Responses to anonymous referee #3

General comments:

This paper proposes a method for determining the supersaturation in a fog or cloud. The topic is highly important due to the crucial role played by supersaturation in determining the concentration and shape of the cloud droplet size distribution. The measurement concept seems novel and robust, and is reasonably well explained in the paper. There are a few places that are not clear, which I note below. I'm not sure why this was submitted to ACP instead of AMT, but that's for the editors to decide regarding appropriateness. I consider the manuscript suitable for publication after the following comments are addressed.

Response: Thanks for your comments, the key reason that we decided to submit this manuscript to ACP not AMT is because that this paper is not about instrument development, but about the concept and theoretical framework behind the effective supersaturation measurements using advanced optical systems, and the system could be achieved based on different ways, for example, totally different optical sensors. Therefore, this manuscript is more about physics not techniques.

Major Comments:

Comment: One key question that arises after reading the paper is how the proposed technique can provide information about supersaturation fluctuations versus mean

supersaturation. The authors nicely describe the importance of quantifying fluctuations, but then it is not clearly explained later how this can be accessed. I believe it is related to the sigma value in Equation 2 (see my comment below) but I did not find a discussion of this topic in the paper, except a brief mention in the Discussion (limitation 1). Please provide more discussion of this topic.

Response: Thanks for your comment, this comment really helped us a lot to add further clarification and refinement.

We added the following paragraph in Sect.2.1 to make it clear what types of supersaturation fluctuations could be measured using the proposed framework:

"Two types of supersaturation fluctuations have been previously identified. The first type involves fluctuations in supersaturation directly governed by water vapor pressure and temperature, as described by Siebert and Shaw (2017). These fluctuations are linked to turbulence and water phase changes that influence water vapor pressure and temperature. The second type concerns fluctuations in effective supersaturation, which are associated with the activation and deactivation processes of aerosols, as noted by Ditas et al. (2012). The first type of fluctuations dictates the instantaneous growth and evaporation of droplets, thereby controlling the activation and deactivation of cloud droplets. As such, the second type of fluctuation is inherently driven by the first type. The theoretical framework proposed in this study enables the measurement of fluctuations in effective supersaturation."

We added the following paragraph in the discussion part to demonstrate the potential applications of the measured effective supersaturation fluctuations:

"As mentioned in Sect. 2.1, the theoretical framework proposed in this study is designed to observe effective supersaturation fluctuations, rather than supersaturation fluctuations themselves. While there are non-negligible uncertainties associated with observing effective supersaturation using the proposed theory, the size and hygroscopicity distributions of total interstitial and activated aerosol

populations remain nearly constant when measured with second-scale or shorter time resolution. The parameter that changes over time is the dynamic exchange between interstitial and activated aerosols. Consequently, fluctuations in the scattering signals of interstitial and activated aerosols can reflect this exchange at high temporal resolution. Since effective supersaturation fluctuations result from underlying supersaturation variations, they could, in principle, provide insights into the causes of these fluctuations, such as turbulence, though this would require further investigation and endeavor. In addition, for size-resolved AR, both σ and MAF are crucial parameters. However, using scattering coefficients at just three wavelengths of Aurora 3000 nephelometer is insufficient for accurately retrieving σ and MAF. If σ and MAF could be measured more precisely through the extended optical framework, it would provide deeper insights into supersaturation fluctuations."

Specific comments:

Comment: Lines 84-85: the statement "estimated from vertical velocity measurements" would be better supported by citing a paper that uses that approach, such as the paper by Cooper 1989 (J. Atmos. Sci.). Also, it would be more correct to state "estimated from vertical velocity and droplet size distribution measurements.

Response: The reference is added, and the sentence is revised accordingly.

Comment: Lines 117-119: "The κ-Kohler theory tells that if the critical diameter of aerosol activation (D_a) and corresponding aerosol hygroscopicity parameter κ are known, the surrounding supersaturation can be retrieved based on air temperature measurements and by assuming σ_s\a the surface tension of water." This statement is correct, but it assumes knowledge that is not stated, such as how kappa and the critical

diameter are related to each other. Please explain more thoroughly.

Response: Thanks for your comment, the following discussions are added after this sentence to explain more:

"Note that D_a and κ are not independent with each other, average κ of aerosols with diameter D_a is needed. Previous studies have shown that the reduction in surface tension (Nozière et al., 2010;Gérard et al., 2016;Ovadnevaite et al., 2017) associated with surfactants in atmospheric aerosols can affect aerosol activation and, consequently, the derivation of effective supersaturation. However, if the derivation of κ (as done in this study) assumes a constant water surface tension, the impact of surface tension changes is minimized, as these effects are already incorporated in the κ calculation. Nonetheless, differences in surface tension between supersaturated and subsaturated conditions (Davies et al., 2019;Petters and Kreidenweis, 2013), and their impact on effective supersaturation, still exist. Additionally, prior research has suggested that slightly soluble components in aerosols can influence κ values under both supersaturated and subsaturated conditions (Ho et al., 2010;Petters and Kreidenweis, 2008;Lee et al., 2022;Han et al., 2022;Riipinen et al., 2015;Wang et al., 2019). Therefore, κ observed under subsaturated conditions would affect the derivation of effective supersaturation."

Comment: Line 126: In describing Equation 2 it is stated that "σ is associated with the slope of the curve near D_a". Please provide a physical interpretation of what factors contribute sigma. Would it be true that for a monodisperse aerosol, and uniform supersaturation, sigma would be zero?

Response: Thanks for your comment, this sentence is revised as:

"σ is associated with the slope of the size-resolved AR curve near D_a and mostly influenced by the heterogeneous distribution of aerosols near D_a as well as supersaturation fluctuations (note that not effective supersaturation fluctuations)"

Comment: Lines 134-136: This part of the sentence is not clear and should be revised: "which brings uncertainty in Da derivations due to that the maximum activation fraction of aerosols larger than Da does not equal to unit although usually very close to (Tao et al., 2018b).

Response: Thanks for your comment, this sentence was revised as:

"which brings uncertainty in D_a derivations due to that not all aerosols larger than D_a are activated, because the MAF in Eq.2 does not equal to unit although usually very close to (Tao et al., 2018)"

Comment: Figure 3: Include brief discussion in the figure caption to explain the underlying concept of the instrument, at least for panel a. For example, explain the purpose of the drier versus the cooler (for controlling humidity).

Response: Thanks for your comment, following sentences are added to explain:

"The heater upstream of the sample is used to reduce the relative humidity (RH) to below 60%, ensuring the evaporation of most of the water content, to make sure the consistency of needed PM_1 cut. The cooler upstream of the 'wet' nephelometer increases the sample RH to approximately 90%, allowing hygroscopicity measurements under conditions close to supersaturation."

Comment: Discussion: some discussion of the expected precision versus accuracy, as well as estimated uncertainties should be included.

Response: Thanks for your comment, the following paragraph was added in Sect.4:

"The uncertainty in effective supersaturation observations using this framework primarily arises from the uncertainties in deriving D_a and κ_{D_a}. The uncertainty in D_a observations using the Aurora 3000 nephelometer as the optical sensor under varying conditions is detailed in Fig. 1c. Factors

affecting the accuracy of κ_{D_a} include: (1) the size dependence of κ of activated aerosols; (2) uncertainties related to surface tension, slightly soluble components, and other factors that lead to differences in κ differences under subsaturated and supersaturated conditions. Based on previous studies on the size dependence of κ (Peng et al., 2020) and the differences between subsaturated and supersaturated conditions (Whitehead et al., 2014;Liu et al., 2018;Tao et al., 2023), a 50% uncertainty (three times the standard deviation) was assumed in the derivation of κ_{D_a} for the uncertainty analysis. Using this approach, the uncertainty in effective supersaturation measurements, estimated through the Monte Carlo method, is shown in Fig. 4. The analysis indicates that applying this framework with the Aurora 3000 nephelometer as the optical sensor results in an uncertainty of approximately 5%. The precision of effective supersaturation measurements is directly linked to the accuracy of the optical sensor's scattering signal. For example, the Aurora 3000 has an accuracy of 1 Mm^{-1}, which leads to different levels of precision in D_a and hygroscopicity measurements depending on the scattering signal strength. If the scattering signal from the total aerosol population is 100 Mm^{-1}, the precision of the observed interstitial aerosol scattering fraction f_{sp} is about 1%. Based on the relationship between f_{sp} and D_a shown in Fig. 1b, this leads to a precision of approximately 3 nm for D_a, which results in an effective supersaturation precision of ~0.01% when supersaturation is near 0.2%, or ~0.0002% when supersaturation is near 0.02%. However, if the scattering signal is lower (e.g., 10 Mm^{-1}), a bias of 1 Mm^{-1} could result in effective supersaturation bias to as much as ~0.07% when supersaturation is near 0.2%, making the measurements unreliable. In summary, while the proposed framework demonstrates the feasibility of observing effective supersaturation with an advanced optical system, the accuracy and precision depend on the resolution of the optical sensors, the scattering parameters being measured, and the scattering signal levels of aerosols in clouds. Enhancing the sensor precision to 0.1 Mm^{-1} or even 0.01 Mm^{-1}, and incorporating ultraviolet wavelengths and multiple scattering angles, might enable high-accuracy supersaturation measurements across a broad range of supersaturation conditions, especially in cleaner environments."

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