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Optical Feedback Linear Cavity Ringdown Spectroscopy

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Optical feedback cavity ringdown spectroscopy is presented with a linear Fabry–Pérot cavity and a cost-effective DFB laser. To circumvent the low coupling efficiency caused by the broad laser linewidth, an optical feedback technique is used, and an enhanced coupling efficiency of 31%, mainly limited by impedance mismatch and mode mismatch, is obtained. The trigger of the ringdown event is realized by the shutoff of the laser driving current, and a novel method with the aid of one electronic switch is applied to avoid the ringdown events excited by the unexpected cavity modes during the process of laser current recovery. As a result, the ringdown signal with a signal-to-noise ratio of 2500 is achieved. Through continuous monitoring, the fractional uncertainty of the empty cavity ringdown times is assessed to be 0.04%. An Allan variance analysis indicates a detection sensitivity of 4.3×10^{-10} cm^{-1} is resulted at an integration time of 120 s, even with a moderate finesse cavity. To further improve the long-term stability, we regularly rectify the empty cavity ringdown time, and an improvement factor of 2.5 is demonstrated.

Keywords: CRDS, laser absorption spectroscopy, high sensitivity, optical feedback, Fabry–Pérot cavity

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INTRODUCTION

Laser absorption spectroscopy is an effective way for trace gas detection due to its advantages of high sensitivity and high stability. To improve the detection sensitivity, a variety of methods, such as cavity ringdown spectroscopy [1, 2], cavity-enhanced absorption spectroscopy [3, 4], thermoelastic spectroscopy [5, 6], and photoacoustic spectroscopy [7, 8], have been proposed. Cavity ringdown spectroscopy (CRDS) is a well-established spectroscopic technique with the merit of high sensitivity [1, 2]. It leverages an optical cavity to prolong the interaction length between the laser and the intracavity gas, and thus, an amplified absorption signal can be obtained. With superior coating technologies, the amplification factor larger than 10^5 and the detection sensitivity for trace gas detection down to 10^{-13} cm^{-1} could be achieved [9, 10]. On the other hand, CRDS deduces the intracavity absorption information from the variation of the ringdown times, rather than the amplitudes of the cavity transmission modes. Thus, it is immune to laser intensity noise and frequency-to-amplitude noise, which is the main limitation of cavity-enhanced absorption spectroscopy (CEAS). In addition, CRDS is a calibration-free technique that can result in absolute gas concentration. Therefore, in recent decades, CRDS has been widely applied in a variety of application fields [11–13].

To provide regular cavity ringdown events, the laser frequency or cavity length, as well as cavity longitudinal mode frequency, is modulated [14]. However, due to the much broader laser linewidth compared to the cavity mode width, only a small proportion of the light could be coupled into the cavity at a time, leading to a rather low coupling efficiency. Simulation shows that the coupling efficiency could be improved by nearly 100 times with the narrowing of laser linewidth in CRDS [15]. However, diode lasers, the most prevalent light source in the CRDS application field, have a linewidth of several MHz, which is much larger than the cavity linewidth. As a result, a feeble light will be observed in the cavity transmission and the performance of CRDS will be limited by detector noise and electronic noise.

To circumvent this problem, frequency-locked CRDS has been proposed [16]. By locking the laser to the cavity, or vice versa, *via* the Pound–Drever–Hall method, the laser linewidth is narrowed, and accordingly, high coupling efficiency is attained. However, for a diode laser, because of its large and broadband frequency noise, it is difficult to lock it to a high finesse cavity.

Optical feedback is an alternative method, which is exclusively suitable for a diode laser. The leakage of the intracavity light from the cavity front mirror, acting as an external superb light source with narrow linewidth, returns to the laser, and an injection frequency locking of the laser to the cavity is realized. Optical feedback CRDS (OF-CRDS) was first proposed by Morville based on a V-shape cavity [17, 18]. The improvement of the coupling efficiency by a factor of more than 20 times and neat cavity modes have been observed. Following the first implementation, most existing OF-CRDS setups are based on a V-shape cavity [19–22]. This is because this cavity geometry could separate the intracavity leak-out light from the unwanted direct reflection at the cavity front mirror, which are suspected to generate optical feedback competition with each other. Recently, our group has introduced a linear Fabry–Pérot cavity-based optical feedback CEAS (OF-CEAS) without special care of unwanted direct reflection [23]. By this, optical feedback linear cavity-enhanced absorption spectroscopy (OF-LCEAS) has been developed [3]. The intracavity absorption is derived directly from the amplitude attenuation of the cavity transmission modes. Compared to the V-shape cavity, the Fabry–Pérot cavity is more universal and less sensitive to mechanical vibration and could possess higher finesse. Therefore, it improves the applicability of OF-CEAS.

In this study, optical feedback linear cavity ringdown spectroscopy (OF-LCRDS) based on a linear Fabry–Pérot cavity and a cost-effective DFB laser is presented. Due to the advantages of CRDS over CEAS, a better performance is expected. The ringdown event is excited by changing the laser current below the threshold of emitting quickly. A novel strategy to avoid the trigger of unexpected ringdown events during the laser current recovery is provided. To improve the long-term stability, the optical feedback phase is actively controlled and the empty cavity ringdown time is rectified with the ringdown time at the laser frequency far away from the absorption. The experimental details are presented at first. A ringdown signal with high fidelity is shown. Also, the detection sensitivity is evaluated by the Allan variance plot [24]. Finally, a spectrometer is used to detect CH_4 concentration and its long-term stability is examined.

EXPERIMENTAL SETUP

The schematic diagram of the experimental setup for OF-LCRDS is shown in **Figure 1**. A distributed feedback laser diode (Eblana, EP1653-7-DM-TO56-A04) with a TO footprint, emitted at a wavelength of 1.65 μm, is utilized, which addresses three overlapping strong CH_4 transitions around 6046.9 cm^{-1} with a line strength of around 1×10^{-21} cm^{-1}/(molecule × cm^{-2}). The output light passes through, in sequence, a mode-matching lens, a half-wave plate (λ/2), two reflectors, a polarization beam splitter (PBS), and a quarter wave plate (λ/4). By rotating the λ/4, the feedback ratio could be adjusted.

The light is then injected into a Fabry–Pérot (FP) cavity. The cavity consists of two identical mirrors with a reflectivity of 99.96%, corresponding to a finesse of around 7850. The two mirrors are separated with a length of 39.4 cm, implying a free spectral range of 380 MHz. The maximum coupling efficiency between the laser and the cavity is measured to be around 31%, mainly limited by impedance mismatch and mode mismatch, by monitoring the cavity reflection when the laser frequency is locked to the cavity mode [25]. To control the feedback phase to be an integer multiple of 2π for an effective optical feedback [23], two strategies are utilized. A translation stage mounting the laser and a PZT adhered to one of the reflectors are responsible for coarse and fine phase adjustments, respectively. A correction voltage is sent to the PZT to realize active and real-time compensation, which is generated from the judgment of the asymmetry of the cavity transmission mode. To avoid the usage of an extra optical switch which will deteriorate the stability of optical feedback by introducing optical phase shift and fluctuation, the ringdown event is excited by directly and abruptly cutting off the laser current using a homemade electronic switch, i.e., ES_1. The cavity transmission and ringdown signals are captured using a PD (Thorlabs, PDA 10CS-EC) and then sent to another homemade electronic switch, i.e., ES_2, which is used to filter out the unexpected cavity modes (detailed information will be given in *Ringdown Signal*). The output of ES_2 is divided into two parts. One is sent to a data acquisition (DAQ) card, and the other is sent to a pulse generator, where the latter generates two pulse signals with different starting times and duty ratios to control the ES_1 and ES_2, respectively.

To get the error signal for the control of the optical feedback phase, the symmetry of the arch-shape cavity mode should be acquired before performing the trigger action. Here, the method similar to that in [18] is adopted to show that the ringdown event is excited on the falling edge of the cavity mode. By changing the laser current below the threshold of the laser emitting quickly, the ringdown event can be observed in the cavity transmission. An exponential function is used to fit the ringdown signal by the least square method.

RESULTS AND DISCUSSION

Ringdown Signal

Special measures to avoid the trigger of unwanted ringdown events during the laser current recovery have been taken, and their effect is illustrated in **Figure 2** by the time sequence of process signals. The black curve in **Figure 2A** is the cavity

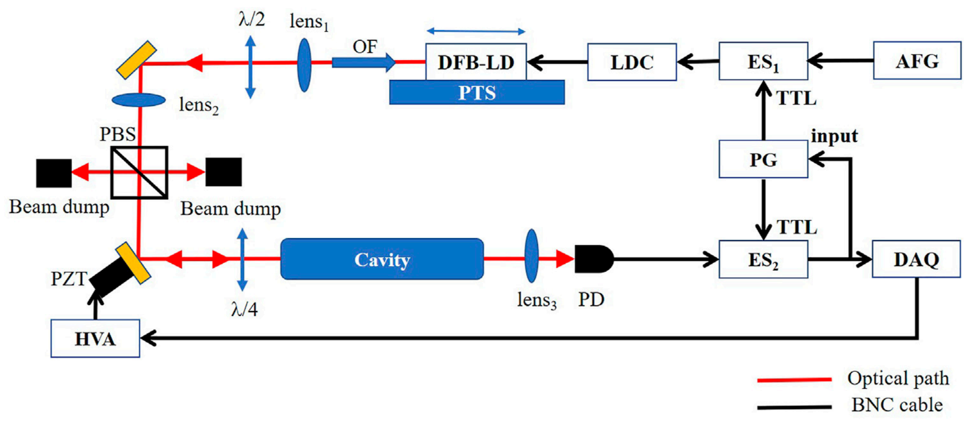

FIGURE 1 | Experimental setup for OF-LCRDS. PG: pulse generator; $ES_{1,2}$: homemade electronic switch; AFG: arbitrary function generator; LDC: laser diode controller; DFB-LD: distributed feedback diode laser; PTS: precision translation stage; λ/2: half-wave plate; PBS: polarization beam splitter; PZT: piezoelectric transducer; HVA: high voltage amplifier; PD: photodetector; and DAQ: data acquisition card.

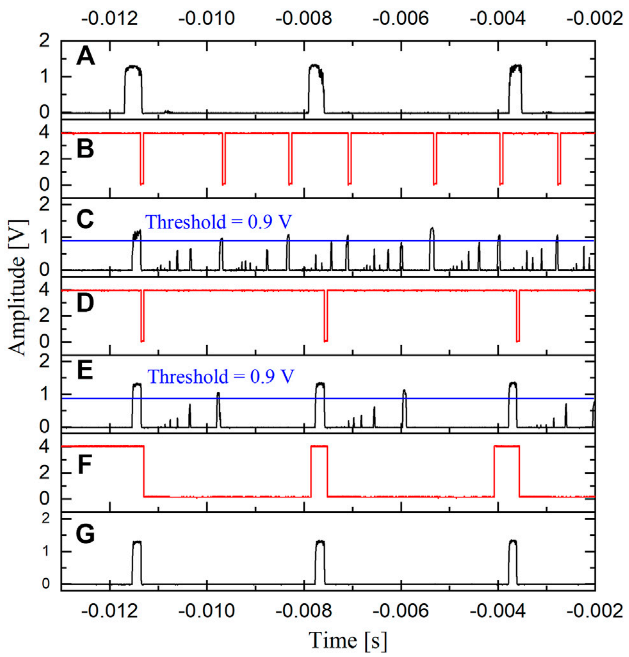

FIGURE 2 | The time sequence of the process signals. **(A)** The cavity transmission modes without the trigger of ringdown event; **(B)** the pulse signal to the ES_1; **(C)** the cavity transmission modes of OF-LCRDS, and a series of messy cavity modes are observed under this situation; **(D)** the pulse signal to the ES_1 with the ES_2; **(E)** the cavity transmission modes of OF-LCRDS with the ES_2; **(F)** the pulse signal to the ES_2 to filter out the unexpected modes; and **(G)** the signal to the DAQ card with the ES_2.

transmission signals for CEAS when the laser current is scanned. There are three successive cavity longitudinal modes, all of which have a typical arch profile. At these points, the laser frequency is briefly locked to the corresponding cavity mode with the time scale of 0.5 ms, even though the nominal linewidth of the laser offered by the manufacturer is 5 MHz and the linewidth of the cavity mode is 54 kHz. This verifies the effect of the optical feedback that could suppress most of the laser frequency noises.

When a falling edge is detected and its amplitude drops below the trigger threshold, a pulse signal with 60 μs duration is generated by the PG and then sent to the ES_1 to cut off the laser current. As a result, one ringdown event is observed. The output signal of the PG is shown in **Figure 2B** with a trigger threshold voltage of 0.9 V for the cavity transmission signal. The curves in **Figure 2C** are the corresponding cavity transmission modes. After 60 μs, the output of the PG would return to its initial state. However, the recovery of the laser wavelength is relatively slow and several unexpected cavity modes might be stimulated during its recovery process, exemplified by the cavity transmission modes with relatively lower amplitude, as in **Figure 2C**. It would also excite the ringdown event if it satisfies the requirement of the trigger. Consequently, a series of messy cavity ringdown events stimulated at uncertain laser frequencies are resulted, just as illustrated in **Figures 2B,C**. This problem could be solved if a higher threshold voltage for the ringdown trigger is set, whereas this strategy is not suitable for the case of large variation of the laser power. For example, a large scanning range of the laser frequency for the detection of a complete absorption spectrum will result in intrinsic variation of the laser power along with the scanning of the laser current.

Here, to address the problem, another electronic switch, i.e., the ES $_2$, is added after the PD, which is controlled by the PG and can filter out these unexpected modes. **Figure 2F** depicts the control signal to the ES_2. Initially, the ES_2 is turned on and the PG can receive the output of the PD. A pulse with a duration of 3.4 ms which lags behind the pulse to the ES_1 is generated by the PG and sent to the ES_2. After the ringdown signal has been recorded using the DAQ card, it will turn off the ES_2 and, thus, cut the link between the PG and the PD. After the laser wavelength gets stable, the ES_2 is turned on again and another ringdown event could be triggered. **Figures 2D,E,G** show the output of the PG to the ES_1, the output of the PD, and the input of the DAQ card by using this new strategy, respectively. It is clear to see, even though

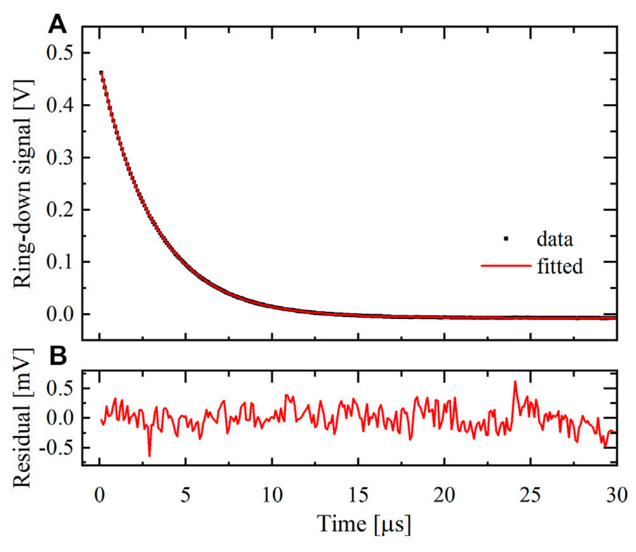

FIGURE 3 | (A) Measured single ringdown event (black dot) and its fitting result by an exponential function (red line). **(B)** The fitting residual.

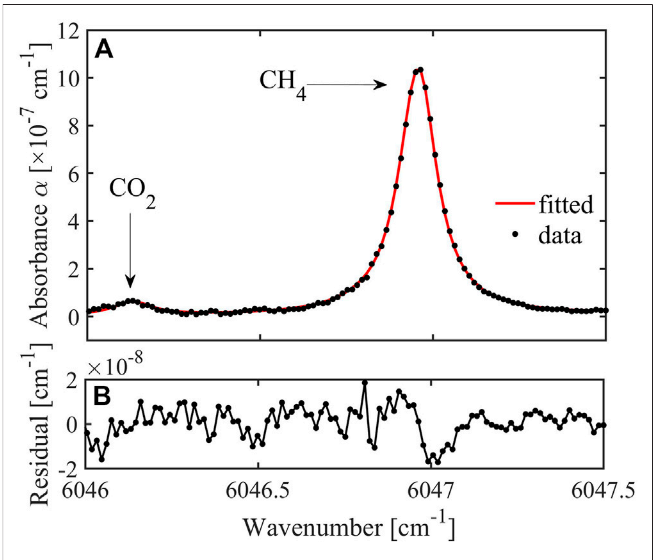

FIGURE 5 | (A) Measured CH_4 absorption spectrum and its fitting result with Lorentz function; **(B)** fitting residual.

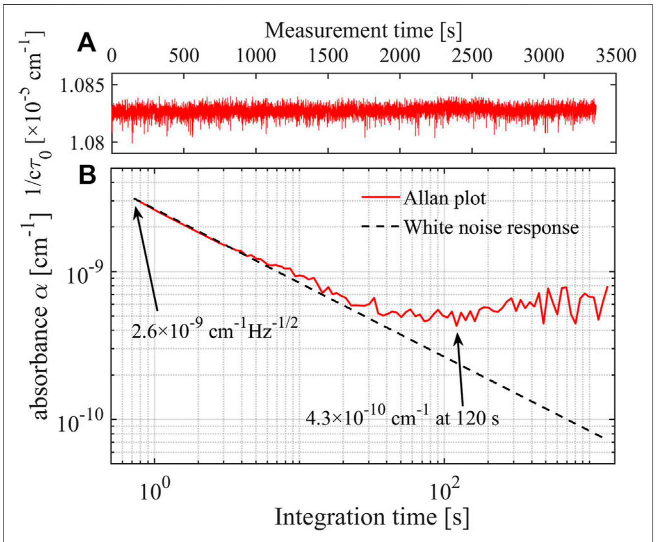

FIGURE 4 | Long-term ringdown time measurement of a single cavity longitudinal mode. **(A)** Empty cavity decay rate; **(B)** Allan variance plot.

the numerous cavity transmission modes are still observed during the recovery of the laser frequency, they do not affect the regular trigger of the desired ringdown event. As a result, three ringdown events corresponding to the three consecutive cavity modes are observed, as shown in **Figure 2G**. With the improvement of the coupling efficiency, large amplitude of the cavity transmission mode as well as the cavity ringdown signal with high repeatability and high signal-to-noise ratio is obtained, illustrated by the black dot in **Figure 3A**. The red curve is the fitting result based on an exponential decay function; and the lower panel is the fitting residual. An excellent signal-to-noise ratio (SNR) of 2500, defined as the ratio of the peak value and the standard deviation of the residual, is obtained. Also, no strong structure has been found in the fitting residual.

Evaluation of the Detection Limit

To estimate the detection sensitivity of OF-LCRDS, a series of ringdown events for a single longitudinal mode of the empty cavity was consecutively measured for around 1 h. This is realized by tuning the laser frequency with a triangle wave signal at a rate of 2 Hz. The red curve in **Figure 4A** depicts the measured empty cavity decay rate, $1/(c\cdot\tau_0)$, over time (c is the speed of light, and τ_0 is the empty cavity ringdown time). It demonstrates a 0.04% fractional uncertainty of the ringdown times, i.e., $\sigma(\tau_0)/E(\tau_0)$ [$\sigma(\tau_0)$ is the standard deviation of τ_0, and $E(\tau_0)$ is the mean value of τ_0], which is among the state-of-the-art of CRDS results. Slow fluctuations can be seen in **Figure 4A**, and it is suspected to be caused by the environmental temperature change which leads to the variation of the cavity length and mechanical vibration which leads to the light hitting the different spots of the cavity mirrors. There is also sparse impulse noise which is attributed to the electric noise and etalon effect. The Allan variance plot [24] is shown as a red curve in **Figure 4B**. A white noise response of 2.6×10^{-9} cm^{-1} Hz$^{-1/2}$ was obtained, which is illustrated by the dash line in black. Also, the system reached its detection limit of 4.3×10^{-10} cm^{-1} at an integration time of 120 s, corresponding to a minimal detection of CH_4 concentration of 1.2 ppb by a CH_4 transition at 6046.9 cm^{-1}.

Measurement of the CH_4 Absorption Spectrum

Then, the absorption spectrum of CH_4 is measured. We filled the cavity with the ambient air of our lab located at Taiyuan,

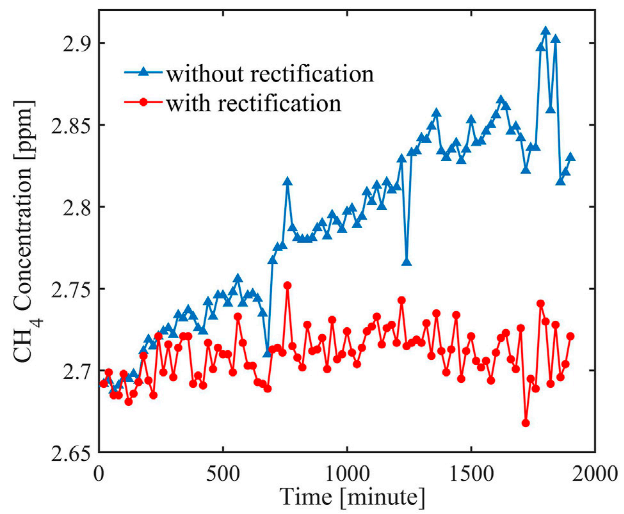

FIGURE 6 | Long-term measurement of CH_4 with constant concentration.

China, which is filtered by using a desiccant first. The intracavity pressure was set to 0.92 atm, i.e., the local atmospheric pressure. In the experiment, each cavity mode was scanned 50 times for average ringdown time. The laser current is scanned by a triangular wave with a frequency scanning range of around 0.5 FSR at a rate of 50 Hz. The shift of the adjacent cavity mode is realized by stepping the central value of the laser current, and it took 7 min to get a whole spectrum. The empty cavity ringdown time is measured at the wavelength of 6047.5 cm^{-1}, which is 18 GHz away from the center of the nearest strong absorption transition. The obtained absorption spectrum is shown as the black dots in **Figure 5A**. With a total laser frequency scanning range of 45 GHz, 118 successive cavity modes are stimulated and a CO_2 absorption line is observed besides the CH_4 absorption line, shown as a small bump in **Figure 5**. The spectral parameters from the HITRAN database [26] and Lorentzian line-shape function are used to fit the measurement spectrum. The fitted curve is displayed as a red line in **Figure 5A**, and the fitting residual is presented in **Figure 5B**. The theoretical model shows a good consistency with the measured spectrum, and the signal-to-residual ratio of 156 is obtained. The retrieved CH_4 concentration is 2.73 ppm.

Rectification of the Empty Cavity Ringdown Time

The variation of the empty cavity ringdown time, owing to temperature fluctuation and mechanical vibration, is the dominant limitation to the long-term stability of CRDS. To solve this problem, the empty cavity ringdown time is rectified regularly by the ringdown time at the laser frequency far away from the gas absorption transition. To verify the effectiveness of this method, a long-term concentration measurement with a time interval of 20 min and a duration of 34 h for CH_4 gas with a constant concentration has been performed. The dotted lines in blue and red in **Figure 6** are the measured concentrations without and with rectification, respectively. Without rectification, the drift range of retrieved concentration is 0.2 ppm within this measurement time. After the rectification, the drift range is suppressed to 0.08 ppm, which shows an improvement with a factor of 2.5.

CONCLUSION

In summary, we have developed optical feedback linear cavity ringdown spectroscopy. The ringdown event is excited by changing the laser current below the threshold of emitting quickly. An effective method to avoid the ringdown events excited by the unexpected cavity modes during the process of current recovery is introduced with one electronic switch. The coupling efficiency of the laser to the FP cavity is improved to 31%, mainly limited by the mode mismatch and impedance mismatch, by optical feedback, and consequently, ringdown signals with high fidelity are achieved. The fractional uncertainty of empty cavity ringdown time is 0.04%, yielding a minimal detection of CH_4 concentration of 1.2 ppb at the integration time of 120 s. The long-term retrieved concentration drift is improved by 2.5 times within 34 h through rectifying the empty cavity ringdown time during the measurement of each spectrum. This novel technique paves the way for the construction of a robust and sensitive CRDS instrument for trace gas detection.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

XW: data curation, formal analysis, methodology, and writing—original draft. GZ: conceptualization, supervision, validation, visualization, writing—review and editing. KJ: formal analysis. BC: software and validation. RK: resources and writing—review and editing. ZC: resources and validation. JL: supervision, funding acquisition, project administration, and writing—review and editing. WM: writing—review and editing.

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