We thank anonymous referee 2 for these excellent remarks

I found the discussion on why mainstream aerosol studies would benefit from higher size-resolution a bit short or maybe not highlighted well enough. I would appreciate it if at some point in the manuscript (the earlier the better) there was a list of bullet points illustrating in what sense mainstream aerosol studies would benefit from HRDMAs.

The opinion's abstract has been extended to indicates that *"... many studies central to aerosol research could be carried out with HRDMAs with considerable advantage in size range, resolution, sensitivity and measurement speed.*" These are the four main bullet points, right at the outset. The reason why they are not developed immediately is that there is a deeply rooted impression in the field that these advantages are either not needed or counterproductive. It seems to me it would be unwise not to give this psychological reality full recognition from the beginning. This point is now stressed with the following two published quotations and a brief remark:

For instance, Kangasluoma et al. 2017 note that "However, these supercritical DMAs were mainly used in laboratory calibrations in previous studies because of their high flow rates of several hundred or thousand liters per minute and the corresponding high maintaining expenses". Similarly, we read in Cai et al. 2017 (2010) that "However, these DMAs are designed to operate at high sheath flowrate (typically several hundreds to thousands lpm) rather than matching the flowrate required by the DEG-UCPC. In addition, large sheath flow recirculation system (including high volume filters and air mover) are required to operate those DMAs, making it troublesome for field measurement". These remarks must be taken most seriously, as they originate from groups working on new particle formation, which are well acquainted with the characteristics of HRDMAs.

Related to the above, I find, that the current highlighting of new particle formation (NPF) and growth as the example for mainstream aerosol studies is weak. It is exactly the NPF community, which typically focuses on the sub-10 nm range, which is the size range where HRDMAs are already in use. In addition, the reference to Kong et al. (2021), which - while important - might not be the only one showing the occurrence of narrow size-distributions in NPF studies (especially as it shows a highly specific case from a laboratory experiment only). The short sentences about studies on viruses and the signal-to-noise remain without reference or illustration (a Figure could nicely show this).

Excellent points. The abstract now incorporates the following remark and references:

"*However, they have not been widely adopted, except in the size range below 10 nm, often in new particle formation studies (Kangasluoma et al. 2020 and Ozon et al. 2021).*"

The revised manuscript now refers to published virus spectra illustrating the relation between signal/noise and resolution.

If signal processing tools are used to obtain size-distribution related parameters such as the new particle formation and growth rate, the paper by Ozon et al. (2021) investigated that a full coverage of the size-distribution with lower resolution can be better than an insufficient coverage with high resolution (e.g. usage of a HRDMA in a DMPS with just a few mobility steps). On the other hand, Kangasluoma et al. (2020) has already highlighted in what sense sub-10 nm studies would benefit from higher resolution in the sense of uncertainty reduction. The ideal instrument would provide all together: high resolution and good coverage of all sizes at high signal (i.e. high detector flow rates). This could be clarified.

Thank you. These two most relevant articles are now cited in the revised *Opinion*.

The *Opinion* (and the already posted comments) already include a very valuable discussion on higher detector flow rates, the usage of better aerosol electrometers and CPCs providing such high detection flow rates. First attempts to increase CPC detection flow rates have been made (but are still far away from "high" flow rates). It was recently shown that such an increased sensitivity through higher detector flow rates indeed provides a significantly improved performance in e.g., NPF studies (Stolzenburg et al. 2023). In addition, I just wanted to point out that for the usage of CPCs together with a HRDMA high CPC detector flow rates are no necessity for extending the Q range. q/Q can also be held constant when a CPC is used by just adding a variable make-up flow downstream of the HRDMA.

Thank you. Stolzenburg at al. is now included in the detector discussion.

I think it is important to add some thoughts on how a HRDMA system which is scanning Q would be calibrated (i.e., the set flow rates to be measured/inferred) and how fast such a system could "scan" or "step" through different flow rates. From my own experience, it seems that the blowers we currently use in our high resolution DMAs have quite a significant lag in their PID control of a new set blower flow rate and the flow to be stable and reliable. In that sense, I am skeptical if scan rates faster than typical voltage scans can be achieved potentially contradicting the response time argument? I would appreciate it if this experimental obstacle would also be shortly discussed in the *Opinion*.

Thank you. The following extended discussion of these important practical considerations is now included:

3.9 Sheath flow rate control

Anonymous referee 2 brings up several important issues relating to the control of the sheath gas. The proposed sweeping over Q rather than V requires a continuous way to measure Q, which cannot be achieved with the same precision and ease as a voltage measurement. There is also the issue of the slower response time of mechanical pumps versus high voltage sources. Nevertheless, an accurate mobility scale requires a precise determination of Q, irrespective of whether one sweeps over Q or over V. Whatever flowmeter is used, the ambiguity in the mobility scale will typically be determined by the flowmeter error, which will not be necessarily greater in a Q-sweep than in a V-sweep. The main difference is that in a V-sweep the Z scale will

be off by a fixed unknown factor, versus a variable equally unknown factor in a Q-sweep. Assuming that the relative error in the flow rate is not greater at high Q than at low Q, it would be of little consolation to know that this error is constant. And this slight advantage will be lost in a conventional sweep if one decides not to be absolutely limited by operating always at fixed Q, irrespective of the size range of interest. These considerations naturally do not remove the need to measure Q as precisely as possible over the whole range of the Q-sweep, which will evidently require a certain development. We are familiar with commercial flowmeters claiming 2% error in Q, and reproducibility within 1%, though only covering the limited range 0-300 L/min. Venturi flowmeters, for instance, can go substantially higher, with comparable precision. These various flowmeters have response times much faster than the pump (limited by the inertia of the rotor), being capable of determining the actual flow rate at any instant during a sweep. The instantaneous pump frequency is also readily measured, and is an alternative marker of the flow rate. There should accordingly be no difficulty in carrying out minute long full Q and V scans. The possibility to achieve wide V-scans in a few seconds has been previously demonstrated (Fernandez de la Mora et al. 2017b) with electrometers, and would be far more problematic on a Q-sweep.

There are, however, other potential difficulties associated to a Q-scan that may not be so simple to handle, or that could limit the scan speed. One of them is that the working pressure in a typical closed sheath flow circuit may depend on the pump speed, while the response time for this pressure depends on how the system pressure is set relative to the external world. No experience is available at present to assess this issue.

One major experimental challenge of HRDMAs is the control of the sheath gas chemical composition, which is currently not mentioned. The *Opinion* only discusses temperature, but relative humidity and purity of the sheath gas are typically also controlled in ambient SMPS/DMPS measurements. To meet the standards which are achieved with *plain* DMAs in most ambient studies, HRDMAs would need to dry and clean the sheath flow at high flow rates. I would appreciate it if this was additionally mentioned besides the temperature issue.

Thank you. The following extended discussion of this important practical consideration is now included.

The ability to control the chemical composition of the sheath gas is relatively limited, as it is unpractical to dry a large flow rate of sheath gas. The most common way of operating a HRDMA is with the sheath gas in closed circuit, such that, after an initial transient, its composition matches that of the entering aerosol. Zinola et al. (2019) have operated a high temperature Half-Mini DMA that sampled hot automobile exhaust gases from the ambient. To avoid vapor condensation from the exhaust gases, they did run the sheath gas at 200º C. This was achieved by taking filtered ambient gas into the pump, blowing it into a heater, then into the DMA sheath gas circuit inlet, and finally exhausting the hot excess gas back to the atmosphere. The temperature was thus controlled, but not the humidity. Note that the pressure at the

polydisperse aerosol inlet slit in the Half-Mini DMA is close to the pressure in the separation region, which is below the pressure at the entry of the sheath gas because the flow moves at relatively high speed (Venturi effect). This pressure is also below the sheath outlet pressure because the DMA is provided with a diffuser. Accordingly, given a suitable diffuser, it is possible to suck ambient aerosol when the sheath gas is exhausting to the atmosphere. On the other hand, none of the HRDMAs built to date to classify particles as large as 60 nm or more has incorporated a diffuser.

In the two previously tested modes, with open and closed sheath gas circuits, the only available steady compositions were either that of the ambient air or that of the aerosol. If one needs a sheath gas drier than the aerosol, one option is to dry the aerosol and operate in close circuit. Another option is to recirculate the sheath flow through a drying medium. It would be far simpler to remove the little humidity brough by the aerosol into the closed circuit than to thoroughly dry a large flow of ambient air. Other humidity controls are feasible. For instance, if the polydisperse aerosol flow is 3 L/min, and one introduces 27 L/min of dry gas into the closed circuit and (after thorough mixing of these two inputs) draws another 27 L/min out of the circuit, the sheath gas will reach an equilibrium humidity $1/10^{th}$ that of the entering aerosol.