**We would like to thank the editor and 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.**

**Part1.**

# **Responses to editor comments:**

Public justification (visible to the public if the article is accepted and published):

This manuscript presents a novel attempt to use low-cost sensors to determine particle hygroscopicity. It requires an additional minor review to address remaining comments by both the reviewers and the editor.

Additional private note (visible to authors and reviewers only):

1. Both reviewers have additional comments that I think need to be addressed by the authors. Referee #2 would like to see further discussion of errors in the calculated hygroscopicity.

A: Please find our response to RC2 listed in part 2.

2. Referee #3 has several comments, including a better discussion of the setup of the AQB.

A: Please find our point-by-point responses to RC3 (Q1-Q3) listed in part 3.

3. Referee #3 also notes that parts of the manuscript are difficult to follow.

A: With the comment (Q5) from Referee #3, the content has been revised. The results and discussion section was re-arranged to improve the logicality and clarification as follows (the content was revised but not shown here): "

3 Results and Discussion 3.1 Performance of AQB systems 3.2 Comparison between OPC and BAM data 3.2.1 Sensitivity coefficient of OPC 3.2.2 Hygroscopicity derivation 3.3 Hygroscopicity derivation using IC data 3.3.1 Composition analysis 3.3.2 E-AIM analysis 3.4 Sensitivity of assumed parameters on derived hygroscopicity"

4. I would like to echo Referee #3's comments and add several of my own. This manuscript has many abbreviations and acronyms, which can make it hard to follow. A glossary would be helpful. At a minimum, the authors need to define each abbreviation or acronym the first time that it it appears in the manuscript. For example, AQB appears in the abstract but needs to be defined again in the main text.

A: Thank you for your suggestion. The content has been revised to ensure that all abbreviations are introduced after their full names when they appear for the first time in the main content. Additionally, a list of principle symbols and abbreviations is provided in Lines 24-60 for clarification.

5. I also share Referee #3's confusion about how the OPC and regulatory data are compared. The manuscript briefly describes a calibration for the OPC from co-location with the BAM (section 2.2). The subsequent hygroscopicity analysis seems to use the calibrated OPC data and compares it to the (dry) PM measured by the BAM. Thus, I don't understand how the comparison between the OPC and BAM can be used both for calibration and the calculation of kappa. The authors should clarify exactly what process they are using at each step.

A: To clarify the calibration process, a detailed description was added to supplementary material as follows:

#### **"Aerosol hygroscopicity derived using OPC and BAM data**

The optical particle counter (OPC) (model: OPC-N2, Alphasense) provides digital outputs of  $PM_1$ ,  $PM_{2.5}$ ,  $PM_{10}$ , and optionally  $PM_{4.25}$ , along with histograms of the particle counts for 16 size bins ranging from 0.38 to 17 μm. This device is designed to monitor ambient aerosol concentrations without any drying system attached to the sampling inlet. In contrast, the Beta Attenuation Mass Monitor (BAM) (model: BAM1020, Met One Instrument) is designed to monitor dry particle mass concentrations of  $PM<sub>2.5</sub>$  and  $PM_{10}$ , using a heating device to ensure the sampling relative humidity (RH) remains below 50%. If the RH of the sampled air stream exceeds 50%, the inlet heater activates, reducing the RH to approximately 35% by warming the air stream downstream before reaching the filter tape. If the RH is below 50%, the heater remains inactive, not altering the sampling flow RH. The technical specifications for the OPC and BAM are summarized in Table S4.

To derive aerosol hygroscopicity  $(\kappa)$ , the sensitivity coefficient of OPC was evaluated first using the data points at RH  $\leq$  50%, as described in Section 2.3 of the main content. Depending on the size range,  $\alpha_{2.5}$ ,  $\alpha_{10}$ , and  $\alpha_{2,5-10}$  represent the sensitivity coefficient of OPC for PM<sub>2.5</sub>, PM<sub>10</sub>, and PM<sub>2.5-10</sub>, respectively. For PM10, a range of hygroscopicity of 0 to 1.2 was applied to Eq. (S1) to obtain *Md,derived,10* and evaluate the mean absolute percentage error (MAPE) between *Md,derived,10* and *MBAM,10*. MAPE as a function of the applied hygroscopicity is plotted to determine the  $\kappa_{10}$  range, which has MAPE  $\leq 1.1 \times$ the lowest MAPE. considering the uncertainty. A similar calculation is applied to  $PM_{2.5}$ , and  $PM_{2.5-10}$  to derive  $\kappa_{2.5}$  and  $\kappa_{2.5}$ <sup>10</sup>, directly.

$$
M_{d,derived,10} = \alpha_{10} \times M_{OPC,10} \times \left[ \left( \frac{S \kappa_{10}}{1 - S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1}
$$
 (S1)

However,  $PM_{10}^*$  in Table 1 considers the size-dependent sensitivity with a mean hygroscopicity.  $\kappa_{10}$  is derived using the following equation:

$$
M_{d,derived,10} = (\alpha_{2.5} \times M_{OPC,2.5} + \alpha_{2.5-10} \times M_{OPC,2.5-10}) \times \left[ \left( \frac{S \kappa_{10}}{1-S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1}
$$
 (S2)

The derived  $\kappa_{10}$  using Eq. (S2) ranges from 0.13 to 0.23, falling between κ values for PM<sub>2.5</sub> and PM<sub>2.5-10</sub>, and is more reasonable than from Eq. (S1), as discussed in Section 3.2.2."

# **Part 2.**

### **Responses to referee#2's comments:**

1. The authors have provided a detailed point-by-point response to the previous round of review comments, resolving most of the issues. Regarding specific comment (3), the authors' response explained the reason why the kappa value of  $PM_{10}$  is higher than those of  $PM_{2.5}$  and  $PM_{2.5-10}$ . However, this explanation also indicates that there is a significant error in estimating aerosol hygroscopicity using AQB detection results, as the estimation does not align with the real atmospheric conditions. This error might originate from the process of calculating the derived sensitivity, such as whether the 50% threshold is too high, or it could be due to the derived sensitivity under dry conditions not being well applicable to humid conditions. The authors should thoroughly analyze the causes of this error and its magnitude in the manuscript.

A: The higher kappa value for  $PM_{10}$  compared to  $PM_{2.5}$  and  $PM_{2.5-10}$  in Table 1 is likely due to significant sensitivity difference between  $PM_{2.5}$  and  $PM_{2.5-10}$  in our system and the simplified calculation using one sensitivity coefficient for the overall PM<sub>10</sub>.  $\kappa$  for PM<sub>10</sub> ( $\kappa_{10}$ ) is derived using the following equation:

$$
M_{d,derived,10} = \alpha_{10} \times M_{OPC,10} \times \left[ \left( \frac{S \kappa_{10}}{1-S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1}
$$
 (R1)

where the parameters with subscript 10 stand for  $PM_{10}$ . As  $PM_{10}$  is divided into  $PM_{2.5}$  and  $PM_{2.5-10}$ , the different estimated sensitivity coefficients suggest that the response of the applied OPC is size-dependent. By applying the more sophisticated sensitivity correction,  $(\alpha_{2.5} \times M_{QPC,2.5} + \alpha_{2.5-10} \times M_{QPC,2.5-10})$  represents the ambient  $PM_{10}$  concentration. The single hygroscopicity parameter of  $PM_{10}$  can be estimated using the following equation:

$$
M_{d, derived, 10} = (\alpha_{2.5} \times M_{OPC, 2.5} + \alpha_{2.5-10} \times M_{OPC, 2.5-10}) \times \left[ \left( \frac{S \kappa_{10}}{1 - S} \right) \times \frac{\rho_w}{\rho_d} + 1 \right]^{-1}
$$
 (R2)

The derived kappa using Eq. (R2) ranges from 0.13 to 0.23, falling between κ values for  $PM_{2.5}$  and  $PM_{2.5}$ <sup>10</sup>. However, because aerosols are inhomogeneous among different particle sizes, we focus on addressing different hygroscopicities between fine and coarse particles and verifying the retrieved values with other methods. OPC has become an affordable aerosol detector and is widely applied for environment monitoring. If the sensitivity is the same across different particle sizes, Eq. (R1) would be sufficient for related calculations. Otherwise, detailed analyses with separated size ranges would provide more comprehensive results. A more detailed deriving process was added to the supplementary material.

Table 1 is revised to include the results retrieved using Eq. (R2) as follows:

	Sensitivity coefficient $(\alpha)$		Hygroscopicity $(\kappa)$			
	$AOB$ #1	$AOB$ #2*	$AOB \#1$	$AOB$ #2	IC (species)	$IC$ (E-AIM)
$PM_2$	$1.26 \pm 0.16$	$1.44 \pm 0.20$	$0.18 - 0.29$	$0.15 - 0.24$	$0.14 - 0.27$	$0.14 - 0.26$
$PM_{10}$	$2.02 \pm 0.34$	$2.20 \pm 0.38$	$0.20 - 0.39$	$0.18 - 0.30$		
$PM_{10}$ <sup>#</sup>	$\alpha_{2.5}, \alpha_{2.5-10}$	$\alpha_{2.5}, \alpha_{2.5-10}$	$0.13 - 0.23$	$0.11 - 0.26$		
$PM_{2.5-10}$	$12.37 \pm 1.33$	$10.58 \pm 2.90$	$0.07 - 0.13$	$0.05 - 0.09$	$0.06 - 0.21$	$0.08 - 0.21$

**Table 1: The sensitivity coefficients and the hygroscopicity for PM2.5, PM<sup>10</sup> and PM2.5-10.**

**# the hygroscopicity derived using different sensitivity coefficients for different size ranges. α2.5 and α2.5-10 are sensitivity coefficients for PM2.5 and PM2.5-10, respectively. More details are provided in the description of the supplementary material. \* the sensitivity of AQB #2 presents the value in the period of sampling flow rates at 3.6-4.2 LPM**

The content of section 3.2.2 (Lines 254-260) is revised to include this concept as follows:

"The dry  $PM_{10}$  derived from OPC through the divided  $PM_{2,5}$  and  $PM_{2,5-10}$  analysis demonstrates better consistency with the reported BAM 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). Moreover, the derived  $\kappa$  for PM<sub>10</sub> with the size-dependent sensitivity coefficient correction ranges from 0.13 to 0.23 (Table 1). This value falls between those for  $PM_{2.5}$  and  $PM_{2.5-10}$  and is more reasonable compared to  $\kappa$  derived with a fixed sensitivity coefficient ( $\kappa = 0.20$ -0.39, higher than those for PM<sub>2.5</sub> and PM<sub>2.5-10</sub>). The results substantiate the importance of considering composition heterogeneity among particle sizes for accurate dry PM derivation. "

# **Part 3.**

# **Responses to referee#3's comments:**

The submitted manuscript presents results of hygroscopicity measurements using an inexpensive set of aerosol analyzers. The work comprehensively describes the measurement results and is very valuable. However, there are several issues that need clarification and elaboration.

Specific Comments:

1. The manuscript lacks a more detailed description of the measurement system. Although the authors presented a photograph of the interior of the setup and a brief description of the devices, there is a lack of description of the sampling system. Did the devices independently sample the measured air from their own inlets, or were they equipped with a common measurement line? I am unable to assess this because no photograph of the setup in its operational state was presented. Furthermore, the placement of the device in relation to the inlet of the reference measurement station is crucial. The effects of boundary layer and local turbulent flows in urbanized areas can significantly influence aerosol measurement results. I believe this should be discussed in greater detail.

A: The home-built Air Quality Box (AQB) system samples the air independently with a fan visible in the upper left of Fig. 1. Additionally, the optical particle counter (OPC-N2) has its own fan ambient air sampling. On the opposite side of the sampling fan, AQB has two openings for air to flow out. The selected TW-EPA station is situated on the roof of a building in a well-ventilated environment. Because the TW-EPA station has a standard operation process to ensure consistency between stations, our system can only be set aside the station (approximately 5 m horizontally and 2 m vertically from the sampling inlet of TW-EPA station) with a photo shown in Figure S1(b). The content of Section 2.1 (Lines 117-121) was revised to describe more detail of the AQB system as follows:

"The entire system is housed in a remodeled enclosure with a dimension of 25 cm  $\times$  16 cm  $\times$  8 cm (L  $\times$  $D \times H$ ) and has well-ventilated openings for sampling and exhaust. The sampling flow rate is primarily controlled by an installed fan at  $\sim 5.6$  L min<sup>-1</sup>, corresponding to a residence time of approximately 34 s in the box. This configuration allows the system to effectively monitor ambient air quality independently without the need of additional inlets."

The other revised paragraph is shown in Section 2.2 (Lines 123-126) as follows:

" The calibration of AQB sensors was carried out by co-locating them with TW-EPA Nanzi station (Fig. S1) in Kaohsiung, Taiwan (22°44'12" N, 120°19'42" E) from 4 to 19 February 2021. Nanzi station is situated on the roof of a 15 m high building in a well-ventilated environment. The primary gaseous components, dry PM2.5 and PM<sup>10</sup> concentrations, and basic meteorological parameters are continuously monitored using standard instruments, as summarized in Table S1."

Additionally, Fig. 1 was revised to include the information of fans to emphasize the sampling flow as follows:



Figure 1: The design of the air quality box (AQB) system.

For the influence of boundary layer and local turbulent flows, the performance of CO monitoring exhibits a high correlation (r=0.976) between AQB and TW-EPA, as depicted in Fig. 2(c), indicating that the two systems are co-located in a similar environment. Therefore, the effects of boundary layer and local turbulent flows in urbanized areas can be assumed negligible for the comparision between OPC and BAM data. The content of section 3.1 (Lines 199-202) was revised as follows:

"Figure 2 shows the time series of the meteorological parameters and pollutant concentrations between calibrated AQB and TW-EPA data from 14 to 17 February 2021. T, RH, CO, and Ox showed a good correlation with  $r > 0.9$ , while NO, NO<sub>2</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> had a moderate correlation ( $r \ge 0.48$ ). The high correlation ( $r=0.976$ ) for CO (with a lifetime of  $\sim 2$  months) indicates a similar air parcel sampled by both AQB and the instrumentation in TW-EPA."

Moreover, Fig. S1 is revised with the photograph of the setup condition of AQB system co-located with TW-EPA station as follows:



**Figure S1. (a) Location of TW-EPA Nanzi station (AQB calibration campaign) site and Fooyin University (2013 sampling campaign). (from © Google Earth 2024 and © Google Maps 2024). (b) Photograph of the setup in AQB system operational state. AQB was located approximately 5 m horizontally and 2 m vertically from the sampling inlet of TW-EPA Nanzi station).**

2. Additionally, the differences between AQB and EPA are not clear to me. It would be beneficial to at least discuss the differences in resolution between the two measurement systems (e.g., in the supplement). The devices used at the reference station should also be calibrated, but how was this done?

A: Table S1 was revised to include more detailed information, including the detection range and resolution for sensors in AQB, and intruments at TW-EPA as follows:





Table S1 shows that the instruments in TW-EPA have a higher detection resolution, making them suitable reference devices for calibrating low-cost sensors. In this study, we focus on deriving aerosol hygroscopicity using two PM instruments, OPC and BAM. Detailed information was added to Table S4 as follows:

#### **Table S4: Technical specifications for OPC and BAM**





As to the calibration process for the reference station, the TW-EPA station operates as a standard air quality monitoring station following standard operation procedures of the U.S. Environmental Protection Agency (EPA). The air monitoring analyzers are equipped to perform automatic zero and span calibrations periodically, making self-adjustments to predetermined readings every midnight. Annual multi-point verification/calibrations are conducted to confirm the linearity and calibration slope of the selected calibration scale. For the PM monitoring instrument, the detection sensitivity, accurate PM<sub>2.5</sub> cut point, temperature, barometric pressure sensors, and flow rate device undergo annual calibration. More detailed information can be found in the document "Quality Assurance Handbook for Air Pollution Measurement Systems" (https://www.epa.gov/sites/default/files/2020- 10/documents/final\_handbook\_document\_1\_17.pdf)

3. In the description and discussion of the results, I missed more concrete references to the measurement results of other authors. While the processes behind the described results are explained, there is a lack of references to other literature values.

A: Thank you for your kind reminder. The measurement results from other authors in reference were added to the results and discussion context. The content of Sect. 3.2.1 (Lines 225-231) were revised as follows: "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  $\mu$ g m<sup>-3</sup>, 10.3  $\mu$ g m<sup>-3</sup> for PM<sub>2.5</sub> and PM10, 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 after proper calibration, consistent with other real-time outdoor field studies, reporting  $R^2$  ranging from 0.34 to 0.97, RMSE ranging from 0.52 to 12.3 μg m-3 , and MAPE about 22% (Gillooly et al., 2019; Demanega et al., 2021; Sá et al., 2022; Crilley et al., 2018)." The content of Sect. 3.2.2 (Line 250-254) were revised as follows: "The higher MAPE might result from the low particle number concentration in the coarse mode, with only about 0.01 to 0.1 particles per bin  $cm^{-3}$  in the size range of 3.0 to 10.0  $\mu$ m. The results are consistent with the findings of Kaliszewski et al. (2020), which showed that the correlation between OPC-N3 (a newer version of OPC-N2) and AeroTrak 8220 (TSI INC., Shoreview, MN, USA) measurement data decreases with particle size, from  $0.3{\text -}0.5$   $\mu$ m (r=0.99) to 5-10  $\mu$ m (r=0.74).". The additional discussion was added on Sect.  $3.2.2$  (Line 241-244) as follows:" With a similar methodology, Crilley et al. (2018) applied κ-Köhler equation to compare measured data between OPC-N2 and tapered element oscillating microbalance (TEOM) and derived the  $\kappa$  ranging from 0.38 to 0.41 and 0.48 to 0.51 for PM<sub>2.5</sub> and PM<sub>10</sub>, respectively, which is within the range for Europe (i.e.,  $0.36\pm0.16$ ) (Pringle et al.,  $2010$ ."

4. Inexpensive aerosol analyzers were created particularly to enable long-term observations. It seems to me that the time series used is relatively short. I am not saying it is insufficient, but more extensive measurements could improve the statistics of the presented results. For example, why did the authors choose to conduct measurements in February? What impact could this have had on the measurements?

A: In this study, we selected the winter season as our research period because air pollutant concentrations are typically higher in Kaohsiung City during winter than in summer. The summer months often experience significant rainfall, including afternoon thunderstorms and typhoons, which reduce PM concentrations through wet deposition. While we agree that more extensive measurements could improve the results, it is important to note that aerosol composition varies seasonally in Kaohsiung City, influenced by monsoonal climates and local circulation patterns. Long-term monitoring data spanning multiple seasons might reveal different aerosol characteristics.

Our monitoring campaign began in early February, transitioning into the spring rainy season in March. Moreover, the availability of our measurement at the studied TW-EPA station was limited, so only two weeks of observation were performed for this study. However, the data covered a wide range of RH for the hygroscopicity derivation. Consequently, the hygroscopicity derived from the collected data can be applied for comparison with that calculated from ion chromatography (IC) analysis during the winter campaigns.

5. Some sections, such as 3.3 and 3.4, are written in a single stream of thought, without a clear division into logical parts in the form of paragraphs. This makes these essentially crucial parts of the work difficult to read and may cause problems for readers in understanding the authors.

A: Thank you for your suggestion. The results and discussion have been re-arranged and revised to improve the logicality and clarification as follows (the content was revised but not shown here): "

### 3 Results and Discussion



Detailed observations:

6. In the text, the parameter k appears earlier (line 52) than its name is introduced (line 104).

A: The content has been revised to ensure that all abbreviations are introduced after their full names when it appears first time in the main content. The content of Lines 87-89 was revised as "Notably, Crilley et al. (2018) improved OPC mass concentration correction by applying the derived hygroscopicity  $(\kappa)$  values of 0.38-0.41 and 0.48-0.51 for PM2.5 and PM10, respectively, achieving a 33% improvement." Additionally, a list of principle symbols and abbreviations is provided in Lines 24-60 for clarification.

7. Please check if all elements of the equations are described. For example, what is Sk in equation (3)?

A: Thank you for your kind reminder. The product of two parameters is revised with adding space. The description of elements is also checked and revised. Eq. (2) is revised as follows:"

$$
S = \frac{D_{amb}^3 - D_d^3}{D_{amb}^3 - D_d^3 (1 - \kappa)} \exp(\frac{4\sigma_{s/a} M_w}{R T \rho_w D_{amb}})
$$
(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 (J m<sup>-2</sup>),  $M_w$  is the molecular weight of water (g mole<sup>-1</sup>), *R* is the gas constant (J mole-1 K<sup>-1</sup>), T is the temperature, and  $\rho w$  is the density of liquid water (1.0 g cm<sup>-3</sup>)."

For Eq. (3), S<sub>K</sub> is revised as "S  $\kappa$ " represents the product of water saturation ratio (S) and hygroscopicity (κ) in the numerator. To avoid the misleading, Eq. (3) is revised as follows:

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

8. In section 2.4, the authors describe the various instruments used in the alternative method. Is this method more important? The devices used in this analysis are listed by name in the main text, while the names of the devices used in AQB and EPA are presented in the supplement. Does this mean they are less important? I sense a lack of consistency from the authors in the description of the equipment used.

A: The methodology described in section 2.4 (IC analysis), which requires aerosol sampling, composition analysis, and the calculation of hygroscopic characteristics, represents a typical approach to assessing the hygroscopicity of aerosols through composition analysis. As low-cost sensors are applied to increase the spatial and temporal measurements, a scientific calibration method might be helpful to provide comparison results to the standard instruments. The hygroscopicity parameter is an important factor in the conversion of ambient particles to dry particles. Even though it is less labor-intensive, the data acquisition requires a broader RH coverage. The report hygroscopicity is a mean value over the studied period. The resolution using composition analysis is higher depending on the sampling period, usually half a day. We agree that referring to the devices as AQB and EPA could diminish their representativeness. Therefore, we have focused our analysis on the comparison between the Optical Particle Counter (OPC) and the Beta Attenuation Mass Monitor (BAM). We reviewed all content for PM-related analysis and discussion and replaced AQB with OPC and TW-EPA with BAM. The content of Section 2.2 (Lines 130- 134) was revised as follows:

"For PM, the reported values by Beta Attenuation Mass Monitor (BAM) in the TW-EPA station reflect the dry-state PM concentration by controlling the measurement at RH less than 50% (i.e., a heating device applied to reduce the sampling flow to 35% water saturation when the ambient RH is  $>$  50%). On the contrary, the optical particle counter (OPC) in AQB directly monitors ambient PM concentration. The difference between **BAM and OPC** data reflects the amount of liquid water content in ambient conditions."

9. I believe the manuscript is worth publishing; however, the authors need to work on the readability and logical flow of their work.

A: We appreciate the constructive comments, which have helped us improve the clarity and logical flow of our manuscript.