Minimizing the Effect of Extraneous Spikes in Open-Path Fourier Transform Infrared Interferograms

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Intense spikes caused by extraneous factors are sometimes found in the interferograms of open-path Fourier transform infrared (OP/FT-IR) measurements. Those spikes result in dominant oscillations in the corresponding spectra and make the true spectral features indistinguishable from the noise. Three techniques were designed to remove the spikes: replacing the affected region by zeroes; grafting the data from the corresponding region on the other side of the centerburst ("homografting"); and grafting the data from the same region on the same side of the centerburst from a neighboring interferogram ("heterografting"). Results showed that all three techniques were effective to remove the spikes, but at the cost of introducing more noise into the corresponding spectrum. The performance of the heterograft technique was the best in terms of noise reduction. The factors affecting the noise level of spectra computed from spike-removed interferograms were also explored. A procedure was proposed to minimize the effect of spikes on OP/FT-IR spectra, and a Matlab program with a graphical user interface is available upon request to facilitate the implementation.

Index Headings: **Open-path FT-IR spectrometry; Fourier transform infrared spectroscopy; Interferogram; Interference; Spike removal; Grafting.**

INTRODUCTION

Open-path Fourier transform infrared (OP/FT-IR) spectrometry is a reliable and effective technique to measure the concentration of small molecules in the atmosphere.[1] The open-path mode has several advantages over laboratory-based measurements, such as convenient operation and exclusion of the sampling step; however, the measurement itself is more susceptible to interference. We have previously investigated five types of interference found in OP/FT-IR interferograms and developed a procedure to reject the corrupted spectra from further analysis that would give erroneous results.[2] In one type of interference, a spike is introduced into the interferogram that gives rise to a sinusoidal interference in the spectrum. In field measurements near agricultural facilities, the spike could be caused by a bird flying through the beam. We have also taken OP/FT-IR measurements under simulated conditions of "battlefield clutter" where incandescent sparks pass rapidly into the beam, leading to sharp spikes in the interferogram.

After Fourier transform of the interferogram, the spike introduces a decaying oscillation into the corresponding spectrum that masks the weaker spectral bands. The decaying oscillation is not necessarily detrimental to spectral analysis, since its profile is determined by the shape of the spike.[3] The amplitude of the oscillation is always greatest at 0 cm^{-1}. If the spike is broad enough, the decay of the corresponding oscillation may be essentially complete by the cut-off frequency of the detector. For OP/FT-IR measurements, the detector is usually mercury cadmium telluride (MCT) with a cut-off frequency of ~700 cm^{-1} so that the effect of broad spikes is usually negligible. However, for a sharp spike, the oscillation extends well beyond 700 cm^{-1} in the corresponding spectrum, making the spectral features of the molecules of interest indistinguishable from the noise.

Although the rejection of interferograms corrupted with sharp spikes could prevent erroneous results,[2] it is at the cost of the information lost at the times at which those rejected interferograms were measured. In practice, even though the rejected spectrum shows high-amplitude oscillations due to the spike(s), the spectral information of the molecules of interest can be retrieved by processing the interferogram rather than the spectrum. The retrieval is feasible because the spike is localized in the affected interferogram, leaving the remaining data points unaffected. Therefore, by removing the spikes from the interferogram by some means, the oscillations in the corresponding spectrum would be reduced significantly, so that the useful spectral information is revealed. In this investigation, we designed three techniques for spike removal, evaluated the performance of each, and finally proposed a procedure to deal with such interferograms.

Interference by spikes is also found in Raman, magnetic resonance, and far infrared and millimeter wave spectra, and several methods had been developed to correct the corrupted data points.[4–6] The basic idea of these correction methods is straightforward. For several methods, the values of the corrupted data points are estimated through interpolation of neighboring data points and then these values replace the corrupted data points.[4] In other approaches, the location and intensity of the spike are obtained by certain means such as Fourier transform[5] or wavelet transform,[6] and the intensity of the spike is then subtracted from the affected region. While these methods are effective in certain cases, there exists a restriction on the number of corrupted data points. For instance, one method requires that the spike occupy no more than two adjacent data channels;[4] in another method the corrupted data points need to be sufficiently distant from each other.[5] Because of such restrictions, previous methods are inapplicable to the spike removal in our investigation in which each spike usually affects the interferogram over a range of more than 100 data points.

Figure 1 shows two interferograms measured consecutively with the emission from a type of firework in the infrared beam; the region of the interferogram that was affected by one spike is shown enlarged in the inset. The difference between the two interferograms in the inset indicates that the effect of the spike is significant over a range of about 100 data points (from 5850 to 5950) and decreases until it cannot be distinguished from an unaffected interferogram only outside a broader range of about

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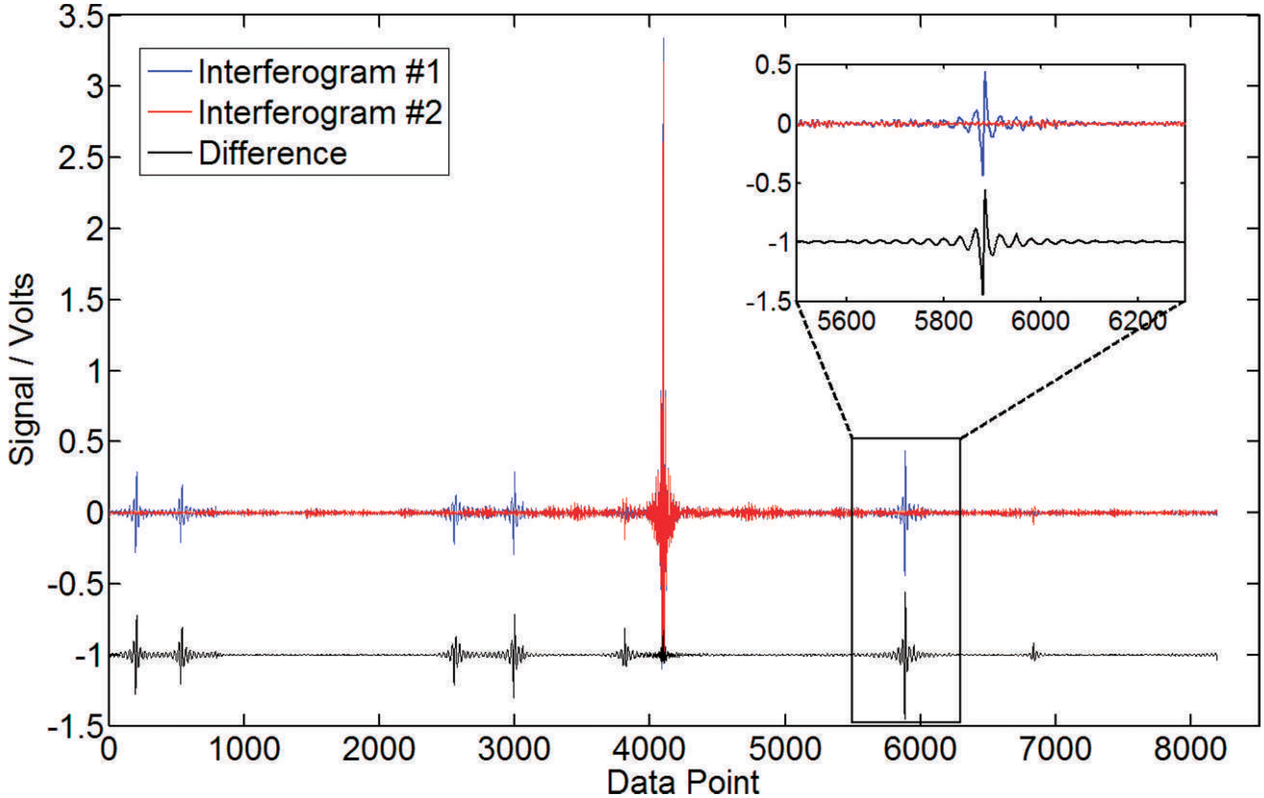

FIG. 1. Two interferograms measured consecutively in the presence of fireworks, and the difference, which is displaced for clarity. The inset shows an enlargement of one spike. The two interferograms were processed with high-pass filtering as described in the paper.

200 points (from 5800 to 6000). With such a large number of corrupted data points, it is unreliable to predict the true values from the neighboring uncorrupted data points. In this case, the corrupted data points must be replaced in a way that is different from previous methods.

MEASUREMENT AND DATA PROCESSING

Experimental. The interferograms were acquired in an experiment to test the applicability of OP/FT-IR spectrometry to the detection of chemical warfare agents under conditions of "battlefield clutter" (smoke, explosions, sparks, etc.).[7] The interferograms were measured at a resolution of 4 cm^{-1} with a Bomem OP/FT-IR spectrometer equipped with a Globar® source and an MB-100 interferometer. A cube-corner-array retroreflector was positioned 27 meters away from the spectrometer to reflect the IR beam back to the MCT detector, which is mounted in the same box as the interferometer. Each measurement resulted from co-adding four interferograms. To simulate battlefield conditions, fireworks were ignited to emit colored smoke and a stream of incandescent particles into the infrared beam of the OP/FT-IR measurement. It was found that fireworks that emitted "crackling" incandescent particles gave rise to intense, usually sharp, spikes in the interferogram. In one series of measurements described in Ref. 7, 32 such interferograms were rejected out of a total of 819. We used these 32 interferograms to test three techniques for spike removal as described below.

Techniques for Spike Removal. If all the spikes in a given double-sided interferogram happen to occur on one side of the centerburst, their effects on the spectrum can be readily eliminated by only using the data on the unaffected side to compute the single-beam spectrum. The downsides are (a) the decrease in the signal-to-noise ratio (SNR) of the computed spectrum by a factor of $\sqrt{2}$ and (b) the fact that all interferograms must be treated in the same way. If the spikes are located on both sides of the centerburst, they must be removed by some other means.

We investigated three techniques to remove the extraneous spikes. The first one, the zero-replacement technique, is to replace all data points in the region of the spike with zeros. The second one, which we call the "homograft" technique, involves replacing every data point of the spike with the corresponding point on the other side of the centerburst; this technique requires that the region used as the graft be unaffected by another spike. The third one, which we call the "heterograft" technique, involves replacing every data point of the spike with the point at the same retardation *and* on the same side of the centerburst of a different interferogram measured shortly before or afterwards. For example, this "donor" interferogram could be the one previous or subsequent to the "acceptor" interferogram (as they were in our measurements). The heterograft technique is somewhat analogous to the method to remove the effect of interference fringes from FT-IR spectra that was reported by Hirschfeld and Mantz.[8]

Each of the three techniques was effective in removing the spike, but differs in its effect on the SNR of the corresponding spectrum. The zero-replacement method works in all cases in which it is applicable, but the SNR is decreased because real spectroscopic information is lost. The homograft technique only functions when the "graft" region on the other side of the centerburst is free of spikes; furthermore, this operation

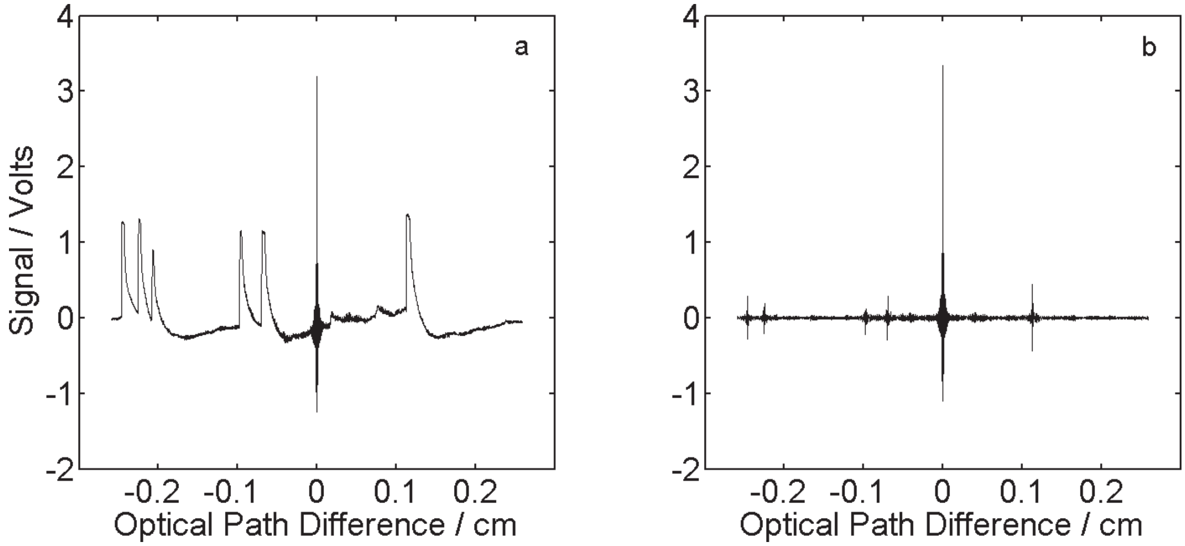

FIG. 2. A measured interferogram with extraneous sharp spikes (**a**) before and (**b**) after high-pass filtering.

introduces phase errors unless the interferogram is perfectly symmetrical. The heterograft technique works in practically all cases when sampling is triggered at a zero-crossing of the laser interferogram, since the chance of encountering a spike at the same retardation of a neighboring interferogram is very slim. Moreover, if data acquisition is started at the same retardation for all interferograms, the "donor" and "acceptor" interferograms should have the same phase spectrum at all retardations; thus, any effect of phase error should be minimal. It should be noted that for FT-IR spectrometers where the interferogram is digitized by a sigma-delta analog-to-digital converter, data acquisition is not started at the same retardation for all interferograms so the heterograft technique would not be applicable. Even though the heterograft technique brings foreign information to the processed interferogram, it is not a serious problem since interferograms usually change very little under rapid-scan conditions.

High-Pass Filtering. The modulation frequency, $f_{\tilde{\nu}}$ Hz of radiation of wavenumber $\tilde{\nu}$ cm^{-1} is given by the product of $\tilde{\nu}$ and the optical velocity, V cm·s^{-1}.[9] Because of the proportionality of $f_{\tilde{\nu}}$ and $\tilde{\nu}$, any interference of frequency f Hz can be converted to its corresponding wavenumber simply by dividing by V. Any interference with a frequency that, after conversion to wavenumber, is less than the cut-off wavenumber for the MCT detector (700 cm^{-1}) can be eliminated by the application of an appropriate high-pass filter. In this investigation, the raw interferograms were preprocessed by application of a high-pass filter with a cut-off of 480 cm^{-1}. After high-pass filtering, it is possible to determine how seriously the absorbance spectrum in the atmospheric windows is affected. After spike removal, each single-beam spectrum was ratioed to a single-beam spectrum that was measured in the absence of interference and then converted to absorbance. The noise level of the absorbance spectrum was calculated as the root mean square (RMS) of the absorbance values between 2700 and 2500 cm^{-1} (where no molecule in air absorbs strongly).

RESULTS AND DISCUSSION

One interferogram with intense spikes caused by the transient presence of incandescent sparkles is shown in Fig. 2a; Fig. 2b shows the result after high-pass filtering. It can be seen that most of the low-frequency interference is removed by the high-pass filtering but sharp spikes remain.

The single-beam spectrum computed from the affected interferogram shown in Fig. 2a is shown in Fig. 3a; it is clear that this spectrum is so noisy as to be of little analytical value. The single-beam spectrum after application of the heterograft technique to the interferogram shown in Fig. 2b is presented in Fig. 3b. For comparison, a spectrum computed from an interferogram measured in the absence of interference is shown in Fig. 3c. A careful comparison of these spectra shows that the noise is slightly higher in Fig. 3b than in Fig. 3c. All three spike-removal techniques have minimal effect on either the shape of background spectrum or the relatively sharp absorption features of water vapor and CO_2.

The Performance of the Spike-Removal Techniques. After high-pass filtering, all 32 interferograms that were rejected from the series described in Ref. 7 were processed with the three spike-removal techniques. The zero-replacement and heterograft techniques were successfully applied for all 32 interferograms, whereas the homograft technique was successful for only 26 cases because of the presence of spikes on both sides of the centerburst at the same retardation in the other six. In order to evaluate the effect of the spike removal on the corresponding spectrum, we calculated the RMS noise level between 2700 and 2500 cm^{-1} in the absorbance spectra computed from the interferograms after spike removal.

We calculated the RMS noise level of 100 absorbance spectra measured in stable air before the fireworks as a benchmark of the noise level. The RMS values were fairly steady with an average of 8.34×10^{-4} a.u. ($\sigma = 0.57 \times 10^{-4}$), so this value reflects the noise level of our OP/FT-IR spectrometer under stable conditions. From the interferograms processed with the three spike-removal techniques, absorbance spectra were computed and the RMS noise levels were obtained, as shown in Fig. 4. It was found that additional noise was introduced by each technique, with the noise introduced by the heterograft technique being generally slightly lower than that by the homograft technique and varying the least. The averages of the RMS noise levels for the zero-replacement, homograft, and heterograft techniques were

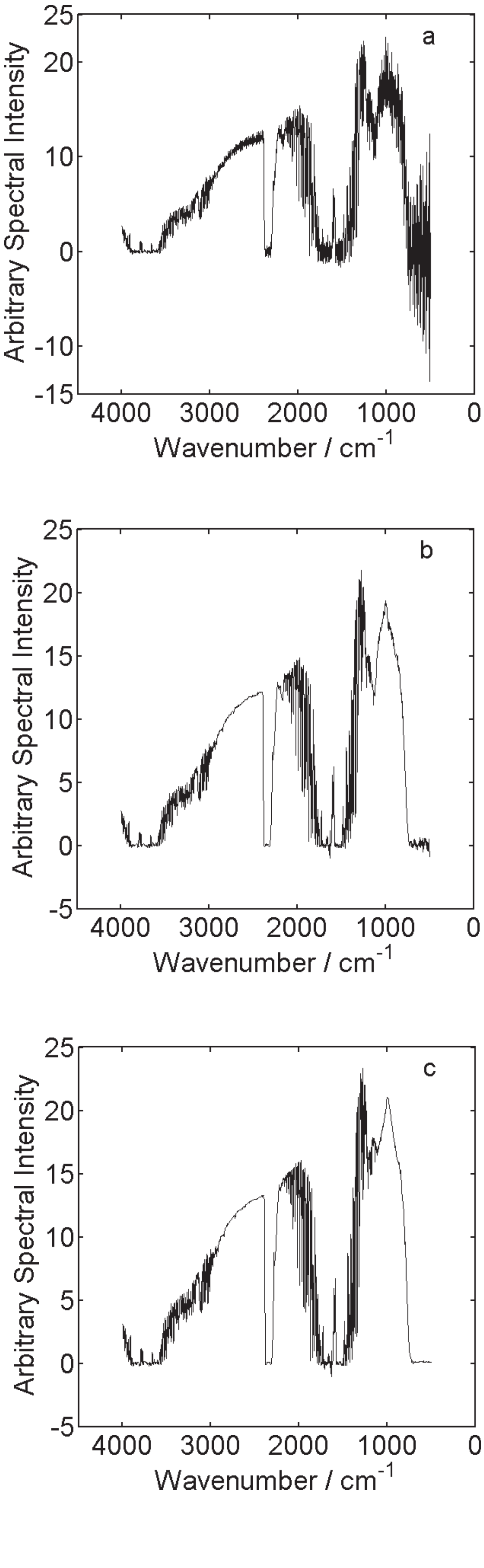

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FIG. 3. Single-beam spectra computed (**a**) from the spike-corrupted interferogram and (**b**) after spike removal with the heterograft technique; (**c**) single-beam spectrum of a measurement in the absence of interference.

calculated to be 41.8×10^{-4} ($\sigma = 29.8 \times 10^{-4}$), 28.5×10^{-4} ($\sigma = 9.56 \times 10^{-4}$), and 25.0×10^{-4} a.u. ($\sigma = 7.10 \times 10^{-4}$), respectively. For four of the spectra that had been processed using the zero-replacement technique, the RMS noise was unusually high, as shown in Fig. 4. In each case, the spike occurred quite close to the centerburst, where the amplitude of the interferogram is higher than at high retardation. Thus, the error introduced by replacing the spike with zeroes is higher than if the spike was at higher retardation. Without the four spectra, the noise level for the zero-replacement technique was 32.2×10^{-4} ($\sigma = 11.1 \times 10^{-4}$). Therefore, when the spike is fairly distant from the centerburst, the performance of the zero-replacement technique is comparable to that of the homograft or the heterograft technique, but when the spike is close to the centerburst its performance degrades more seriously than the other two techniques. This is because the zero-replacement technique changes the amplitude of the interferogram more than the homograft or the heterograft technique does, particularly at low retardation where the amplitude is high.

In summary, in terms of the RMS noise level in the absorbance spectrum, the heterograft and homograft techniques outperform the zero-replacement technique, with the heterograft technique slightly outperforming the homograft technique. However, the noise level is still increased by an average of about a factor of three in comparison to the noise of an unaffected interferogram.

Factors Affecting the Performance of the Spike Removal. Even though the heterograft technique performed the best in spike removal, its performance varies in processing different interferograms, as shown by the RMS noise levels in Fig. 4. It was found that the lowest RMS noise level was 13.0×10^{-4}, which is close to the benchmark of 8.34×10^{-4}, while the highest was about four times higher (38.0×10^{-4}). In order to find the factors that relate to the performance of the heterograft technique, we plotted the RMS noise level versus the optical path difference of the spike that is nearest to the centerburst; the results are shown in Fig. 5. In order to include the effect of another factor into the same figure, we set the size of each dot in Fig. 5 to be proportional to the number of spikes found and removed in the interferogram.

Figure 5 shows that the large dots tend to aggregate in the upper region, while the small ones are in the lower region, i.e., the larger the number of spikes in the interferogram, the higher the noise introduced by the spike removal into the absorbance spectrum. Regardless of the size of the dots, Fig. 5 indicates that the closer the spike is to the centerburst, the higher the noise of the absorbance spectrum.

In addition to the above two factors, another one apparently affecting the noise level is the range of spike, since it determines how many data points are corrupted. It is practically helpful to know which of the three factors plays a dominant role in affecting the noise level. In this respect, we used synthetic interferograms by adding artificial spikes to an interferogram free of any interference, and in those interferograms we changed the location, the range, and the number of the spikes. After spike removal, the RMS noise levels were calculated from corresponding absorbance spectra, and the

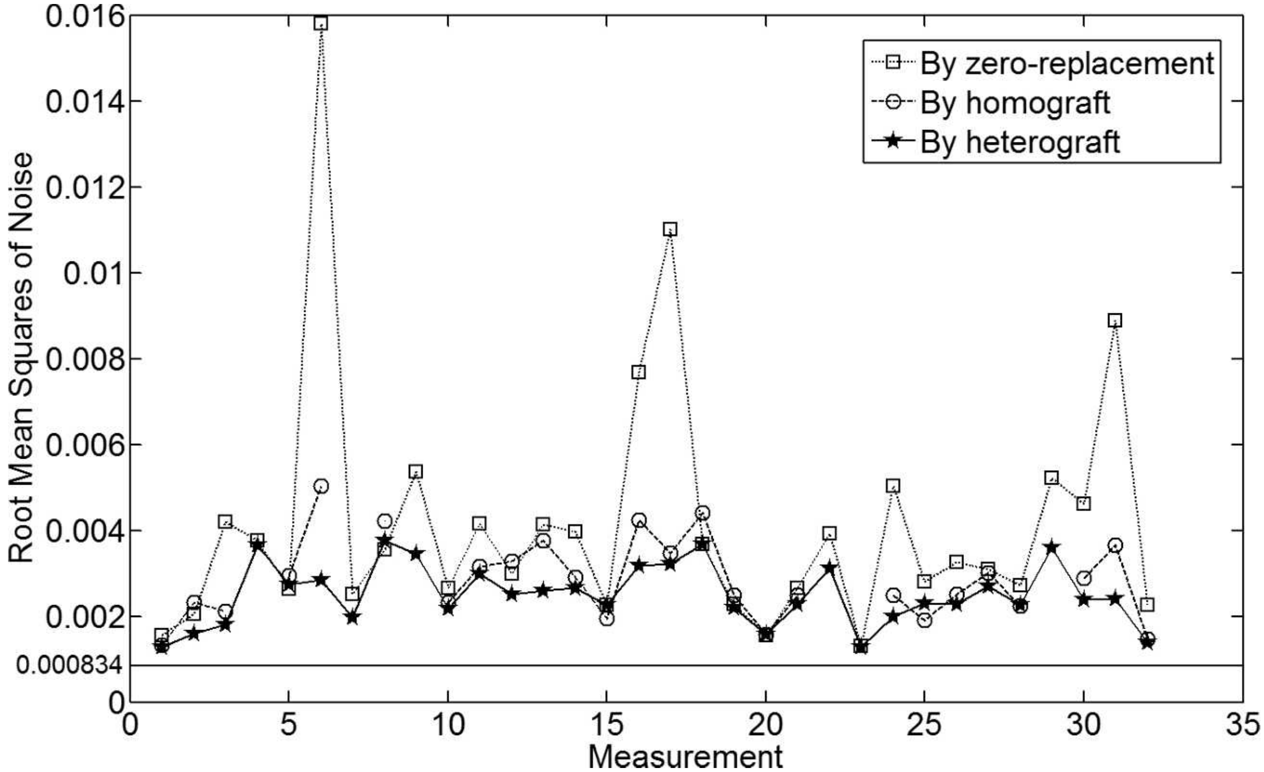

FIG. 4. The RMS noise levels of spectra computed from interferograms processed with three spike-removal techniques. The horizontal line represents the benchmark noise level, which is 8.34×10^{-4} a.u.

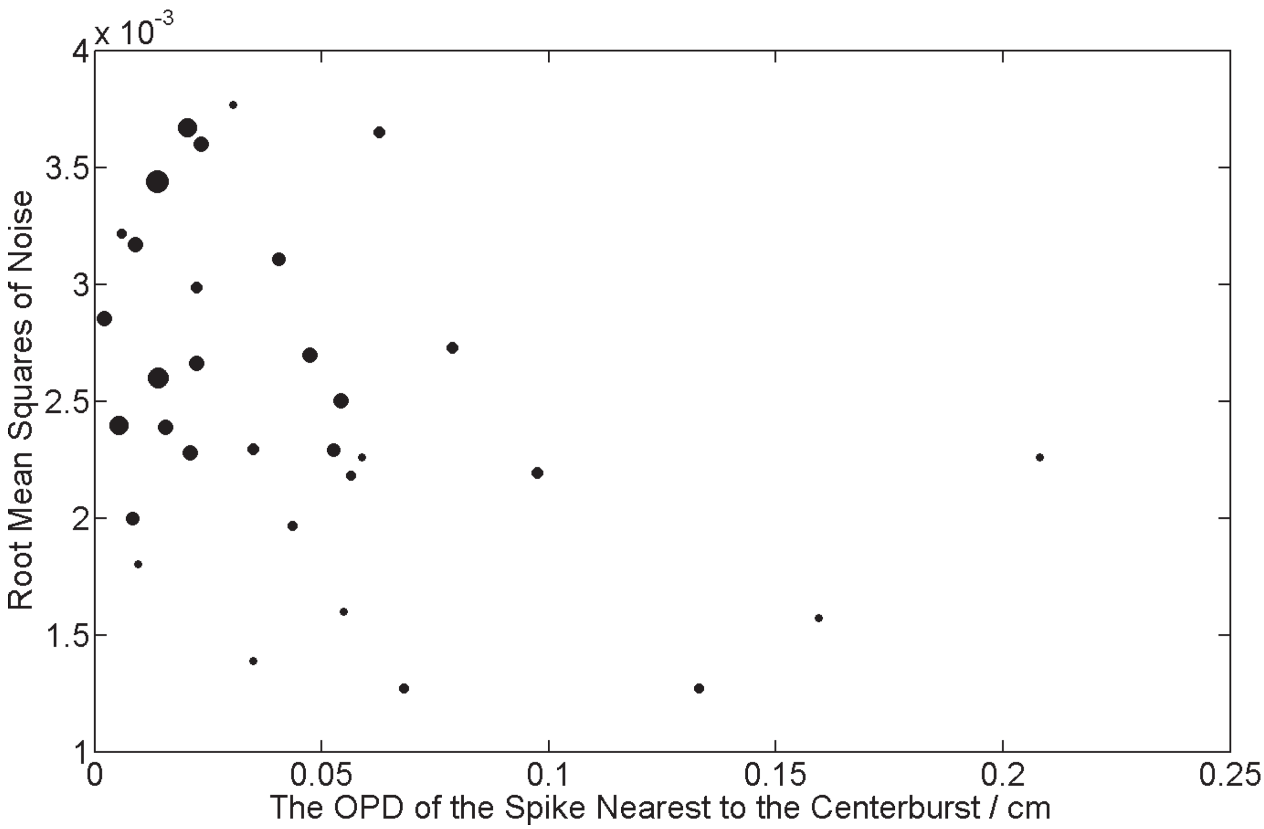

FIG. 5. The RMS noise levels of spectra from the interferograms processed with the heterograft technique. Each dot represents one interferogram, and the size is proportional to the number of spikes removed.

TABLE I. Average RMS noise level of spectra computed from synthetic interferograms after spike removal by three techniques. Artificial spikes of various locations, ranges, and quantities were introduced into those interferograms.

	RMS noise level ($\times 10^{-4}$) due to different factors of spike		
Technique	Location of spike	Range of spike	Number of spikes
Heterograft	8.47	8.64	9.00
Homograft	8.49	9.29	9.35
Zero-replacement	16.0	10.9	12.2

results are listed in Table I. The RMS data in Table I demonstrate that in the heterograft technique, the effect of the number of spikes on the noise level is higher than, but somewhat close to, those of the other two factors, which are the range and the location of spikes. A similar conclusion was drawn regarding the homograft technique. With respect to the zero-replacement technique, the most serious effect on noise level is from the location of the spike, which is expected since the most intense data points are near the centerburst, and it is affected the most by the zero-replacing process. From Table I, it could be found that for each of the three factors, the noise level of the heterograft technique is the lowest and the noise level of the zero-replacement technique is the highest, which is consistent with the result obtained in processing the real data in the previous section.

CONCLUSION

The presence of sharp spikes in an OP/FT-IR interferogram does not mean the measurement is invalid and should be discarded. The true spectral information is often retrievable by the application of a high-pass filter to the interferogram and employing the heterograft technique proposed in this paper to remove the spikes. At this point, the implementation of the spike-removal technique is manual; a Matlab program with a friendly graphical user interface is available upon request.

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