Performance test of an aerosol concentration measurement system based on quartz crystal microbalance

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Abstract. QCM or quartz crystal microbalance is a well-known sensor technology that generates cycles of oscillation related to mass change on the crystal’s surface. This crystal works well when it has a frequency counter and an oscillator to drive the crystal and count the oscillation, and a good airflow regulator. This study developed a measurement system for aerosol concentrations with a diameter of less than 2.5 micrometers. The system consists of QCM sensors, an oscillator, a frequency counter, and an airflow regulator. The system was tested inside an exposure chamber with a constant emission source for the different velocity speeds, namely $v_1$, $v_2$, $v_3$, $v_4$, and $v_5$. The test was conducted every 10 seconds due to the saturated time of the QCM related to the mass loading effect of aerosol. The results show that the system can drive the QCM sensor with a frequency of 5 MHz. The measurement system works well to measure aerosol concentration after the preload duration often seconds and every sixty seconds in which the durations are related to the optimum QCM’s response at $v_1$ and $v_2$. The optimum performance was found to be in the laminar regime, with the sample rate of 0.6 m/s to 1.0 m/s.

1. Introduction
Air pollution is a major environmental pollution problem affecting everyone in many countries. This pollution generally can be divided into gaseous and particulate emissions. Different classification can vary in terms of chemical compounds and structures depending on the primary emission sources. Gaseous emission can be found in SO$_2$, NO$_x$, NO$_2$, CO, and many others [1]. Besides, particulate emission, known as aerosol or particulate matter, consists of a mixture of solid particles and liquid droplets (both organic and inorganic compounds) suspended in the ambient air [2]–[4].

In terms of aerosol emission, they can be classified into some types depending on their average diameters. Studies indicate that fine (as known as PM$_{2.5}$; particles with aerodynamic diameter ≤ 2.5 µm) and coarse particles (known as PM$_{10}$; particles with aerodynamic diameter ≤ 10 µm) ubiquitous in atmospheric air. The smallest one, ultrafine particle (PM$_{0.1}$; particles with aerodynamic diameter ≤ 10 µm), even become the most contributor to air pollution in terms of aerosol emission. Another study
also divides aerosols peculiarly into superfine (PM$_{0.18}$, PM$_{0.32}$, and PM$_{0.56}$) and ultrafine particles (PM$_{0.056}$, and PM$_{0.1}$) [2].

All of them are recognized as significant contributors to respiratory illness and even mortality, mainly resulting from many combustion processes. They can be generated from biomass burning [5], industrial combustion process [6], coal combustion [7], [8], diesel-powered vehicles [3], [9], [10], motorcycle exhaust [11], and many others. In urban areas, road traffic constitutes the primary source of particulate matters, as reported in Japan [3], Helsinki, Finland [12], Lucknow, India [2], New York, United States [13], and in Budapest, Vienna, and Prague (European cities) [1].

Aerosol measurements are typically reported in terms of annual mean or daily concentrations. These types of routine measurements describe such PM concentrations in terms of milligrams per cubic meter (mg/m$^3$), micrograms per cubic meter (µg/m$^3$), and nanograms per cubic meter (ng/m$^3$) [2]. The concentrations are interpreted as the mass of detected particles per cubic meter (particles/cm$^3$) due to a big diameter size, especially for fine and coarse particles. Ultrafine particles, which have the smallest diameter, use the number of detected particles per cubic centimeter, neglecting the mass distribution [3]. All of them can be measured using many methods, such as a fast-scanning TSI SMPS (Scanning Mobility Particle Sizer) spectrometer [1], [13], TSI P-Trak Ultrafine Particle Counter [14], [15], low-pressure impactors [3], and Miniature DiffusionSize Classifier (miniDiSC).

Based on the explanation above, various methods and instruments have been recently used for aerosol measurements. However, most of the systems use an optical principle that is well-known to have excellent performance but has extra attention and maintenance. Most of them are relatively grouped into high-cost instrumentations that need more budgets. Still, they have a limit for measurement in an extreme environment, such as a high humidity condition. Recently, a quartz crystal microbalance, QCM sensor, has gained a great concern because it can measure the mass change due to the sensor frequency shift with nanoscale resolution. Also, QCM sensor has the advantage of fast building time, while the performance depends on the coated materials on the surface of QCM. It is also relatively considered a low-cost sensor. Hence, it can be developed as an aerosol sensor to measure the concentration with different diameter sizes. However, there is a limited study reported so far for a QCM aerosol sensor. In this paper, we report the aerosol measurement system assembled with a bare quartz crystal microbalance. This study is a preliminary study for developing an aerosol measurement system with a more complex subsystem.

2. Materials and methods

2.1. Sensing element

Bare QCMs (uncoated) with 5 MHz fundamental frequency (AT-cut, 8 mm in diameter) were used as the aerosol sensors. The diameter of the electrodes (silver electrode) was 4 mm (area $A = 1.26 \times 10^{-5}$ m$^2$). All uncoated QCMs were washed with acetone, ethanol, and deionized water, consecutively. After that, they were dried for a whole night (at room temperature) before being used [16]. The electrodes were connected to a self-developed oscillator to drive the sensor, combined with a signal filter circuit. The resulted frequency was measured using a frequency counter.

2.2. Performance test

A series of aerosol particle concentrations were generated from cigarette burning (constant mass, 1.501 gram) as the aerosol sources. They were varied into five different velocities: $v_1 (0.6$ m/s), $v_2 (1.0$ m/s), $v_3 (1.2$ m/s), $v_4 (1.8$ m/s), and $v_5 (2.0$ m/s). This emission was exposed to a collecting chamber (exposure chamber, 14 cm in width x 15 cm in length x 10 cm in height, volume = 2,100 cm$^3$) for 12 minutes (this duration was chosen due to the time needed to inject emission from the burnt cigarette sample completely, according to the preliminary study) with a constant suction pump (velocity = 2.0 m/s). Then, the emission was channeled into an exposure chamber containing a fine particle filter. This exposure chamber (fine particle chamber) had a Whatman™ filter paper Grade 5 to filter the emission and only let particulate matter with a diameter less than 2.5 µm pass through this filter (Figure 1).
After exposing the emission, the aerosol concentration inside the exposure chamber was monitored using a Handheld Air Tester (Hinaway, Model CW-HAT200S). Then, the frequency shift of the QCM, $\Delta f$, was recorded per 10 seconds as the sampling time (a self-fabricated data-logger, which was connected to the frequency counter). The measurement was conducted until the aerosol mass concentrations inside the chamber after being exposed ($C$) were similar to the original concentration ($C_0$, before being exposed by the emission). These treatments were repeated three times for each velocity variation. As an important control parameter, all treatments were conducted in a controlled exposure chamber with a temperature of 25±0.3°C and 65±0.3% of humidity.

![Figure 1. Schematic diagram of the aerosol concentration measurement system.](image)

The tested system's sensitivity was defined as the response frequency ($f$) to the target [17]. Frequency shift ($\Delta f$) was obtained from $f - f_0$, where $f_0$ was the frequency benchmark (fundamental frequency), and $f$ was the frequency response of the QCM exposed to certain $C$. Response time and repeatability were evaluated by repeating each test using a similar condition ($n = 3$). Each test was repeated three times, in which the standard room temperature ±25°C.

2.3. Statistical analysis

All results were interpreted as mean±SEM (standard error of the mean), in which the parameter of the statistic was the mean. The sampling distribution of the mean was generated by repeated sampling ($n = 3$). An ANOVA statistical test was conducted to determine the differences in the mean response between each aerosol concentration. Thus, $p < 0.05$ was determined as statistically significant [8].

3. Results

3.1. The waveform of the QCM sensors

Figure 2 shows the system's waveform observed from a digital oscilloscope at five stages: $v_1$, $v_2$, $v_3$, $v_4$, and $v_5$ before and after injecting the aerosol mass concentrations. All tests were performed in a closed-system (inside an exposure chamber). This figure shows that each test has a similar fundamental frequency (initial condition). After injecting the aerosol mass concentration, the frequency decreases, whether in $v_1$, $v_2$, $v_3$, $v_4$, or $v_5$. It can be seen that the initial condition ($f_0 = 4.99773$ MHz) has a higher
frequency than the final condition \(f_0\). This final condition refers to the condition in which the QCM sensor reaches the saturated zone.

![Image](https://example.com/image1)

**Figure 2.** The observed waveforms of the bare QCM sensors' output signals at all treatment stages: (a). initial condition (fundamental frequency \(f_0\); and (b). after exposure.

3.2. Frequency shift
The aerosol concentration measurement system test has been preliminarily performed to evaluate the characterization of the velocity experimentally. The velocities are varied at five different speeds. After pumping in the exposure chamber containing aerosols, the highest velocity \(v_5\) has the fastest saturation time (Figure 3). The test duration was performed for 100 seconds, in which the lowest frequency was 4.99765 MHz. It means that \(v_5\) has 47 Hz of frequency shift \((\Delta f_5)\) related to the fundamental frequency \(f_0\) (4.99773 MHz). The duration is only 40 seconds (frequency is 4.99768 MHz), and it gets saturated at the 50th second (saturated frequency is 4.99770 MHz). The second position refers to \(v_4\) with the frequency shift \(\Delta f_4\) of 47 Hz, having 4.99768 MHz of the lowest frequency for 50 seconds of performance test duration.
Figure 3. The experimental and curve-fitting results with different sucking velocities. These results show a linear response of the miniature monitoring system using a bare QCM.

The longest duration belongs to the lowest velocity, \( v_1 \). This variation needs more than 60 seconds to reach saturation time. This variation has \( \Delta f_1 \) of 60 Hz, having 4.99765 MHz of the lowest frequency. According to Figure 3, \( v_2 \) has more than 60 seconds to get the saturation time, showing 60 Hz of the frequency shift \( \Delta f_2 \) (\( f_t = 4.99765 \) MHz). In the middle position, \( v_3 \) needs 60 seconds to get saturated with the frequency of 4.99768 MHz. It has a frequency shift of 50 Hz. This figure also shows
a significant correlation between performance test duration and the frequency shift for all velocity variations ($R^2 > 0.75$ and $p < 0.05$).

4. Discussion
The developed system was characterized. The results show that aerosols' concentrations inside the exposure chamber were constant since only one constant source was used in this study (cigarette smoke). The measurement lasted 100 seconds (for the maximum stage), and the frequency was recorded in every sampling time (every 10 seconds) from a digital oscilloscope. The data were plotted on a polynomial regression (in a series of times). The performance test result indicates that the aerosol mass loading on the surface of the QCM decreases the fundamental frequency $f_0$. These results imply that the QCM sensor linearly responds to the mass concentration ($\Delta m$) as particles are continuously injected into the exposure chamber.

During the measurement time $\Delta t$, the frequency shift was about $47 – 60$ Hz. According to the equation below, we can get the particle mass concentration $\Delta m$ (in the unit of mass, $\mu g$) corresponded to the frequency shift,

$$\Delta f = \frac{2 f_0^2}{A \sqrt{\mu q \cdot \rho q}} \Delta m$$ \hfill (1)

Based on the equation above, we can calculate the mass concentration of the injected aerosol. The results, $\Delta m$, are shown in Table 1 below. These results can also be used to calculate the sensitivity of the QCM, as the ratio of response frequency ($\Delta f$) to the target [17]. The sensitivity of the QCM was calculated using [18]:

$$\text{Sensitivity (Hz/µg)} = \frac{\Delta f}{\text{aerosol mass change}}$$ \hfill (2)

| Velocities | $\Delta f$(Hz) | $\Delta m$ ($\mu g$) | Sensitivity (Hz/µg) |
|------------|----------------|----------------------|---------------------|
| $v_1$      | 60             | 13.48                | 4.45104             |
| $v_2$      | 60             | 13.48                | 4.45104             |
| $v_3$      | 50             | 11.23                | 4.45236             |
| $v_4$      | 47             | 10.48                | 4.48473             |
| $v_5$      | 47             | 10.48                | 4.48473             |

According to the equation above (Sauerbrey equation), the aerosol debit $Q$ does not influence the mass fluctuation. A constant flowrate will drain particle concentration in a certain volume $V$ (we used a constant volume for a control parameter). Otherwise, a nonconstant flowrate will have an impact on the mass loading effect since:

$$Q = A \nu$$ \hfill (3)

and:

$$Q = \frac{\nu}{t}$$ \hfill (4)

so we can get:

$$V = A \nu t$$ \hfill (5)

$A$ is the sample inlet area ($\text{cm}^2$), $\nu$ is the sample rate, and $t$ is the length of the sample flow.
According to the equations above, the volume V is influenced by the time used to test the system performance (testing duration). A longer time is needed for a slower velocity since we used a constant V. It means that a faster rate will have a faster saturation time. This saturation time is when the crystal QCM will no longer be good for the system. In other words, the used QCM sensor has reached the maximum response in accordance with the injected particle mass.

The precision sector for measuring the aerosol concentration also depends on the minimum detectable mass loading effect of the aerosol on the QCM surface. Due to the noise of the system (such as the oscillator circuit and the QCM housing), the frequency will fluctuate during the sensing of the mass loading effect. For example, we found an unstable stage at the first measurement time. In a short-term duration (<10 seconds), the system fluctuated. The fluctuation was found to be 10 Hz over this period. Therefore, the system's mass loading resolution is obtained to be more than 10 seconds as a stable measurement time (preload) for minimum detectable aerosol mass.

The QCM sensor output also depends on the flow pattern. The nozzle used in this system is one of the most important design parameters. To obtain a significant correlation of the performance test, laminar flow in the pumping system is required[19-20]. This condition is related to the velocity that is being characterized. It needs the Reynolds number (Re) in the range from 500 to 3000 by using:

\[ Re = \frac{4pWho}{\mu (w+h)} \]  

(6)

The air density is shown by ρ (1.2 kg/m³). W refers to the width of the injection nozzle width (8 mm), while h is the nozzle’s height (55 mm). The dynamic viscosity of a particle is shown by μ (1.18 x 10⁻³ Pa x s). Following the above equation procedure, the Re number is calculated to be 1705, 2841, 3409, 5114, and 5682 (laminar and turbulent regimes), respectively, for \( v_1 \), \( v_2 \), \( v_3 \), \( v_4 \), and \( v_5 \). Only \( v_1 \) and \( v_2 \) are there in the laminar regime. Although \( v_5 \) is not in the laminar regime, it still has a good sensitivity.

5. Conclusion

In conclusion, a correlation between test duration and frequency shift of a QCM sensor in the developed system has been tested under different air flow speed. A faster flowrate (2.0 m/s) has a more quick response, resulting in a smaller response time (40 seconds) and lower sensitivity (4.48 Hz/µg). The slowest flowrate affects the longest response time (60 seconds), and it has the highest sensitivity (4.45 Hz/µg). A bare QCM is reliable for measuring aerosol mass concentration with a diameter of fewer than 2.5 micrometers in a quick response.

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