \cdot$ s, $T = 295$ K; liquid phase = hexane, gas phase = helium, tracer = pentane.

Figure 2 gives the reactor impulse response in the Fourier transform domain (Figure 2a) and the time-domain (Figure 2b) when the deconvolution is attempted without filtering, i.e., $\gamma = 0$ in Eq. 4. Both of these responses are dominated by noise over the entire range of frequency and time at which a solution is expected. Evaluation of the numerical Fourier transforms using Filon quadrature (Davis and Rabinowitz, 1975) in separate numerical experiments yielded similar results. This suggests that the noise was not artifically introduced as a result of the larger truncation errors associated with the discrete Fourier transform by Eq. 5, which is based upon trapezoidal quadrature. Graphical examination of the discrete Fourier transforms of the input-output tracer responses obtained by either method, namely, $X(n/NT)$ and $Y(n/NT)$, shows that the real and imaginary parts are smooth, well-behaved functions. In addition, they have a damped, oscillatory behavior which approach a limiting value of 0 as $\omega \rightarrow \infty$ as expected for pulse response systems. Even so, the impulse response in the Fourier transform domain obtained by division of these curves randomly oscillates with increasing frequency. Thus, the simple deconvolution method of dividing numerical Fourier transforms fails to produce the impulse response in this instance. When the experimental input and output data were smoothed using cubic splines, the resulting impulse responses were similar to the ones shown in Figure 2. This approach apparently fails to remove the required amount

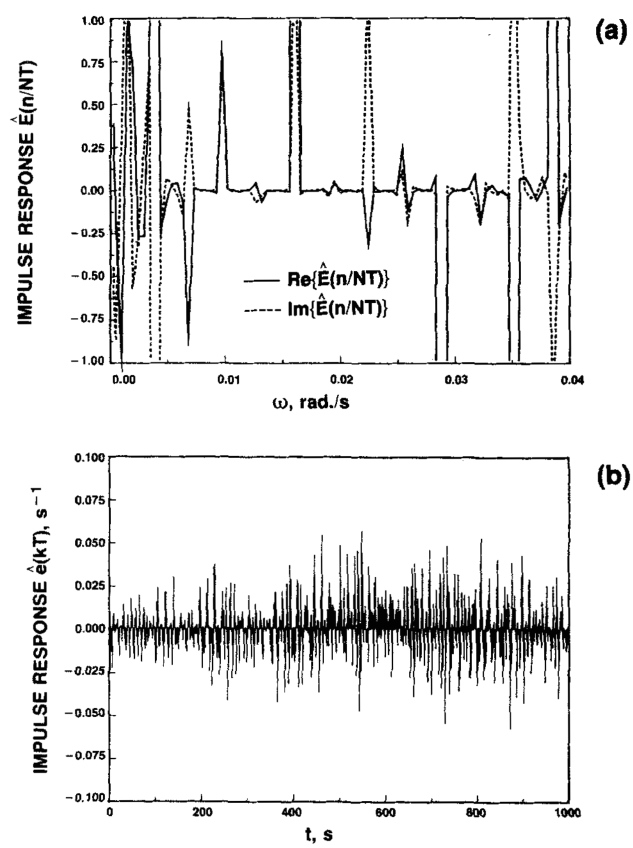


Figure 2a: Fourier-transform domain impulse response $\hat{E}(n/NT)$.
Figure 2b: Time-domain impulse response $\hat{e}(kT)$.

**Figure 2. Deconvolution of the input-output tracer response data using the conventional approach without filtering.**

Parameters: $\Delta t = 1.0$, $N = 2{,}048$, $\gamma = 0$.

of noise from the data. To obtain acceptable results, digital filtering to remove the ill-effects of noise is essential.

The results shown in Figure 2 can be readily explained by referring to Eq. 2. The estimate of the Fourier-transformed impulse response $\hat{E}(j\omega)$ is defined as the sum of the noise-free response $E(j\omega)$ and the contribution due to noise $N(j\omega)/X(j\omega)$. The Fourier-transformed input response $X(j\omega)$ is a damped oscillatory function that approaches a limiting value of zero as the frequency $\omega$ approaches infinity. The Fourier transform $N(j\omega)$ of the noise function $n(t)$ has no predicable dependence on the frequency $\omega$. It represents random errors that are introduced during the experimental measurements of $x(t)$ and $y(t)$, as well as random background noise associated with the tracer detector during normal operation. These errors do not approach a limiting value of zero with increasing frequency $\omega$ so that the quotient $N(j\omega)/X(j\omega)$ represents the dominant term when compared to $E(j\omega)$. A generalized method for a priori estimation of $n(t)$ is not available, so that graphical inspection of the impulse response obtained without digital filtering ($\gamma = 0$) is suggested as one method for determining whether or not filtering is required.

Figure 3 shows the impulse responses in the Fourier transform domain (Figure 3a) and the time domain (Figure 3b) obtained from Eqs. 4 and 6 using the proposed deconvolution method. When compared to the corresponding impulse re-

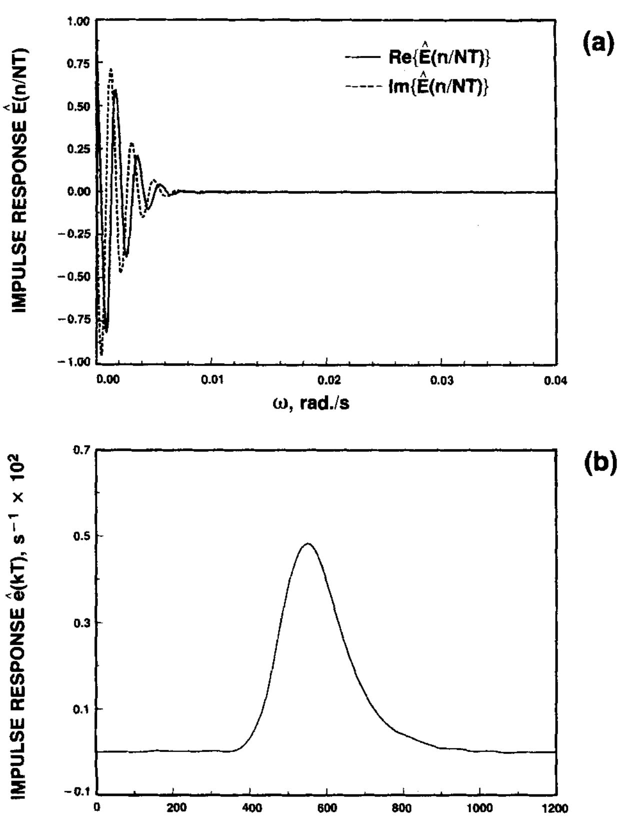


Figure 3a: Fourier-transform domain impulse response $\hat{E}(n/NT)$.
Figure 3b: Time-domain impulse response $\hat{e}(kT)$.

**Figure 3. Deconvolution of the input-output tracer response data using the proposed filtering method.**

Parameters: $\Delta t = 1.0$, $N = 2048$, $\gamma = 10^3$.

**Table 1. Comparison of Reactor Impulse Response Curve Moments Obtained by Two Methods**

| Moment Value | Method 1* $\mu_1, \sigma^2$ | Method 2** $\hat{\mu}_1, \hat{\sigma}^2$ | Error† |
|---|---|---|---|
| $\mu_1$, s | 582.8 | 579.1 | 0.64 |
| $\sigma^2$, $s^2$ | 10,275 | 10,921 | 6.28 |

*Based on the differences between the moments of the output-input curves according to Eqs. 8 and 9.

**Based on Eqs. 12–14 where the reactor impulse response is obtained by deconvolution with $\gamma = 1.0 \times 10^3$.

†Defined as $100|(\mu_1 - \hat{\mu}_1)/\mu_1|$ and $100|(\sigma^2 - \hat{\sigma}^2)/\sigma^2|$ where ^ denotes the Method 2 value.

sponses in Figure 2, the results are quite dramatic since smooth curves have emerged from what otherwise appeared to be random noise.

As mentioned above, the moments of the reactor impulse response curve shown in Figure 3b, and defined according to Eqs. 12–14, should be in reasonable agreement with those obtained by differences in moments of the output-input tracer response curves if the smoothing parameter $\gamma$ has been properly selected. Table 1 compares the first absolute moment $\mu_1$ and the variance $\sigma^2 = \mu_2 - (\mu_1)^2$ of the reactor impulse response obtained by these two methods. Values for $\mu_1$ and $\sigma^2$ of the reactor impulse response obtained by deconvolution have less than 1 and 7% relative error, respectively, which is quite acceptable.

Figure 4 shows the dependence of the square of the relative errors defined by Eqs. 7a and 7b on the smoothing parameter $\gamma$. The one based upon the first moment $\epsilon_1$ (Figure 4a) is monotonic over values of $\gamma$ spanning more than three orders-of-magnitude

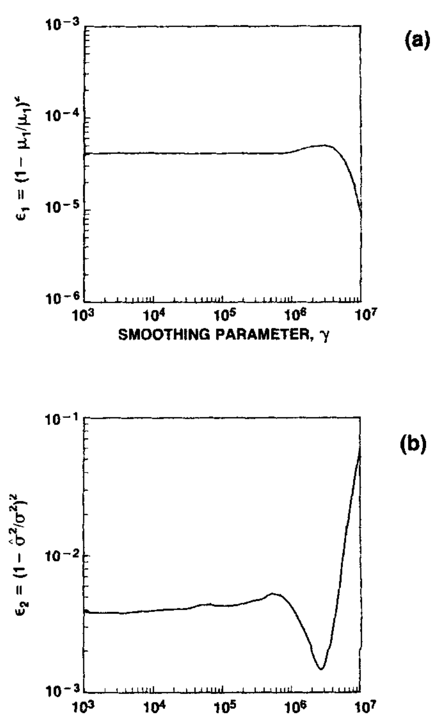


**Figure 4. Effect of the smoothing parameter $\gamma$ on the squared relative error in the first absolute moment (Figure 4a) and the variance (Figure 4b) of the impulse response.**

until $\gamma \rightarrow 10^6$ where it increases and then decreases. The squared relative error of variances defined by Eq. 7b is also relatively monotonic until $\gamma \rightarrow 5 \times 10^5$ where it increases, decreases to a local minimum at $\gamma = 2.6 \times 10^6$, and then increases again. Taken collectively, these results suggest an upper bound of $\gamma \simeq 5 \times 10^5$, with smaller values being acceptable as long as the resulting impulse responses appear reasonably smooth. Due to the ill-posed nature of solving Volterra convolution integral equations with noisy data (Baker, 1977), more than one value of the smoothing parameter $\gamma$ may yield acceptable results. The results given in Figure 3 are based upon $\gamma = 10^3$ which produces smooth solutions for the impulse responses in the Fourier transform and time domain. Development of suitable flow models and parameter estimation using these impulse responses as the basis represents one possible application of the results.


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## Notation

$c(kT)$ = discrete form of the smoothing function
$C(n/NT)$ = discrete form of the Fourier transformed smoothing function
$\hat{e}(kT)$ = discrete form of the estimated time-domain impulse response, $s^{-1}$
$e(t)$ = continuous form of the exact time-domain impulse response, $s^{-1}$
$\hat{e}(t)$ = continuous form of the estimated time-domain impulse response, $s^{-1}$
$\hat{E}(n/NT)$ = discrete form of the estimated Fourier-transformed impulse response
$E(j\omega)$ = continuous form of the exact Fourier-transformed impulse response
$j = \sqrt{-1}$
$k$ = summation index
$n(t)$ = continuous form of the time-domain noise function
$N$ = total number of points
$N(j\omega)$ = continuous form of the Fourier-transformed noise function
$t$ = time, s
$T$ = time measuring period, s
$x(t)$ = continuous form of the normalized system input response, $s^{-1}$
$X(n/NT)$ = discrete form of the Fourier-transformed system input response
$X(j\omega)$ = continuous form of the Fourier-transformed system input response
$y(t)$ = continuous form of the normalized system output response, $s^{-1}$
$Y(n/NT)$ = discrete form of the Fourier-transform system output system response
$Y(j\omega)$ = continuous form of the Fourier-transform system output response

### *Greek Letters*

$\epsilon_1$ = squared relative error based upon first absolute moments of the system impulse response
$\epsilon_2$ = squared relative error based upon the variances of the system impulse response
$\mu_1$ = first absolute moment of the system impulse response obtained from the input-output responses, s
$\hat{\mu}_1$ = first absolute moment of the system impulse response obtained by deconvolution of the input-output responses, s
$\sigma^2$ = variance of the system impulse response obtained from the input-output responses, $s^2$
$\hat{\sigma}^2$ = variance of the system impulse response obtained by deconvolution of the input-output responses, $s^2$

### *Subscripts*

$x$ = refers to the system input response
$y$ = refers to the system output response

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