

We thank the reviewers for their comments and suggestions and have provided point by point responses.

### Referee 1:

General comments:

Authors proposed a new single parameter hygroscopicity representation for insoluble aerosol surfaces, and have done comparisons with traditional TK or FHK models. The proposed model might be extended to atmospherically relevant insoluble particles and findings of this search reveal that water-insoluble aerosol can adsorb water if their surfaces have been oxidized or functionalized with polar groups, thus of importance to atmospheric aerosol research. I only have some minor and specific comments.

1. The logic of the introduction is not clear, and hard to follow. For example, a lot of discussions about the FHK model in the results part, but very few descriptions in the introduction. In my opinion, both the FHK and TK should be introduced before the discussions of FHH-AT.

We have revised the introduction to improve flow and clarity. Specifically, our primary intent is to discuss the importance of the adsorption model (FHH). TK is the simplest and most widely used model and is discussed for comparison. We thought twice about including FHK. FHK is less known but should be applied to water-soluble polymers. PSL is a polymer but not water-soluble and as we show later on, should not be used on PSL particles. Hence, a too lengthy discussion of FHK or TK will distract from the primary FHH and single hygroscopicity parameter message. We use FHK and TK, two water-soluble models, compared with the FHH-AT to show the importance of applying the right theory to specific compounds. In the new revision of the article, we have added sentences as requested.

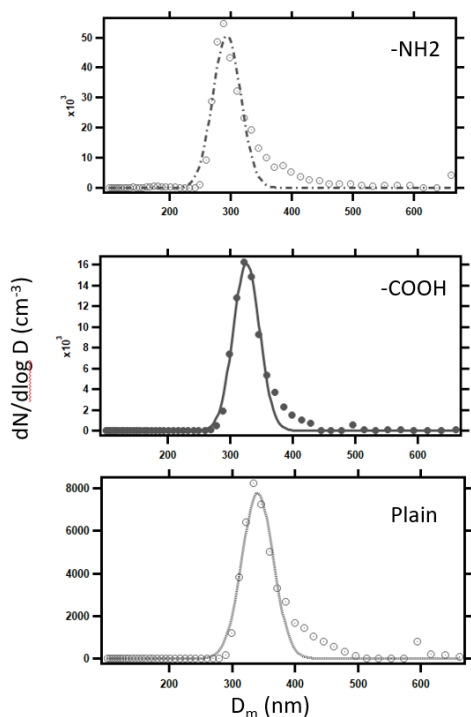
The text has revised sentences and an additional paragraph has been added, and is as follows:

“Flory Huggins Köhler (FHK)(Petters et al., 2009) is one example of a droplet growth model specifically applied to the water soluble polymers. FHK has been shown to work well for long-chained polymers such as gelatin, polyethylene glycol and polylactic acid (Mao et al., 2021; Petters et al., 2006, 2009). It uses a one fitting parameter that describes solvation and most recently was incorporated into a single-parameter hygroscopicity term that describes the water-uptake of water-soluble aerosol (Mao et al., 2021).”

2. The TK model directly gives the relationship between aerosol growth factor and relative humidity (saturation ratio), suggest authors also present a direct formula that links RH,  $D_d$ (dry diameter) ,  $D_w$  (wet diameter) and the single hygroscopicity parameter.

It should be noted that we do not show results from the sub-saturated regime in this paper. PSL does not grow in the subsaturated regime. Thus, equations related to growth factor and RH are not used or presented in this paper. However, FHH does extend into the subsaturated regimes, and thus we refer the author to relevant papers that show sub-saturated FHH work. Furthermore, we encourage the reviewers to explore our companion paper Gohil et al, 2022 (ACPD) that is a hybrid activation adsorption and solubility model that is more robust, can be applied to subsaturated measurements, and has direct equations for the subsaturated environment.

We provide the HTDMA result (RH=91%) of the PSL around 300 nm and show that PSL particles are actually hydrophobic. The growth factor for all three particles are all around 1, showing that the particles do not grow under sub-saturation regime.



HTDMA-91%-300nm

	-NH2	-COOH	-plain
<u>Dd</u>	278	331	310
HTDMA	294.1±22.8612 8	340.37±20.971 37	326.39±24.781 97
HGF	1.057914	1.028308	1.052871

**Specific Comments:**

L37-38, “for water-soluble particles....., TK can accurately predict their water uptake behavior”, I am not sure whether use “accurately predict” is correct. Even the aerosol particle is water soluble, the performance of TK still depends highly on the solubility 1.

The word “accurately” has been deleted. The sentence now reads:

“For water-soluble particles like inorganic ammonium sulfate (Rose et al., 2008) and sucrose (Dawson et al., 2020; Gohil and Asa-Awuku, 2022), TK can predict their water uptake behavior”

L40 “partially water soluble corresponding to very small solubility” or has other physical understanding?

The value of solubility was added to the text. The sentence now reads:

“However, TK does not work so well for atmospherically relevant and abundant particles that are partially water soluble or water insoluble, less than a concentration of  $5 \times 10^{-4}$  (Kumar et al., 2009; Petters and Kreidenweis, 2008; Tang et al., 2016). Thus, alternative droplet growth models for the partial and insoluble particles are needed. “

L44, BET does not appear again in the following, is the abbreviation necessary?

Abbreviation deleted. The sentence now reads:

“Brunauer, Emmett, and Teller adsorption isotherm models are typically applied for multilayer adsorption analysis of water uptake on clays (Hatch et al., 2012) and fly ash (Navea et al., 2017).”

L95-96, should use TK and FHK?

Changed. The sentence now reads:

“Two single parameter hygroscopicity representations have been previously derived using TK (Petters and Kreidenweis, 2007) and FHK (Mao et al., 2021) assumptions.”

L119 flowrate of L/min is better

Corrected. The sentence now reads:

“The PSL particles are then passed through a Condensation Particle Counter (CPC, TSI 3776) with a flow rate of  $0.3 \text{ L min}^{-1}$  and a CCNC. “

L159 the van't Hoff factor is missing

Corrected.

The sentence now reads:

“ $\kappa_{int} = v (M_w \rho_s) / (M_s \rho_w)$  (Sullivan et al., 2009). Where  $M_w$  is the molecular weight of water;  $M_s$  is the molecular weight of the dry particle;  $\rho_w$  is the density of water;  $\rho_s$  is the density of the dry particle;  $v$  is the van't Hoff coefficient that is one.”

L227 change “and” to “;” before AFHH?

Corrected. The sentence now reads:

“The fit results for plain type PSL estimate  $A_{FHH} = 0.17$  and  $B_{FHH} = 0.99$ ;  $A_{FHH}$  is 0.3 and  $B_{FHH}$  is 1.08 for carboxyl functional group modified PSL;  $A_{FHH}$  is 0.11 and  $B_{FHH}$  is 0.83 for amine functional group modified PSL.”

L258 “derived is”, delete “derived”

Corrected. The sentence now reads:

“the derived hygroscopicity is problematic and nonsensical (therefore not shown).”