

## Response to Reviewers for "Deriving the hygroscopicity of ambient particles using low-cost optical particle counters".

We would like to thank the anonymous reviewers for the comments that significantly improved the clarity and readability of the manuscript. Our point-by-point responses are found below in blue ink. The revised content is highlighted in yellow.

### RC1

1. You stated that "AQB-monitored PM concentration can be converted to dry particle mass concentration, aligning well with EPA data after OPC sensitivity correction. The derived hygroscopicity provides the relationship between ambient relative humidity and particle water content. By dividing PM<sub>10</sub> into PM<sub>2.5</sub> and PM<sub>2.5-10</sub>, considering the composition heterogeneity, we achieved more precise dry PM<sub>10</sub> concentrations with lower MAPE." Please state differences clearly with 2 different observing packages.

A: Thank you for your constructive comments and for emphasizing the need for clarity in differentiating between the two observational packages used in our study. The EPA stations (using METONE BAM1020) report dry-state PM concentrations by controlling the measurement environment to maintain relative humidity (RH) below 50%. In contrast, the optical particle counter (OPC) in the AQB directly monitors ambient PM concentrations. The comparison of these two datasets illustrates the sensitivity variation of low-cost OPC sensors and the influence of hygroscopicity. The content of Section 4 (Lines 288-294) was rewritten to clarify this issue as follows: "In the PM analysis, PM<sub>10</sub> was divided into PM<sub>2.5</sub> and PM<sub>2.5-10</sub> to account for compositional heterogeneity among different particle sizes. Comparing the AQB-monitored ambient PM data and the TW-EPA data (for dry particles) at RH ≤ 50%, the derived sensitivity coefficients ( $\alpha$ ) for PM<sub>2.5-10</sub> (10.58 - 12.37) were higher than those for PM<sub>2.5</sub> (1.26 - 1.44) likely due to the significant sensitivity variation in the OPC over time. By considering hygroscopicity with the  $\kappa$ -Köhler equation and assuming a constant composition density for sensitivity-corrected AQB data, the derived dry particle mass concentrations show improved consistency with TW-EPA data compared to the simple linear regression approach."

2. Provide units for the parameters in the equations.

A: Thank you for your kind reminder. All equations are labeled with units to ensure correct use. The adjusted revisions for the descriptions of equations (Eqs.1 and 2) in the paragraph as follows: "

$$\alpha = \frac{M_{EPA}}{M_{OPC}} \quad (1)$$

where  $M_{EPA}$  and  $M_{OPC}$  are PM concentrations ( $\mu\text{g m}^{-3}$ ) measured by TW-EPA and OPC, respectively.

$$S = \frac{D_{amb}^3 - D_d^3}{D_{amb}^3 - D_d^3(1-\kappa)} \exp\left(\frac{4\sigma_{s/a}M_w}{RT\rho_w D_{amb}}\right) \quad (2)$$

where  $D_{amb}$  and  $D_d$  are the diameters (m) of the ambient and dry particulate matter, respectively,  $\sigma_{s/a}$  is the surface tension of the particle ( $\text{J m}^{-2}$ ),  $M_w$  is the molecular weight of water ( $\text{g mole}^{-1}$ ),  $R$  is the gas constant ( $\text{J mole}^{-1} \text{K}^{-1}$ ), and  $\rho_w$  is the density of liquid water ( $1.0 \text{ g m}^{-3}$ ). The first term is the solute effect, while the second term is the Kelvin effect."

In Eqs.3 and 4, units for the parameters in the equations have already been provided in Eqs. 1 and 2 or the paragraph. For Eq.5, the volume mixing ratio ( $\epsilon$ ) and hygroscopicity ( $\kappa$ ) is a dimensionless quantity.

3. How accurate your hygroscopicity calculation that needs to be discussed.

A: In this study, the low-cost sensors acquired data for a certain period to cover a more comprehensive RH range for the hygroscopicity calculation. In the studied case, the analysis successfully showed different mean  $\kappa$  values between  $\text{PM}_{2.5}$  and  $\text{PM}_{2.5-10}$  based on the data of the monitored period. Even though there was no intensive filter sample collection during the studied period, the comparison of two IC analysis results from other studies (2013 and 2021 winter campaigns) shows similar hygroscopicity of  $\text{PM}_{2.5}$ , ranging from 0.18 to 0.25, which might represent a typical winter  $\text{PM}_{2.5}$  hygroscopic characteristics in Kaohsiung city. The consistent results between the two winter campaigns might suggest the overall soluble species fraction in  $\text{PM}_{2.5}$  is generally within a similar range. Therefore, the derived mean  $\kappa$  range under the specific assumption (solute density and ignorance of Kelvin effect) from AQB data was discussed for the range consistency compared to the results of available two winter campaigns for  $\text{PM}_{2.5}$  and the 2013 winter campaign for  $\text{PM}_{2.5-10}$ . However, the accuracy improvement of dry particle mass concentration derived from the AQB monitored data after applying the OPC sensitivity coefficient and derived  $\kappa$  values can be evaluated as illustrated in Section 3.2 and summarized in a new table (Table 2).

**Table 2: Performance metrics of different calibration methods for  $\text{PM}_{2.5}$ ,  $\text{PM}_{2.5-10}$ , and  $\text{PM}_{10}$ .**

	$\text{PM}_{2.5}$			$\text{PM}_{2.5-10}$			$\text{PM}_{10}$			
	RH $\leq$ 50% Only <sup>a</sup>	All data (no $\kappa$ )	All data ( $\kappa=0.29$ )	RH $\leq$ 50% Only <sup>a</sup>	All data (no $\kappa$ )	All data ( $\kappa=0.09$ )	RH $\leq$ 50% Only <sup>a</sup>	All data (no $\kappa$ )	All data ( $\kappa=0.36$ )	( $\text{PM}_{2.5} + \text{PM}_{2.5-10}$ ) <sup>c</sup>
applied $\alpha$	1.26 $\pm$ 0.16	1.04	1.40	12.37 $\pm$ 1.33	10.77	13.16	2.02 $\pm$ 0.34	1.69	2.36	—
MAPE (%)	21.3 (12.8)	48.8	24.8	15.9 (11.5)	37.9	31.8	32.8 (18.5)	62.5	29.2	18.2
RMSE ( $\mu\text{g cm}^{-3}$ )	20.5 (3.7)	29.1	11.3	4.9 (2.8)	9.4	9.1	42.6 (10.3)	54.7	26.9	15.9
$R^2$ <sup>b</sup>	0.55 (0.51)	-3.49	0.32	0.31 (0.78)	0.57	0.59	-4.18 (-0.58)	-4.74	-0.38	0.51

<sup>a</sup> Only for data points at RH  $\leq$ 50%. The value in parentheses is the performance result without two significant outliers shown in Fig. 3

<sup>b</sup> Coefficient of determination ( $R^2$ ) is calculated as the proportion of variation in the calibrated dry mass concentration.

<sup>c</sup> The combination of calibrated data from  $\text{PM}_{2.5}$  All data ( $\kappa=0.29$ ) and  $\text{PM}_{2.5-10}$  All data ( $\kappa=0.09$ ).

To clarify this approach, Sections 3.2 and 3.3 were revised as follows: in Section 3.2 (Lines 172-211): “Figures 3(a) and 3(c) show the scatter distribution of the mass concentrations between AQB #1 (with no calibration) and TW-EPA data for PM<sub>2.5</sub> and PM<sub>10</sub>, respectively. Overall, the PM mass concentrations measured by AQB system appear to be higher than those reported by TW-EPA. The results reveal an apparent influence of ambient RH, indicating the contribution of water content. The red-shaded area represents a regression line with a slope corresponding to the inverse of the sensitivity coefficients ( $\alpha$ ) derived from data points at ambient RH  $\leq$  50% (17 out of 356 points, 5%). The notable deviation of the red shaded area from the 1:1 line towards the right side indicates the requirement of  $\alpha > 1$  corrections, contributed by the different measurement principles and calibration techniques, which may result from the assuming particle density and refractive index (RI) (dust, density: 1.65 g cm<sup>-3</sup>, RI: 1.5 + 0i). The estimated  $\alpha$ , as summarized in Table 1, are higher for PM<sub>10</sub> than for PM<sub>2.5</sub>, i.e.,  $2.02 \pm 0.34$  vs  $1.26 \pm 0.16$ , which are reasonably conclusive as tested with more data points selected at higher RH thresholds (Fig. S2). The  $\alpha$  difference between PM<sub>2.5</sub> and PM<sub>10</sub> might be attributed to the complex composition of ambient particles, which differs from the samples used for instrument calibration, as well as possible sensitivity variations in OPC over time. With sensitivity calibration, the performance at ambient RH  $\leq$  50% exhibits a strong correlation with MAPE at 12.8%, 18.5%, and Root Mean Squared Error (RMSE) at 3.7  $\mu\text{g m}^{-3}$ , 10.3  $\mu\text{g m}^{-3}$  for PM<sub>2.5</sub> and PM<sub>10</sub>, respectively, as summarized in Table 2 excluding the two significant outliers (shown as hollow circles in Fig. 3). The results confirm the effectiveness of OPCs in capturing PM concentrations, consistent with previous real-time outdoor field studies (Gillooly et al., 2019; Demanega et al., 2021; Sá et al., 2022; Crilley et al., 2018). Additionally, the OPC sampling flow rate has an impact on measurement performance. AQB #1 maintained a steady rate at  $3.6 \pm 0.2$  LPM, whereas AQB #2 exhibits two distinct time periods with sampling flow rates of 3.6-4.2 LPM for the first period and 3.2-3.6 LPM for the second period. ... With the derived  $\alpha$ , the hygroscopicities were retrieved using Eq. (3), resulting in  $\kappa$  ranging from 0.18 to 0.29 for PM<sub>2.5</sub> and 0.20 to 0.39 for PM<sub>10</sub> (Table 1) during the studied period. Figures 3(d) and 3(f) show the scatter distribution of the derived dry concentration vs. TW-EPA concentration for PM<sub>2.5</sub> and PM<sub>10</sub>, respectively. The results from the two AQB systems exhibit slight differences but are consistent overall. Considering both the sensitivity coefficient and hygroscopicity, the performance of AQB in deriving dry PM concentration is significantly improved with lower MAPE, RMSE, and higher R<sup>2</sup> than the results obtained using only the sensitivity coefficient, as summarized in Table 2. ... The lower  $\kappa$  for PM<sub>2.5-10</sub> might suggest a significant contribution from dust or other less hygroscopic species, consistent with the IC analyses in Table 3 and discussed further in Sect. 3.3. With the retrieved  $\alpha$  and  $\kappa$  for PM<sub>2.5</sub> and PM<sub>2.5-10</sub>, Fig. 3(e) shows the scatter distribution between the derived dry PM<sub>2.5-10</sub> from AQB data and TW-EPA data, exhibiting a MAPE of 31.8%, more significant than the 24.8% for PM<sub>2.5</sub>. ... Detection efficiency may be influenced by notable spatial variations, aligning with the findings of Kaliszewski et al. (2020), which showed a reduced correlation between OPC-N3 measurements and reference instruments for larger particles. The dry PM<sub>10</sub> derived from AQB through the divided PM<sub>2.5</sub> and PM<sub>2.5-10</sub> analysis demonstrates better consistency with the reported TW-EPA data than the direct calibration method. This is evidenced by a lower MAPE in Fig. 3(g) (18.2%) compared to Fig. 3(f) (29.2%) and a significant improvement than the simple linear regression method, which has a higher MAPE at 62.5% (Table 2).

This substantiates the importance of considering composition heterogeneity among particle sizes for accurate dry PM derivation. “; and in Section 3.3 (Lines 222-235): “A similar analysis for the winter of 2021 yielded a consistent  $\kappa$  range for PM<sub>2.5</sub>, as illustrated in Fig. S5. This consistency across distinct study periods indicates typical ambient PM<sub>2.5</sub> hygroscopic characteristics in Kaohsiung City during winter, which can be applied for further discussion with the AQB data. For coarse particles, the more significant variability in  $\kappa$  for PM<sub>2.5-10</sub> compared to PM<sub>2.5</sub> can be attributed to the significant fluctuations in the soluble composition of coarse particles, primarily driven by substantial quantities of thenardite (Na<sub>2</sub>SO<sub>4</sub>) and halite (NaCl) (Tang et al., 2019). ... The derived  $\kappa$  value for PM<sub>2.5</sub> from IC analysis (0.14-0.27) is consistent with that obtained from AQB analysis (~0.22), while the  $\kappa$  value for PM<sub>2.5-10</sub> from IC analysis (0.06-0.21) is relatively higher than that from AQB analysis (~0.09) (Table 1 and Fig. 4(a)). The  $\kappa$  differences between the IC and AQB analyses could be attributed to the spatial and temporal variations in aerosols, as well as the different campaign years and locations (~20 km apart, as shown in Fig. S1). These differences might also be influenced by technique uncertainties, such as ammonia and nitrate sampling evaporation during filter sampling (Hering and Cass, 1999; Chen et al., 2021), as well as OPC detection uncertainties and the required parameter assumption in the calculation. Overall, the derived  $\kappa$  values from the OPC data in AQB likely reflect the mean hygroscopicity of both integrated fine and coarse particles. “. Additionally, Fig. S5 is revised as follows:

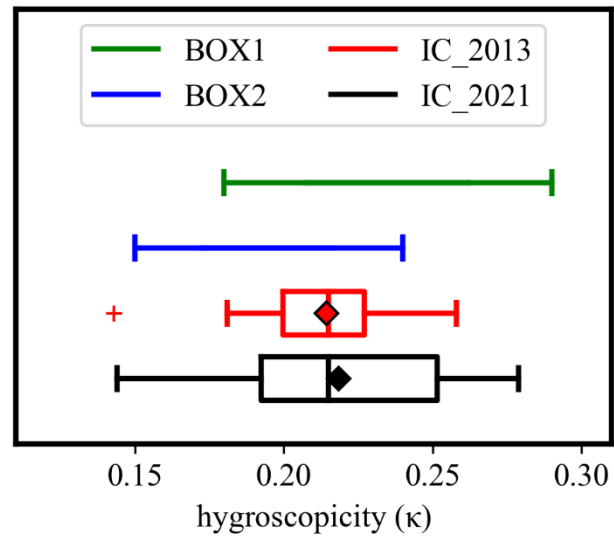


Figure S5: The hygroscopicity of PM<sub>2.5</sub> derived from AQB and IC data with an assumed particle density of 1.2 g cm<sup>-3</sup>. The IC\_2021 is from 2021 samples collected at the National Kaohsiung University of Science and Technology (22°46'22.4" N 120°24'03.4" E) in Kaohsiung for the period of 8 – 18 December 2021 (diamond: mean value; outliers: < 1st quartile Q1-1.5 interquartile range (IQR) or > 3rd quartile Q3+1.5 IQR).

4. please provide how did you convert ppm to mass for various species?

A: For particulate matter, OPC in the AQB and BAM1020 in the TW-EPA station monitor PM mass concentrations in micrograms per cubic meter ( $\mu\text{g}/\text{m}^3$ ); hence, there is no need for unit conversion from ppm to mass concentration. For gaseous species monitored in this study, the concentration is calibrated and expressed in volume mixing ratio as ppm or ppb, the same as EPA data. In our study, conversions from ppm to mass were not performed for gaseous species.

If there is a need for the unit conversion of monitored gas species, it can be calculated using the ideal gas equation as follows:

$$\text{Mass concentration} = C \times \frac{M_{\text{gas}} \cdot P}{R \cdot T}$$

where the unit of *Mass concentration* here uses  $\mu\text{g m}^{-3}$  as an example,  $C$  is the volume mixing ratio (ppmv),  $M_{\text{gas}}$  is the molecular weight of selected gas ( $\text{g mole}^{-1}$ ),  $R$  is the gas constant ( $\text{J mole}^{-1} \text{K}^{-1}$ ),  $T$  is the temperature of air parcel (K), and  $P$  is the pressure of air parcel (Pa). AQB system monitors meteorological parameters temperature (T) and pressure (P), which can convert the gaseous air pollutant concentration from volume mixing ratio to mass concentration. Furthermore, we replaced "ppm" by "ppmv" and "ppb" by "ppbv" to avoid any potential misunderstanding throughout the whole manuscript.

5. please provide your final conclusions in an itemized list.

A: Thank you for your suggestion to present the final conclusions of our study in an itemized list. In this study, we emphasize our study in three main points:

1. Effectiveness of low-cost systems: The performance of home-built Air Quality Box (AQB) systems was evaluated, demonstrating their effectiveness in capturing meteorological parameters and various pollutant concentrations.
2. Sensitivity analysis and hygroscopicity derivation:  $\text{PM}_{10}$  was divided into  $\text{PM}_{2.5}$  and  $\text{PM}_{2.5-10}$  to account for sensor detection sensitivity and compositional heterogeneity among different particle sizes. With the consideration of sensor sensitivity and hygroscopicity of particles, the derived dry particle mass concentrations showed improved consistency with TW-EPA data than those derived from simple linear regression.
3. Derived hygroscopicity and error discussion: The derived hygroscopicity values align with results from the soluble composition analysis using ion chromatography. This study also emphasizes the need for careful consideration of uncertainties and calibration techniques to interpret low-cost sensor data in atmospheric research accurately.

We prefer to choose the narrative conclusion section by reconstructing the logic in revision: " In this study, we evaluated the performances of home-built Air Quality Box (AQB) systems equipped with low-cost sensors and focused on the ambient variability of particulate matter (PM) concentrations to derive the hygroscopicity of PM and the conversion to dry particle concentrations. The AQB systems revealed their effectiveness in capturing meteorological parameters and most pollutant concentrations with high

correlations ( $r \geq 0.96$ ) for temperature, relative humidity, CO, and Ox ( $O_3 + NO_2$ ) and moderate correlations ( $r \geq 0.48$ ) for NO<sub>x</sub> and PM, as compared to TW-EPA data. In the PM analysis, PM<sub>10</sub> was divided into PM<sub>2.5</sub> and PM<sub>2.5-10</sub> to account for compositional heterogeneity among different particle sizes. Comparing the AQB-monitored ambient PM data and the TW-EPA data (for dry particles) at  $RH \leq 50\%$ , the derived sensitivity coefficients ( $\alpha$ ) for PM<sub>2.5-10</sub> (10.58 - 12.37) were higher than those for PM<sub>2.5</sub> (1.26 - 1.44) likely due to the significant sensitivity variation in the OPC over time. By considering hygroscopicity with the  $\kappa$ -Köhler equation and assuming a constant composition density for sensitivity-corrected AQB data, the derived dry particle mass concentrations show improved consistency with TW-EPA data compared to the simple linear regression approach. The derived  $\kappa$  values range from 0.15 to 0.29 for PM<sub>2.5</sub> and 0.05 to 0.13 for PM<sub>2.5-10</sub>, consistent with those from IC soluble composition analysis (0.14 to 0.27 for PM<sub>2.5</sub> and 0.06 to 0.21 for PM<sub>2.5-10</sub>) and primarily influenced by the proportion of soluble components, ~53% in PM<sub>2.5</sub> and ~30% in PM<sub>2.5-10</sub>. The sensitivity analysis of various parameters showed that the effects of chosen deliquescence relative humidity (DRH) thresholds and Kelvin effects had a minor impact on  $\kappa$  values (less than 1%). Conversely, recalculating particle densities for PM<sub>2.5</sub> ( $1.42 \pm 0.03 \text{ g cm}^{-3}$ ) and PM<sub>2.5-10</sub> ( $1.34 \pm 0.07 \text{ g cm}^{-3}$ ) led to higher  $\kappa$  values by approximately 17% and 9%, respectively, compared to the results assuming  $1.2 \text{ g cm}^{-3}$ . Overall, the AQB systems are helpful in understanding the temporal and spatial variability of air quality by effectively monitoring pollutant concentrations and providing the capability for hygroscopicity derivation. This study also emphasizes the need for careful consideration of uncertainties and calibration techniques to accurately interpret low-cost sensor data in atmospheric research.”

## RC2

**We would like to thank the anonymous reviewer for the comments that significantly improve the clarity and readability of the manuscript. Our point-by-point responses are found below in blue ink. The revised content is highlighted in yellow.**

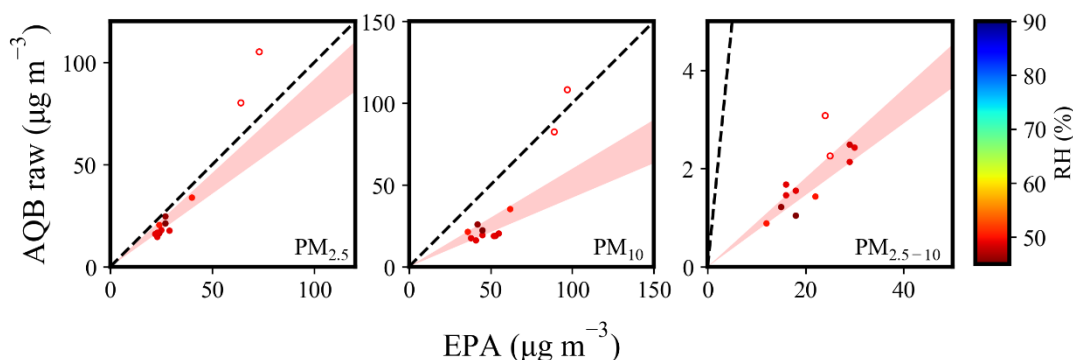
General Comments:

The normalization and portability of atmospheric pollutant monitoring are crucial for in-depth research into local variations in atmospheric environment and pollution. This manuscript establishes a low-cost air quality monitoring device and applies it to estimate the hygroscopicity parameters of aerosols, comparing the results with site observation data. The study holds certain scientific and application value and aligns with the publication scope of the Atmospheric Measurement Techniques journal. However, there are some scientific and technical issues within the manuscript itself, suggesting major revisions before reconsidered.

Specific Comments:

1) The accuracy and error range of AQB in detecting aerosols and the application of AQB to estimate aerosol hygroscopicity parameters should be two separate research components with a sequential order. However, in this manuscript, the authors often fail to clearly distinguish between the two. The authors adopt a method of setting RH thresholds to classify AQB observation results into dry aerosols and humidified aerosols, and then compares and calibrates the observations of dry aerosols with EPA station data, which is a feasible approach. However, in Sections 3.1, 3.2, and Figures 2 and 3, the authors do not classify or analyze the data based on the RH threshold set by themselves. Meanwhile, as shown in Figure 2(b), the occurrence of RH below 50% during the observation period is rare. Can such a limited amount of data support the examination of the reliability of AQB detection?

A: We appreciate the reviewer's insightful comments. In response to concerns, Fig R1, similar in configuration to Figs. 3(a-c), shows the data points at  $RH \leq 50\%$ , which were applied to determine the sensitivity coefficient ( $\alpha$ ).



**Figure R1.** The correlation of mass concentration between TW-EPA and OPC in AQB #1 (raw data) for  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ , and  $\text{PM}_{2.5-10}$  at  $RH \leq 50\%$ . The hollow points are the two significant outliers mentioned in Table 2.

Although these data points constitute about 5% of the total (i.e., 17 out of 356 points), a high correlation between the AQB and TW-EPA measurements was observed. Figure 2(b) shows only part of the campaign to reveal the temporal comparison between AQB and TW-EPA data. As stated in section 2.3, the statistical distribution of  $M_{EPA}$  to  $M_{OPC}$  ratios at  $RH \leq 50\%$  was analyzed to evaluate a sensitivity coefficient ( $\alpha$ ) as the mean value  $\pm 0.5\sigma$  ( $\sigma$ : standard deviation). The shaded area in Fig. R1 represents the distribution of  $\alpha$ , covering most of the data points at  $RH \leq 50\%$ . The same calculation but for higher RH thresholds (up to 60% to have more data points, 51 out of 356) summarized below shows a similar  $\alpha$  range, indicating sufficient data points at  $RH \leq 50\%$  for a conclusive  $\alpha$ . The following figure is added to the supplementary as Fig. S2 to clarify this issue. In the content, Lines 179-181 were revised as follows: “The estimated  $\alpha$ , as summarized in Table 1, are higher for  $\text{PM}_{10}$  than for  $\text{PM}_{2.5}$ , i.e.,  $2.02 \pm 0.34$  vs  $1.26 \pm 0.16$ , which are reasonably conclusive as tested with more data points selected at higher RH thresholds (Fig. S2).”

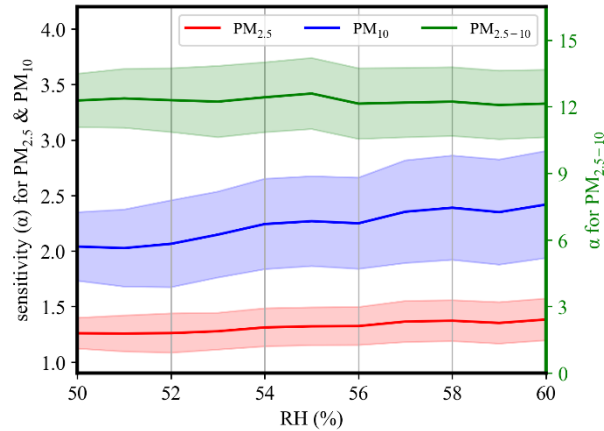


Figure S2: The determined sensitivity as a function of RH thresholds for PM<sub>2.5</sub> (red), PM<sub>10</sub> (blue) and PM<sub>2.5-10</sub> (green). The shading area is the mean value  $\pm 0.5\sigma$

As to the accuracy of applied methods, the comparison analysis between fitted data and TW-EPA data is summarized in a new table (Table 2) added to the content as follows:

Table 2: Performance metrics of different calibration methods for PM<sub>2.5</sub>, PM<sub>2.5-10</sub>, and PM<sub>10</sub>.

	PM <sub>2.5</sub>			PM <sub>2.5-10</sub>			PM <sub>10</sub>			
	RH≤50% Only <sup>a</sup>	All data (no κ)	All data (κ= 0.29)	RH≤50% Only <sup>a</sup>	All data (no κ)	All data (κ= 0.09)	RH≤50% Only <sup>a</sup>	All data (no κ)	All data (κ= 0.36)	(PM <sub>2.5</sub> +PM <sub>2.5-10</sub> ) <sup>c</sup>
applied α	1.26±0.16	1.04	1.40	12.37±1.33	10.77	13.16	2.02±0.34	1.69	2.36	—
MAPE (%)	21.3 (12.8)	48.8	24.8	15.9 (11.5)	37.9	31.8	32.8 (18.5)	62.5	29.2	18.2
RMSE (μg cm <sup>-3</sup> )	20.5 (3.7)	29.1	11.3	4.9 (2.8)	9.4	9.1	42.6 (10.3)	54.7	26.9	15.9
R <sup>2</sup> <sup>b</sup>	0.55 (0.51)	-3.49	0.32	0.31 (0.78)	0.57	0.59	-4.18 (-0.58)	-4.74	-0.38	0.51

<sup>a</sup> Only for data points at RH ≤ 50%. The value in parentheses is the performance result without two significant outliers shown in Fig. 3

<sup>b</sup> Coefficient of determination (R<sup>2</sup>) is calculated as the proportion of variation in the calibrated dry mass concentration.

<sup>c</sup> The combination of calibrated data from PM<sub>2.5</sub> All data (κ= 0.29) and PM<sub>2.5-10</sub> All data (κ= 0.09).

The MAPE and RMSE for RH ≤ 50 % are 12.8 % and 3.7 μg cm<sup>-3</sup> for PM<sub>2.5</sub>, 11.5 % and 2.8 μg cm<sup>-3</sup> for PM<sub>2.5-10</sub>, without considering the two significant outliers. The performance of AQB for PM<sub>2.5</sub> and PM<sub>10</sub> through different calibration methods was discussed in Section 3.2 (Lines 172-211) as “Figures 3(a) and 3(c) show the scatter distribution of the mass concentrations between AQB #1 (with no calibration) and TW-EPA data for PM<sub>2.5</sub> and PM<sub>10</sub>, respectively. Overall, the PM mass concentrations measured by AQB system appear to be higher than those reported by TW-EPA. The results reveal an apparent influence of ambient RH, indicating the contribution of water content. The red-shaded area represents a regression line with a slope corresponding to the inverse of the sensitivity coefficients (α) derived from data points at ambient RH ≤ 50% (17 out of 356 points, 5%). The notable deviation of the red shaded area from the 1:1 line towards the right side indicates the requirement of α > 1 corrections, contributed by the different measurement principles and



calibration techniques, which may result from the assuming particle density and refractive index (RI) (dust, density:  $1.65 \text{ g cm}^{-3}$ , RI:  $1.5 + 0i$ ). The estimated  $\alpha$ , as summarized in Table 1, are higher for  $\text{PM}_{10}$  than for  $\text{PM}_{2.5}$ , i.e.,  $2.02 \pm 0.34$  vs  $1.26 \pm 0.16$ , which are reasonably conclusive as tested with more data points selected at higher RH thresholds (Fig. S2). The  $\alpha$  difference between  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  might be attributed to the complex composition of ambient particles, which differs from the samples used for instrument calibration, as well as possible sensitivity variations in OPC over time. With sensitivity calibration, the performance at ambient  $\text{RH} \leq 50\%$  exhibits a strong correlation with MAPE at 12.8%, 18.5%, and Root Mean Squared Error (RMSE) at  $3.7 \mu\text{g m}^{-3}$ ,  $10.3 \mu\text{g m}^{-3}$  for  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ , respectively, as summarized in Table 2 excluding the two significant outliers (shown as hollow circles in Fig. 3). The results confirm the effectiveness of OPCs in capturing PM concentrations, consistent with previous real-time outdoor field studies (Gillooly et al., 2019; Demanega et al., 2021; Sá et al., 2022; Crilley et al., 2018). Additionally, the OPC sampling flow rate has an impact on measurement performance. AQB #1 maintained a steady rate at  $3.6 \pm 0.2 \text{ LPM}$ , whereas AQB #2 exhibits two distinct time periods with sampling flow rates of 3.6-4.2 LPM for the first period and 3.2-3.6 LPM for the second period. ... With the derived  $\alpha$ , the hygroscopicities were retrieved using Eq. (3), resulting in  $\kappa$  ranging from 0.18 to 0.29 for  $\text{PM}_{2.5}$  and 0.20 to 0.39 for  $\text{PM}_{10}$  (Table 1) during the studied period. Figures 3(d) and 3(f) show the scatter distribution of the derived dry concentration vs. TW-EPA concentration for  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ , respectively. The results from the two AQB systems exhibit slight differences but are consistent overall. Considering both the sensitivity coefficient and hygroscopicity, the performance of AQB in deriving dry PM concentration is significantly improved with lower MAPE, RMSE, and higher  $R^2$  than the results obtained using only the sensitivity coefficient, as summarized in Table 2. ... The lower  $\kappa$  for  $\text{PM}_{2.5-10}$  might suggest a significant contribution from dust or other less hygroscopic species, consistent with the IC analyses in Table 3 and discussed further in Sect. 3.3. With the retrieved  $\alpha$  and  $\kappa$  for  $\text{PM}_{2.5}$  and  $\text{PM}_{2.5-10}$ , Fig. 3(e) shows the scatter distribution between the derived dry  $\text{PM}_{2.5-10}$  from AQB data and TW-EPA data, exhibiting a MAPE of 31.8%, more significant than the 24.8% for  $\text{PM}_{2.5}$ . ... Detection efficiency may be influenced by notable spatial variations, aligning with the findings of Kaliszewski et al. (2020), which showed a reduced correlation between OPC-N3 measurements and reference instruments for larger particles. The dry  $\text{PM}_{10}$  derived from AQB through the divided  $\text{PM}_{2.5}$  and  $\text{PM}_{2.5-10}$  analysis demonstrates better consistency with the reported TW-EPA data than the direct calibration method. This is evidenced by a lower MAPE in Fig. 3(g) (18.2%) compared to Fig. 3(f) (29.2%) and a significant improvement than the simple linear regression method, which has a higher MAPE at 62.5% (Table 2). This substantiates the importance of considering composition heterogeneity among particle sizes for accurate dry PM derivation.”.

2) Introducing aerosol chemical composition observations into a thermodynamic equilibrium model to calculate aerosol hygroscopic growth and comparing it with optical observations is a common research approach. However, contrasting different field experiments conducted at different times (with an 8-year difference) and different underlying surfaces by the authors doesn't have much significance.

A: Thank you for highlighting the concerns regarding the comparison of derived hygroscopicity between two different field studies conducted in different years. We acknowledge that the hygroscopic characteristics of ambient particles can vary spatially and temporally. Our comparison of the chemical composition of PM<sub>2.5</sub> from two winter sampling campaigns in 2013 and 2021 revealed that the composition concentration might be different, but the derived hygroscopicity was consistent across these years, demonstrating typical ambient PM<sub>2.5</sub> hygroscopic characteristics in Kaohsiung City during winter. Even though there was no intensive filter sample collection during the studied period, the derived mean  $\kappa$  range under the specific assumption (solute density and ignorance of Kelvin effect) from AQB data was discussed for the range consistency compared to the results of available two winter campaigns for PM<sub>2.5</sub> and 2013 winter campaign for PM<sub>2.5-10</sub>. However, the temporal resolution for the derived hygroscopicity from IC data is higher than that derived from AQB data since the AQB analysis required a longer time period to cover a comprehensive range of RH to have the particle growth profile. The derived hygroscopicity then represents a mean value over a longer period. To clarify this approach, Section 3.3 (Lines 222-235): “A similar analysis for the winter of 2021 yielded a consistent  $\kappa$  range for PM<sub>2.5</sub>, as illustrated in Fig. S5. This consistency across distinct study periods indicates typical ambient PM<sub>2.5</sub> hygroscopic characteristics in Kaohsiung City during winter, which can be applied for further discussion with the AQB data. For coarse particles, the more significant variability in  $\kappa$  for PM<sub>2.5-10</sub> compared to PM<sub>2.5</sub> can be attributed to the significant fluctuations in the soluble composition of coarse particles, primarily driven by substantial quantities of thenardite (Na<sub>2</sub>SO<sub>4</sub>) and halite (NaCl) (Tang et al., 2019). ...The derived  $\kappa$  value for PM<sub>2.5</sub> from IC analysis (0.14-0.27) is consistent with that obtained from AQB analysis (~0.22), while the  $\kappa$  value for PM<sub>2.5-10</sub> from IC analysis (0.06-0.21) is relatively higher than that from AQB analysis (~0.09) (Table 1 and Fig. 4(a)). The  $\kappa$  differences between the IC and AQB analyses could be attributed to the spatial and temporal variations in aerosols, as well as the different campaign years and locations (~20 km apart, as shown in Fig. S1). These differences might also be influenced by technique uncertainties, such as ammonia and nitrate sampling evaporation during filter sampling (Hering and Cass, 1999; Chen et al., 2021), as well as OPC detection uncertainties and the required parameter assumption in the calculation. Overall, the derived  $\kappa$  values from the OPC data in AQB likely reflect the mean hygroscopicity of both integrated fine and coarse particles. “. Additionally, the corrected version of Fig. S5 is shown as:

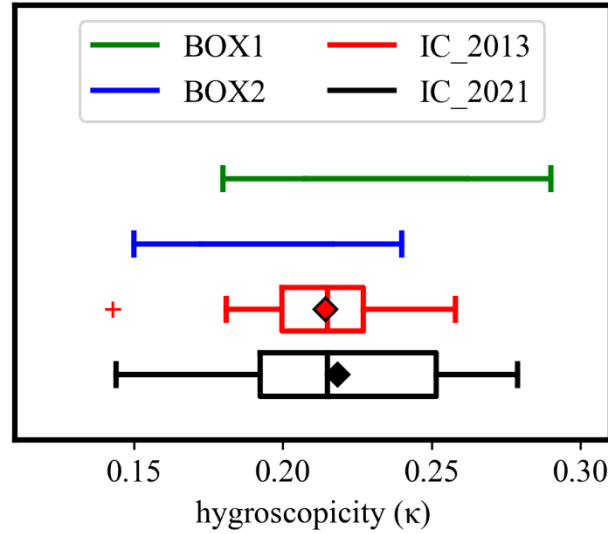


Figure S5: The hygroscopicity of PM<sub>2.5</sub> derived from AQB and IC data with an assumed particle density of 1.2 g cm<sup>-3</sup>. The IC\_2021 is from 2021 samples collected at the National Kaohsiung University of Science and Technology (22°46'22.4" N, 120°24'03.4" E) in Kaohsiung for the period of 8 – 18 December 2021 (diamond: mean value; outliers: < 1st quartile Q1-1.5 interquartile range (IQR) or > 3rd quartile Q3+1.5 IQR).

3) In Table 1, the hygroscopicity parameter kappa for PM<sub>2.5</sub> and PM<sub>2.5-10</sub> is smaller than the hygroscopicity parameter kappa for PM<sub>10</sub>, which is abnormal. PM<sub>10</sub> is the sum of PM<sub>2.5</sub> and PM<sub>2.5-10</sub>, and its hygroscopicity should be intermediate between the two.

A: The higher hygroscopicity of PM<sub>10</sub> than those of PM<sub>2.5</sub> and PM<sub>2.5-10</sub> is due to a significantly high portion PM<sub>2.5</sub> in PM<sub>10</sub> observed in AQB combined with the positive correlation between the derived sensitivity ( $\alpha$ ) and hygroscopicity ( $\kappa$ ), as shown in Eq. (3) for the ambient and dry PM mass concentration conversion.

$$M_{d,derived} = (\alpha \times M_{OPC}) \times \left[ \left( \frac{S\kappa}{1-S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1} \quad (3)$$

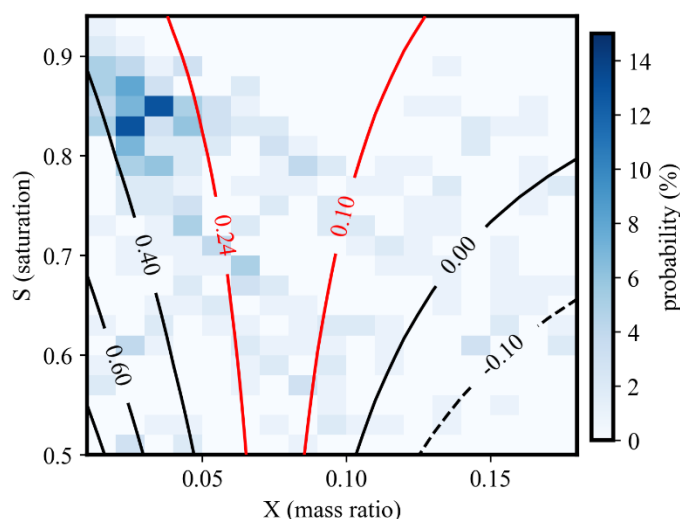
For a given  $M_{opc}$ , a higher  $\alpha$  would require a higher  $\kappa$  to have the same  $M_{d, derived}$ . Because the estimated  $\alpha$  for PM<sub>10</sub> is higher than that for PM<sub>2.5</sub>, a higher  $\kappa$  might be expected if PM<sub>2.5</sub> portion is dominant in PM<sub>10</sub>. This can be evaluated through the following calculation using an ideal system. The equation in the following shows the derived dry PM<sub>10</sub> concentration is the sum of the derived PM<sub>2.5</sub> and the derived PM<sub>2.5-10</sub> concentration.

$$\begin{aligned} (\alpha_{10} \times M_{10}) \times \left[ \left( \frac{S\kappa_{10}}{1-S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1} \\ = (\alpha_{2.5} \times M_{2.5}) \times \left[ \left( \frac{S\kappa_{2.5}}{1-S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1} + (\alpha_{2.5-10} \times M_{2.5-10}) \times \left[ \left( \frac{S\kappa_{2.5-10}}{1-S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1} \end{aligned}$$

where  $M$  is  $M_{opc}$  for different size ranges (indicated in the subscript). By assuming  $X$  as the ratio of  $M_{2.5-10}$  to  $M_{2.5}$  monitored by AQB, the relationship can be rewritten in the following:

$$\alpha_{10}(1 + X) \times \left[ \left( \frac{S\kappa_{10}}{1 - S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1} = \alpha_{2.5} \times \left[ \left( \frac{S\kappa_{2.5}}{1 - S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1} + (\alpha_{2.5-10} \times X) \times \left[ \left( \frac{S\kappa_{2.5-10}}{1 - S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1}$$

where the  $\rho_w$  and  $\rho_d$  are constants (assumed in this study), the sensitivity coefficients are given as  $\alpha_{2.5} = 1.26$ ,  $\alpha_{10} = 2.02$ , and  $\alpha_{2.5-10} = 12.37$  from Table 1, and hygroscopicity is given as a median value in derived results of PM<sub>2.5</sub> and PM<sub>2.5-10</sub> ( $\kappa_{2.5} = 0.24$ , and  $\kappa_{2.5-10} = 0.10$ ). The derived  $\kappa_{10}$  can be evaluated with a function of  $X$  and  $S$  in the following figure and is mainly affected by  $X$ . A higher  $\kappa_{10}$  than  $\kappa_{2.5}$  (0.24) is expected as  $X < 0.05$ , i.e., low  $M_{2.5-10}/M_{2.5}$ . The observed data from AQB has a higher probability of having  $X \sim 0.04$  with  $S \sim 0.8-0.9$  which leads to a higher derived  $\kappa_{10}$  than  $\kappa_{2.5}$  (0.24).



**Figure R2.** The distribution of the derived hygroscopicity of PM<sub>10</sub> in the condition of given sensitivity coefficients ( $\alpha_{2.5} = 1.26$ ,  $\alpha_{10} = 2.02$ , and  $\alpha_{2.5-10} = 12.37$ ) and hygroscopicity ( $\kappa_{2.5} = 0.24$ , and  $\kappa_{2.5-10} = 0.10$ ). The contour is the derived hygroscopicity of PM<sub>10</sub>, and the shading is the data point distribution of the AQB monitored data.

Overall, the estimated higher derived  $\kappa_{10}$  is possible to happen due to a significant amount of data points having a low portion PM<sub>2.5-10</sub> in PM<sub>10</sub> observed in AQB.

4) In Section 2.1, the author introduces the photo-ionization detector for monitoring VOCs, but in Figure 1 and subsequent manuscripts, the abbreviation used by the author is NMHC. These two abbreviations are not entirely equivalent.

The Alphasense PID-AH2 measures volatile organic compounds (VOCs) in the air using photoionization detection (PID), as stated in its datasheet. This device utilizes a lamp that emits high-energy UV photons. When a VOC molecule absorbs a photon, it generates electrically charged ions, creating an electric field. The detector then monitors the resulting current, which is proportional to the ambient VOC concentration. Notably, the PID-AH2 in this study uses a Krypton lamp with a photon energy of about 10.6 eV, capable of detecting some C2, and most C3, C4+ VOCs. In contrast, the Horiba APHA-360, a VOC gas analyzer used by TW-EPA, continuously analyzes THC, CH<sub>4</sub>, and non-CH<sub>4</sub> (NMHC) in ambient air using a flame ionization detector and cross flow modulation. Since the

ionization potential of methane is approximately 13.7 eV, the PID cannot detect its concentration, making NMHC the closest comparable data from TW-EPA for our purposes. Additionally, the sensitivity of the PID varies with the type of VOC detected; for example, toluene generates approximately twice the response of isobutylene. Consequently, as shown in Fig. 2(g), the sensor captures some peak NMHC concentrations, but not all temporal variations are detectable. To ensure consistency, the label in Fig.1 is revised as "VOCs". Furthermore, to avoid any potential confusion regarding the capabilities of the PID sensor, the following information is added in Section 2.1 (Lines 71-73): " The PID sensor, equipped with a Krypton lamp providing a photon energy of about 10.6 eV, cannot detect methane, which has a higher ionization potential of ~13.7 eV (Glockler, 1926). Therefore, the data of non-methane hydrocarbons (NMHC) from TW-EPA is more comparable to PID data in our analysis."

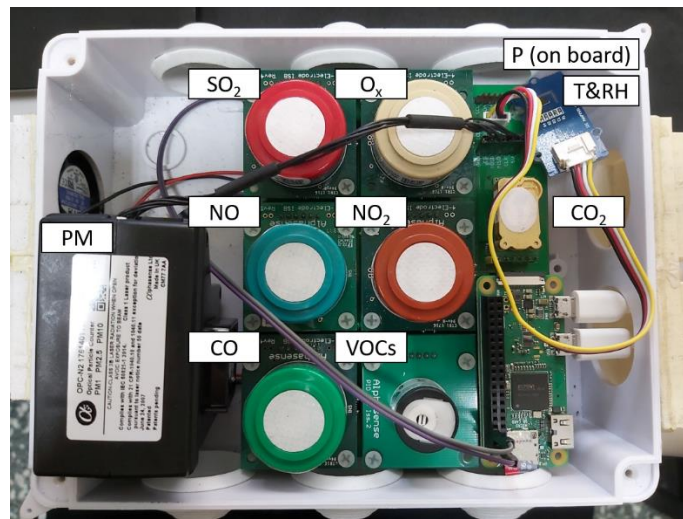


Figure 1: The design of the AQB system.