Response to reviewers

Thanks for reviewers' careful reading and constructive comments and suggestions. We made every effort to respond to reviewers' questions point to point, and revised our manuscript and appendix according to their comments. For clarity, reviewers' comments are shown in *black italic font*. The response is shown in blue normal font. The modified content in the manuscript and/or the appendix is shown in **green bold font**.

Anonymous Referee #2

Referee comment on "Impact of water uptake and mixing state on submicron particles deposition in the human respiratory tract (HRT): Based on explicit hygroscopicity measurements at HRT-like conditions" by Ruiqi Man et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-256-RC2, 2022

Summary:

In this work the authors describe measurement of the hygroscopic growth of externally mixed particles from the North China Plain. They use these data in conjunction with a lung deposition model to predict the effect of hygroscopic growth on deposition in the respiratory tract. The results show that in total, dose was reduced when hygroscopic growth effects were considered as the more numerous smaller particles, that deposit via diffusion mechanisms, deposited less effectively. Variations were seen across the size range, with smaller particles showing a reduced likelihood to deposit, while larger particles were more likely to deposit.

Overall, this paper goes some way towards showing the importance of considering hygroscopic growth, but the extent of new insights is limited. The effects are reported to be rather small so an improved sensitivity analysis and consideration of uncertainties is needed to validate and support the conclusions. Some specific points towards this are detailed below:

1. Deposition fraction is on a particle number basis, and the conclusions connect the dose with the number of particles. The authors should consider reporting dose on a mass deposition basis, which will significantly increase the contributions of the larger particles on deposited dose.

[Response]: Thanks for the reviewer's suggestion. The particle deposition can be weighed by particle number concentrations, mass concentrations, and/or surface area concentrations. Evaluating the deposition dose on a mass basis is definitely an important task in exploring the health risk of particles (such as in toxicological studies). However, due to the lack of the measurement of the PNSD and hygroscopicity of coarse particles, the deposition mass dose of larger particles cannot be calculated in this study. We used the particle number dose as a weight for the reason that the measurement object was submicron particles rather than coarse particles. As the predominant particle type by number in ambient submicron particles, ultrafine particles contribute insignificantly to mass (Xia et al., 2009), but they do great harm to human health (Elsaesser and Howard, 2012; Englert, 2004; Oberdorster, 2001; Sioutas et al., 2005). Therefore, the deposition number dose can highlight the health risk of ultrafine particles. As mentioned above, we used the deposition number dose of particulate matters as a measure to study the particle deposition. The relevant statement was added in the manuscript.

[Revise]: The statement was added in Line 454-459 in the manuscript:

"Besides, the deposition pattern of particles with diameters larger than 1 µm was not discussed here due to the lack of the measurement of the PNSD and hygroscopicity of coarse particles. While, as an important part of the ambient particle mass, coarse particles may also make a significant contribution to the particle deposition in the HRT (Figure S9). The related research to find out the impact of the particle hygroscopicity on the deposition mass dose of coarse particles ought to be carried out in the future."

Figure S9 and related content were added in Line 90-95 in the appendix:

"As shown in Figure S9, a peak appeared at D_p = 2 - 3 µm in the DF curves of the head and P regions, which resulted in a peak in the total DF curve. It implied that particles with larger diameters may also have a significant contribution to the particle deposition in the human respiratory tract.

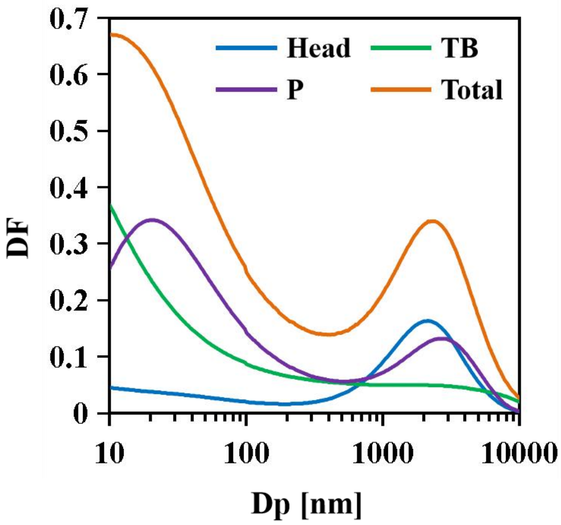

Figure S 9. The size-resolved regional and total DFs for the adults group. The particle density was set as 1.0 g/cm^3."

2. Does the lung deposition model change the density of the particles as they grow due to water uptake? A density of 1.5 g/cm3 is high for hygroscopic particles at >90% RH. I suggest a sensitivity analysis be performed to compare the difference in deposition for 1.0 and 1.5 g/cm3 particle distributions.

[Response]: (1) Thanks for the reviewer's comment. The change of the particle density during the water uptake was not included in the MPPD model.

(2) Due to the lack of the particle density measurement during the sampling period, we compared the differences between the size-resolved DFs of particles with ρ_p = 1.0 g/cm^3 vs. ρ_p = 1.5 g/cm^3 for adults. The results and discussions of the sensitivity analysis of the particle density were added as follows.

[Revise]: The statement was added in Line 291-293 in the manuscript:

"It should be noted that the particle density would change during hygroscopic growth, which was not considered in the calculation due to the lack of the measurement of the particle density. The sensitivity analysis of the particle density on the regional DFs was shown in Figure S4."

Figure S4 and related statement were added in Line 51-68 in the appendix:

"The particle density mainly affects the probability of inertial impaction during the particle deposition process, which can be evaluated by using the dimensionless Stokes number (Stk), defined as Eq (S1) (Pramod et al., 2011):

$$\mathbf{Stk = \frac{\rho_p {d_p}^2 C_c U}{18 \eta d_f}} \quad \text{(S1)}$$

where ρ_p is the density of the particle. The Stokes number is the basic parameter describing the inertial impaction mechanism. A larger Stokes number implies a higher probability of deposition by impaction (Pramod et al., 2011).

Due to the lack of the density measurement of particles during the sampling period, the differences between the size-resolved DFs of particles with ρ_p = 1.0 g/cm^3 vs. ρ_p = 1.5 g/cm^3 for adults were compared. As displayed in Figure S4, the particle density has great influence on the particle deposition in the head and P regions for larger submicron particles. The average DF differences in the head, TB, P, and the whole HRT were (11.1 ± 13.9)%, (0.5 ± 0.8)%, (3.8 ± 6.4)%, and (4.2 ± 6.5)%, respectively. Therefore, the measurement or estimation of the particle density during the particle hygroscopic growth is of great importance in calculating the particle deposition in human bodies.

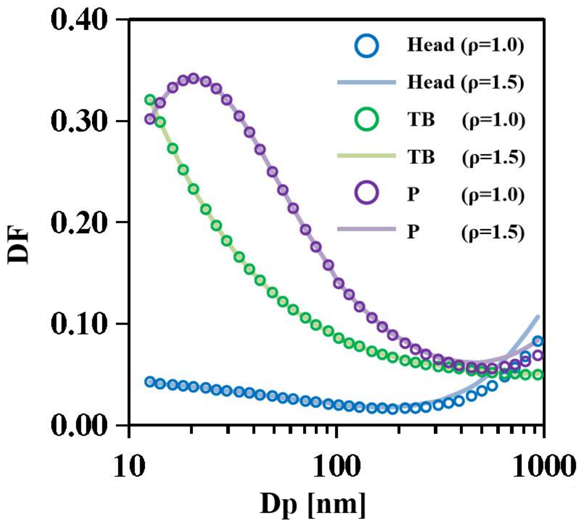

Figure S 4. Size-resolved regional deposition fractions (DFs) of particles with density (ρ) = 1.0 vs. 1.5 g/cm^3 for the adults group. The blue, green, and purple markers represent the DFs of particles with ρ = 1.0 g/cm^3 in the head, TB, and P, respectively. The blue, green, and purple lines represent the DFs of particles with ρ = 1.5 g/cm^3 in the head, TB, and P, respectively."

The statement was added in Line 452-454 in the manuscript:

"Due to the limited measurements and physiological parameters, some vital factors which may have effect on the particle deposition in the HRT were not considered in this study, such as gender, the exercise level, and the particle density."

3. How was the dry size of the particles determined in the hygroscopic growth measurements? Were any shape correction factors considered?

[Response]: (1) As seen in Figure R1, the H-TDMA consists of two differential mobility analyzers (DMAs) and two condensation particle counters (CPCs). The monodisperse aerosol sizes cut with mobility diameters (30, 50, 100, 150, 200, and 250 nm in this study) were selected in turn by the first DMA under dry conditions (RH < 10%). Then, the aerosols passed through a humidifier with a controlled higher RH, and the size distributions over wet mobility diameters were measured with the second DMA (Duplissy et al., 2011).

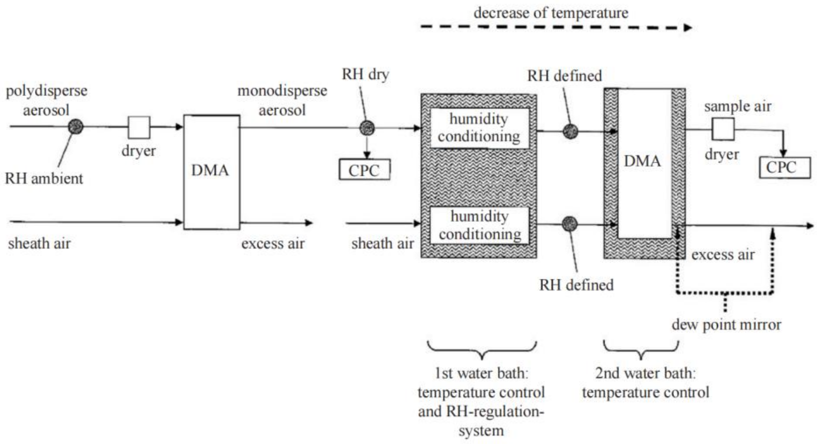

Figure R 1. Set-up of the H-TDMA (Hennig et al., 2005)

(2) No shape correction factor was considered in this study. The ambient aerosols at this sampling site mainly consisted of secondary aerosols (Wu et al., 2017), which are mostly spherical. Irregular particles, such as sea salt and dust, contributed little to aerosols collected at the sampling site. In addition, irregular particles generally exist in coarse mode particles rather than submicron particles. Therefore, The shape factor $\mathcal{X}$ was set as 1.

[Revise]: The content was added in Line 148-155 in the manuscript:

"To match the particle size range in the MPPD model, the electrical mobility diameter was converted to aerodynamic diameter by Eq (4) (Khlystov et al., 2004):

$$\mathrm{d_a} = \mathrm{d_m}\sqrt{\mathcal{X} \times \frac{\rho \times C_{c(dm)}}{C_{c(da)}}} \quad (4)$$

where $\mathrm{d_a}$ and $\mathrm{d_m}$ is the particle aerodynamic diameter (nm) and electrical mobility diameter (nm), respectively. ρ is the particle density (1.5 g cm^{-3} in this study (Hu et al., 2012)). $\mathcal{X}$ is the shape factor. C_c is the Cunningham slip correction factor for a certain diameter. Similar to other studies, the shape factor $\mathcal{X}$ is assumed as 1 and C_c is neglected in the calculation (Khlystov et al., 2004; Hu et al., 2012). Therefore, the electrical mobility diameter (in the range of 10.3 - 756.6 nm) was converted to the aerodynamic diameter (in the range of 12.6 - 926.6 nm)."

4. How accurate is the RH measured in the HTDMA? How stable is the RH? At the high RH of these measurements, even fractions of a % of RH can lead to significant changes in the size of the particles and will introduce uncertainty in the results.

[Response]: The accuracy and stability of the HH-TDMA were studied by Hennig et al (2005). The RH in the second DMA reached an absolute accuracy of ±1.2% for 98% and a long-term stability of ± 0.1-0.4% of set point values (Hennig et al., 2005).

[Revise]: The content was added in Line 114-116 in the manuscript:

“The RH in the second DMA reached an absolute accuracy of ±1.2% for 98% and a long-term stability of ± 0.1-0.4% of set point values (Hennig et al., 2005).”

5. *On line 103, HH-TDMA is referred to – what does the second “H” stand for?*

[Response]: The HH-TDMA is the abbreviation of the high humidity tandem differential mobility analyzer (please refer to Line 91-92 in the manuscript). Therefore, the second ‘H’ stands for ‘humidity’.

6. *Line 84 – a constant value of kappa with RH does not indicate an ideal solution. It indicates that the effective molar volume of the solute does not vary with RH.*

[Response]: Thanks for the reviewer’s correction. The related expression was modified according to the reviewer’s advice.

[Revise]: Line 83-84 in the manuscript:

“It was further assumed that κ was independent of RH on the premise that the effective molar volume of the solute does not vary with RH.”

Reference

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