Supplemental Material

A set-up for respiratory tract deposition efficiency measurements (15-5000 nm) and first results for a group of children and adults

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S1. The RESPI$_2$ set-up

The system used for respiratory tract deposition measurements in this study is flow-through type. The system is based on that described previously in Löndahl et al. (2006) with improvements. All improvements made were aimed at expanding the upper size range of the instrument, from 500 nm up to 5 µm. In order to do so, we introduced an APS (Aerodynamic Particle Sizer, model 3321, TSI®) in the set-up for characterizing the particle number size distribution in the size range of 800 nm up to 5 µm in equivalent aerodynamic diameters (d_{ae}). The APS itself can, according to the manufacturer, classify particles from 0.5 to 20 µm (d_{ae}). However, the data in the range 0.5 to 0.8 are uncertain, and even after redesigning the system to minimize particle losses, 5 µm was considered the upper limit for reliable measurements. For larger sizes, the particle losses in the system are considered too high to perform accurate measurements.

Using two analyzing instruments (SMPS and APS) running in parallel instead of one, as in the previous version of the instrument (incorporating only an SMPS), required some changes to the flow lines that lead the samples of the aerosol from the two tanks to the analyzing equipment. The valve system was redesigned using two 3-way valves as shown Fig. S1. Aerosol samples from the two different tanks were alternatingly led to the SMPS and the APS. The switching of valves was controlled electronically by the RESPI$_2$ software, as was the SMPS system.

The respiratory flow pattern was measured and logged using two pneumotachographs – one placed after the inhalation tank and one at the outlet of in the exhalation tank (Fig. 1). The logged flows were adjusted to take into account the instrumental flows (1 L/min) and calibrated regularly throughout the measurements that were performed over approximately two years. The tidal volumes (converted to BTPS) and average breathing frequencies were calculated from the respiratory flow pattern.

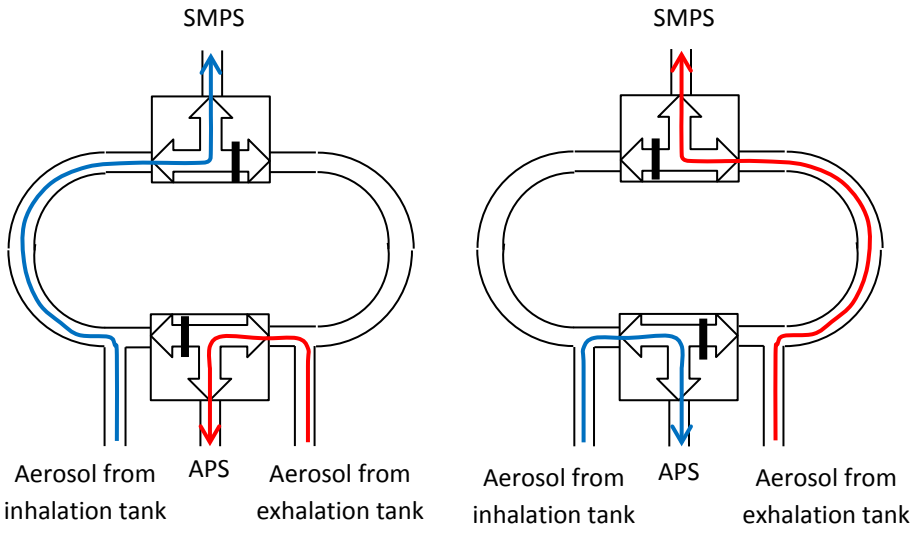

Figure S1. Schematic picture of the air flows through the 3-way valves in the RESPI$_2$.

The SMPS used for the set-up was built in-house and consisted of a bipolar charger, a differential mobility analyzer of Vienna type (length 28 cm, inner/outer radius 25/32.4 mm, aerosol flow rate 0.95 l/min and sheath flow rate 9 l/min) and a condensation particles counter (CPC, model 3010, TSI, Inc.). The SMPS software was integrated into the RESPI$_2$-software. Scanning time was 90 s and alternated

between scanning up and down in voltage. The aerosol flow to the APS was also 0.95 L/min. The APS was regularly calibrated using a standard of monodisperse polystyrene spheres of 5 μm and the SMPS was calibrated using polystyrene latex particles of 100 nm.

S2. Instrumental particle losses (supplemental figure)

The particle losses were measured using carnauba wax and glass spheres. Example of typical size dependent losses in the system is shown in Fig. S2.

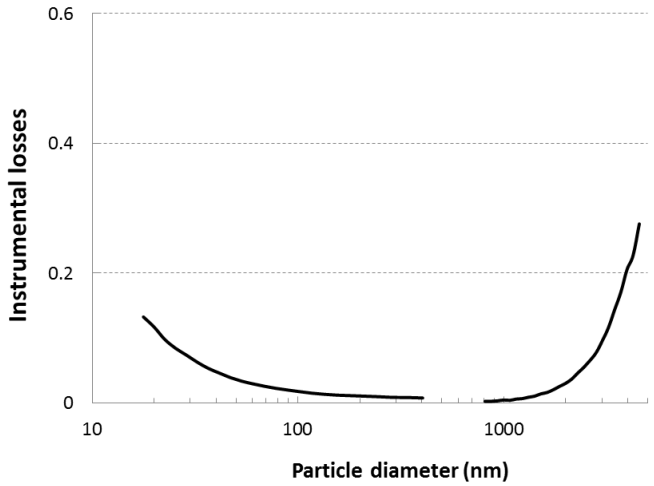

Figure S2. Instrumental particle losses at 6 L/min.

S3. Particle deposition fraction (supplemental figure)

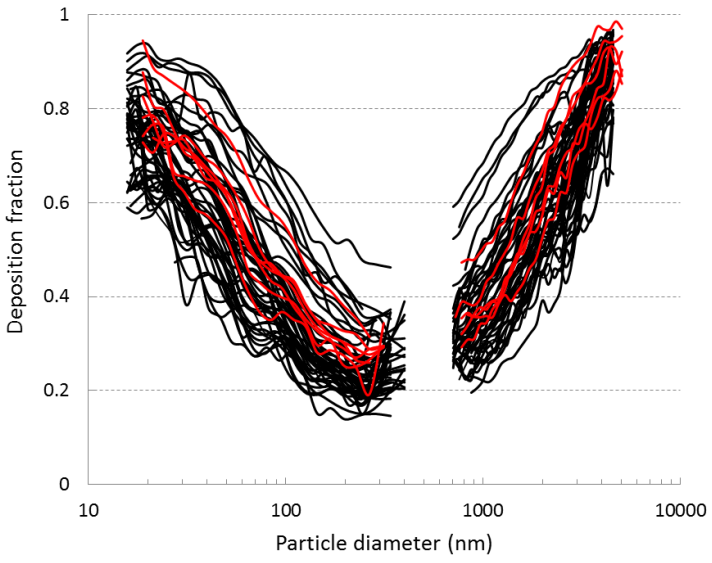

Figure S3. All individual DF(d_p) curves. Black lines correspond to adult subjects and red lines to children.

S4. Test aerosol

The aerosol generated for the lung deposition measurements was composed by carnauba wax in the 10-500 nm diameter range, while in the range 500-5000 nm by glass spheres. A schematic figure of the aerosol generation is shown in Fig. S4. In order to get a broad size distribution of the carnauba wax particles, covering 15-500 nm, three flow lines with different residence times were introduced at the outlet of the furnace, illustrated in Fig. S4. Typically, the carnauba wax particles were tri-modal, while the glass spheres were composed by one broad mode (Fig. S5).

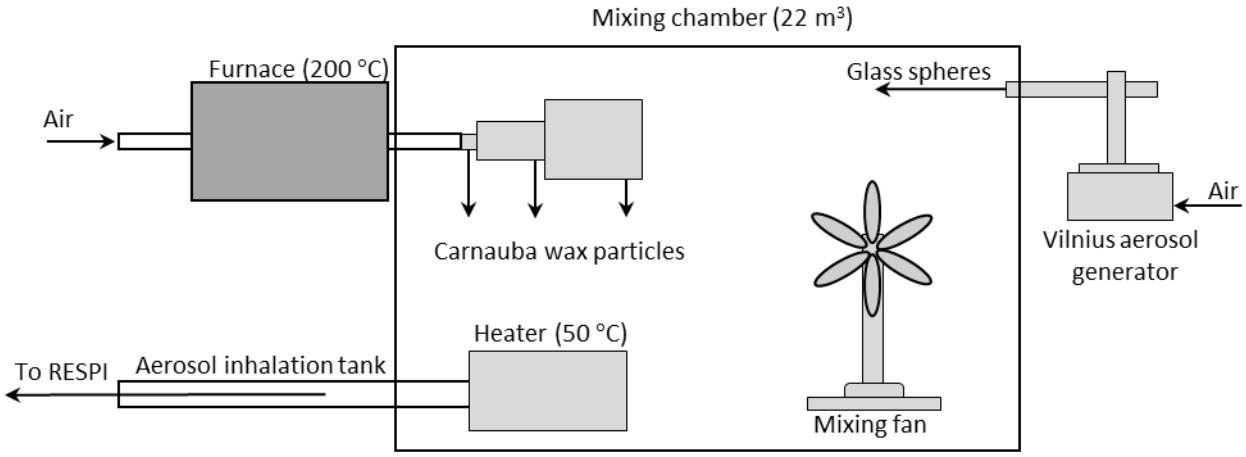

Figure S4. Schematic picture of the aerosol generation.

As described in the review by Löndahl et al. (2014) for lung deposition measurements using polydisperse aerosol particle distributions, any small size shift of the particles before and after inhalation will result in an error in the derived lung deposition fraction. This is illustrated in Figure S6. Due to the characteristic aerosol size distribution used for this study, with the multiple peaks of the carnauba wax particles, any shift in the distribution became visible in the derived deposition pattern, as demonstrated in Fig. S6. The errors introduced for a more unimodal and smooth distribution may be just as large; however, it is not as obvious from the deposition pattern. This was regarded as an advantage in this study since it allowed us to control if size shifts were an issue or not. In the preparation and test carried out prior to the study, there were obvious problems with size shifts. To avoid this, a heater was introduced before the particles entered the inhalation tank of the RESPI (Fig. S4).

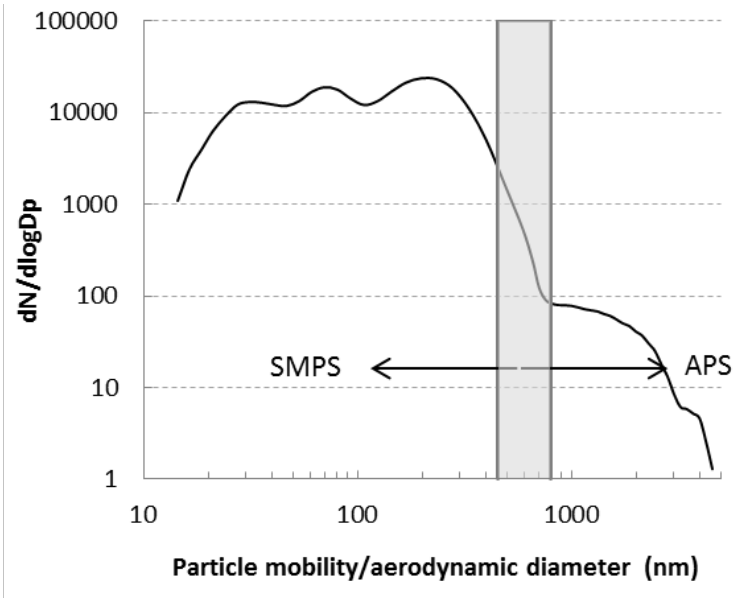

Figure S5. The graph shows a typical size distribution of the aerosol used in the study. The grey area shows the cut-point between the size region of the SMPS and APS.

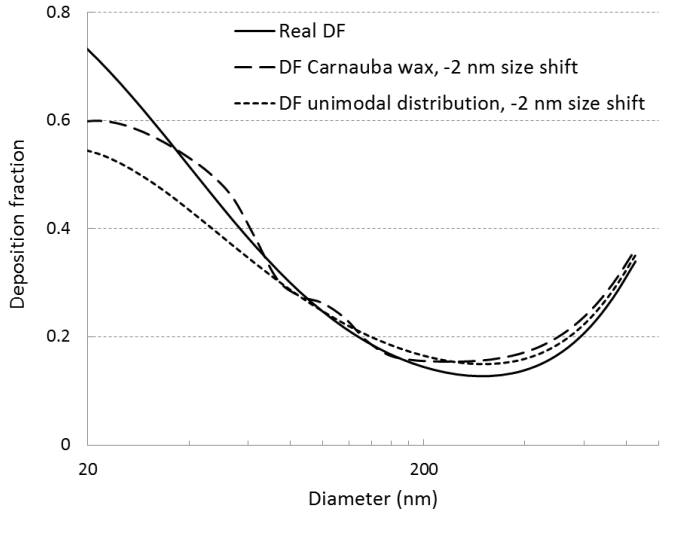

Figure S6. Illustrates the error caused by a negative diameter size shift of 2 nm of the exhaled particles. The solid line corresponds to the true DF, dashed line to the resulting measured DF in the case of a size shift in the carnauba wax particle size distribution, and the dotted line corresponds to the measured DF in the case of a size shift of a fictive distribution with a geometrical mean diameter of 100 nm (geometrical standard deviation 1.7).

References

Löndahl, J., Möller, W., Pagels, J.H., Kreyling, W.G., Swietlicki, E. and Schmid, O. (2014). Measurement Techniques for Respiratory Tract Deposition of Airborne Nanoparticles: A Critical Review. *Journal of aerosol medicine and pulmonary drug delivery* 27: 229-254.

Löndahl, J., Pagels, J., Swietlicki, E., Zhou, J.C., Ketzel, M., Massling, A. and Bohgard, M. (2006). A Set-up for Field Studies of Respiratory Tract Deposition of Fine and Ultrafine Particles in Humans. *Journal of Aerosol Science* 37: 1152-1163.