Supplementary Information: Diffusional Transfer Function for the Scanning Electrical Mobility Spectrometer (SEMS)

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1 Streamline Approach in Evaluating Static DMA Transfer Function

In this section, we review the method that Knutson and Whitby (1975) (KW) and Stolzenburg (1988) applied to derive the non-diffusional and diffusional static DMA transfer functions, respectively.

The conceptual scheme of the DMA is shown in Figure 1 in the main manuscript. We present a similar to that of Stolzenburg (1988) to account for particle diffusion in the scanning DMA in this manuscript.

Knutson and Whitby (1975) developed an elegant model for the transmission of non-diffusive particles in terms of the fluid flow streamfunction, ψ, which is defined such that

$$ru_r = \frac{\partial \psi}{\partial x}, \qquad ru_x = -\frac{\partial \psi}{\partial r} \tag{S1}$$

where r and x are the radial and axial coordinates, and u is the fluid velocity, and the electric flux function, ϕ, which, for the steady-state DMA, can be related to the electric field through

$$rE_r = \frac{\partial \phi}{\partial x}, \qquad rE_x = -\frac{\partial \phi}{\partial r} \tag{S2}$$

where E is the electric field. Non-diffusive, charged particles follow trajectories of constant particle stream function, Γ, i.e.,

$$\Gamma = \psi + Z\phi$$

which is related to the particle velocity, v, via

$$rv_r = \frac{\partial \Gamma}{\partial r}, \qquad rv_z = -\frac{\partial \Gamma}{\partial z} \tag{S3}$$

Particles that do not diffuse are advected by the gas and migrate under the action of the electric field, maintaining a constant value of Γ along their trajectories. Thus,

$$\Gamma_e - \Gamma_i = \Delta\Gamma = 0 = \Delta\psi + Z\Delta\phi$$

along the particle trajectory. Thus, the mobility that corresponds to particles crossing a range of fluid streamlines, $\Delta\psi$, and of electric flux function, $\Delta\phi$, is

$$Z = -\frac{\Delta\psi}{\Delta\phi} \tag{S4}$$

Applying the KW definition of the characteristic trajectory of the classified particles within the static DMA as that which enters the classification region at the center of mass of the incoming aerosol sample, and exits at the center of mass of the outgoing classified aerosol flow, we define

$$Z^* = -\frac{\Delta\psi^*}{\Delta\phi}$$

This result applies generally for a static DMA, and is not dependent upon the specific geometry.

For a high aspect ratio (large length relative to distance between electrodes) cylindrical DMA (CDMA), the flow may be approximated as flowing parallel to the coaxial, cylindrical electrodes, in which case, $u_r = 0$, and $\psi(r, x) = \psi(r)$ (Figure 1). Multiplying Eq. (S1) by 2π and integrating from radial position r to the outer radius, R_2, where the aerosol enters the DMA, we find

$$2\pi\psi(r) = \int_r^{R_2} 2\pi r u_z(r) dr = Q(r) \tag{S5}$$

which is the volumetric flow rate between the radial position and the outer electrode. The aerosol flow rate is Q_a, so the value of the streamfunction at the center of mass of that flow, i.e., at the radial position r^* where $Q(r_i^*) = \frac{Q_\mathrm{a}}{2}$, is $\psi_i^* = \frac{Q_\mathrm{a}}{4\pi}$; that at the center of mass of the classified aerosol outlet is $\psi_e^* = \frac{2Q_\mathrm{ex}+Q_\mathrm{c}}{4\pi} = \frac{Q_\mathrm{sh}+Q_\mathrm{ex}+Q_\mathrm{a}}{4\pi}$, where Q_sh, Q_ex, and Q_c are the sheath, exhaust, and classified sample flow rates, respectively. Thus,

$$\Delta\psi^* = \frac{Q_\mathrm{sh} + Q_\mathrm{ex}}{4\pi}$$

The electric field in an ideal, constant voltage, cylindrical DMA is

$$E(r) = -\frac{V_e}{r \ln \frac{R_2}{R_1}}$$

assuming the central rod voltage is negative, so

$$\Delta\phi^* = \int_0^L -\frac{V_e dx}{\ln \frac{R_2}{R_1}} = -\frac{V_e L}{\ln \frac{R_2}{R_1}}$$

and the mobility of the particle following the characteristic trajectory becomes

$$Z^* = \frac{Q_{\text{sh}} + Q_{\text{ex}}}{4\pi L V_e} \ln \frac{R_2}{R_1} \tag{S6}$$

where L is the effective coaxial length of the DMA. While the voltage is kept constant during the classification, we have explicitly noted that the voltage is that at the instant when the particle leaves the classification region, to be consistent with the definition here in the case of scanning DMA.

Stolzenburg (1988) estimated the form of the transfer function for diffusive particles by the stream function method. Define s as the coordinate along the length of the trajectory, and y as the local coordinate that is orthogonal to the trajectory. Brownian diffusion leads to a mean-square increment in displacement from this kinematic (non-diffusive) trajectory in a time increment, dt, as

$$d\sigma_y^2 = 2\mathcal{D}dt \tag{S7}$$

where $\mathcal{D}$ is the particle diffusivity. The local velocity of the particle is the time derivative of the vector from the origin of a suitable fixed coordinate system, i.e., $\vec{\rho}$, to the particle position, which can be decomposed to the local components in the (s, y) coordinate system, such that

$$\vec{v} = \frac{d\vec{\rho}}{dt} = v_s \frac{\partial \vec{\rho}}{\partial s} + v_y \frac{\partial \vec{\rho}}{\partial y}$$

For small displacements, we may approximate v_s and v_y by Taylor series about the non-diffusive trajectory,

$$\begin{aligned} v_s &\approx v_s|_{y=0} + \left.\frac{\partial v_s}{\partial y}\right|_{y=0} y \\ v_y &\approx v_y|_{y=0} + \left.\frac{\partial v_y}{\partial y}\right|_{y=0} y \end{aligned}$$

Neglecting cross-stream shear, $\left.\frac{\partial v_s}{\partial y}\right|_{y=0} = 0$, and $v_s \approx v_s|_{y=0}$. By definition of our local coordinate system, $v_y|_{y=0}$.

Further assuming that $\left.\frac{\partial v_y}{\partial y}\right|_{y=0} = 0$, we have

$$\begin{aligned} v_s &\approx \left. v_s \right|_{y=0} \approx |v| \\ v_y &\approx \left. v_y \right|_{y=0} \approx 0 \end{aligned}$$

which is strictly valid only for high aspect ratio DMAs, i.e., those for which $L/(R_2 - R_1) \gg 1$. This model further neglects distortions to particle concentration profiles due to the presence of walls, such that particles are allowed to deviate from the classified aerosol outlet flow by diffusing through the walls.

We are not concerned with the spatial deviations of particles, but rather with the extent to which they deviate across the flow from the inlet particle stream function, Γ_i. Error propagation shows the incremental change in the stream function variance to be

$$d\sigma_\Gamma^2 = \left(\frac{\partial \Gamma}{\partial y}\right)^2 d\sigma_y^2$$

Following Stolzenburg (1988), it can be shown that, at $y = 0$, and with the assumptions described above,

$$\frac{\partial \Gamma}{\partial y} = rv$$

so, with Eq. (S7),

$$d\sigma_\Gamma^2 = 2v^2 r^2 \mathcal{D} dt$$

For a high aspect ratio CDMA, such as that originally described by Knutson and Whitby (1975) and commercialized as the TSI Model 3081 long column DMA, and relatively large particles that are classified at voltages in excess of a few tens of volts, neglecting the losses to the classification region walls, as is implicit in this model, is reasonable because particles are sufficiently far from the walls through most of their transit through the DMA. For highly diffusive particles, which are classified at low voltage, diffusional losses may become important. Scaling σ_Γ^2 with respect to $(\Delta\psi^*)^2$ yields a dimensionless transfer function broadening parameter that can be related to an integral along the characteristic trajectory as

$$\tilde{\sigma}^2 \equiv \frac{\sigma_\Gamma^2}{(\Delta\psi^*)^2} = \frac{2\mathcal{D}}{(\Delta\psi^*)^2} \int_{\Gamma^*} v^2 r^2 dt \tag{S8}$$

Defining a new radial variable $\omega = \left(\frac{r}{R_2}\right)^2$ and a new radial parameter $\gamma = \left(\frac{R_1}{R_2}\right)^2$, and invoking the flow rate ratios of Knutson and Whitby (1975) provides new parameters

$$\beta = \frac{Q_\mathrm{a} + Q_\mathrm{c}}{Q_\mathrm{sh} + Q_\mathrm{ex}} \tag{S9}$$

$$\delta = \frac{Q_\mathrm{c} - Q_\mathrm{a}}{Q_\mathrm{c} + Q_\mathrm{a}} \tag{S10}$$

Casting the integral in EQ. (S8) in nondimensional form, the variance becomes

$$\tilde{\sigma}^2 = \left(\frac{4\pi}{Q_\mathrm{sh} + Q_\mathrm{ex}}\right)^2 2\mathcal{D}\bar{U}^2 R_2^2 t_m \int_{\Gamma^*} \tilde{v}^2 \omega d\tau,$$

where t_m is the mean gas residence time

$$t_m = \frac{2\pi R_2^2 L(1-\gamma)}{(Q_\mathrm{sh} + Q_\mathrm{ex} + Q_\mathrm{a} + Q_\mathrm{c})} \tag{S11}$$

$\tau = \frac{t}{t_m}$, and $\bar{U} = \frac{L}{t_m}$. $\tilde{\sigma}^2$ can be expressed in terms of dimensionless variables as

$$\tilde{\sigma}^2 = 4\left(\frac{1+\beta}{1-\gamma}\right)\frac{1-\sqrt{\gamma}}{\mathrm{Pe}^*_\mathrm{mig}}\zeta \int_{\Gamma^*} \tilde{v}^2 \omega d\tau \tag{S12}$$

Then the migration Péclet number of a particle of diffusivity $\mathcal{D}^*$ in a CDMA is defined as

$$\mathrm{Pe}^*_\mathrm{mig} = \frac{v^*_{E_2}(R_2 - R_1)}{\mathcal{D}^*}$$

$\zeta = \frac{\mathcal{D}}{\mathcal{D}^*}$ accounts for the difference of the diffusivity of the particle in question from that of a particle of mobility Z^*, and $v^*_{E_2} = Z^* E(R_2, V_e) = \frac{Q_\mathrm{sh}+Q_\mathrm{ex}}{4\pi L R_2}$ is the migration velocity of a non-diffusive particle traveling along the characteristic trajectory that exits the classification region at time t_e (or dimensionless time τ_e). The mobility is related to the diffusivity through

$$Z = \frac{ne}{kT}\mathcal{D},$$

where k is the Boltzmann constant, and T is the temperature, so that $\zeta = \frac{\mathcal{D}}{\mathcal{D}^*} = \frac{Z}{Z^*}$.

The integral of Eq. (S12) is evaluated by noting that

$$v^2(r,t) = u(r,t)^2 + v^*_{E_2}(t)^2 \frac{R_2^2}{r^2},$$

so

$$\int_{\Gamma^*} (\tilde{v}(\omega,\tau))^2 \omega d\tau = \int_{\Gamma^*} (\tilde{v}_{E_2}(\tau))^2 d\tau + \int_{\Gamma^*} (\tilde{u}(\omega(\tau),\tau))^2 \omega d\tau = I_r + I_z \tag{S13}$$

where the two integrals are denoted as I_r and I_z, and $\tilde{v}_{E_2}(\tau)$ is the electrophoretic migration velocity of a particle at $\omega = 1$. The two integrals, which correspond to the components of the particle motion in the coaxial and radial directions, respectively, can be evaluated for either the steady-state DMA, or one in which the voltage changes with time given the characteristic trajectory for a given DMA and voltage profile. These results may be combined to express the dimensionless variance as

$$\tilde{\sigma}^2 = \frac{G_{\text{DMA}}}{\text{Pe}^*_{\text{mig}}}\zeta,$$

where

$$G_{\text{DMA}} = 4\left(\frac{1+\beta}{1-\gamma}\right)(1-\sqrt{\gamma})\,(I_z + I_r)$$

Thus, the Stolzenburg (1988) analysis makes it possible to evaluate $\tilde{\sigma}^2$, provided the center of mass of an ensemble of particles of a given mobility follows a consistent kinematic trajectory through the classification region of the DMA. The original analysis was performed for the static (constant voltage) DMA; for size distribution measurements, this mode of operation corresponds to the so-called differential mobility particle sizer (DMPS). Other modes of operation that yield consistent trajectories are also amenable to this analysis as we shall show below. The key hypothesis underlying this transfer function estimation is that particles deviate from the non-diffusive trajectory that begins at ψ_i and ends at ψ_e as if there were no walls present in the DMA. This leads to a delta function distribution about the non-diffusive trajectory and a Gaussian distribution about the diffusive trajectory. To be transmitted from the incoming aerosol flow to the outgoing aerosol flow, $0 \le \psi_i \le \frac{Q_{\text{a}}}{2\pi}$ and $\frac{Q_{\text{sh}}}{2\pi} \le \psi_e \le \frac{Q_{\text{total}}}{2\pi}$, the transfer function is then

$$\Omega(Z, Z^*) = \int_0^{\frac{Q_{\text{a}}}{2\pi}} \left[\int_{\frac{Q_{\text{sh}}}{2\pi}}^{\frac{Q_{\text{sh}}+Q_{\text{ex}}+Q_{\text{a}}+Q_{\text{c}}}{4\pi}} f_{\text{trans}}(\psi_e, \psi_i)\,d\psi_e\right] f_{\text{inlet}}(\psi_i)\,d\psi_i \tag{S14}$$

where $f_{\text{inlet}}(\psi_i) = \frac{2\pi}{Q_{\text{a}}}$ is the inlet probability function, and $f_{\text{trans}}(\psi_e, \psi_i) = \delta_D(\psi_e - \psi_i + Z\Delta\phi)$ for the non-diffusive trajectory, where $\delta_D(x)$ is the Dirac delta function, and $f_{\text{trans}}(\psi_e, \psi_i) = \frac{1}{\sqrt{2\pi}\sigma_\Gamma}\exp\left[-\frac{(\psi_e-\psi_i+Z\Delta\phi)^2}{2\sigma_\Gamma^2}\right]$ for the diffusive trajectory. For the steady-state DMA, all particles experience the same $\Delta\phi$, so integrating first over ψ_e, and then over ψ_i yields the transfer function in dimensionless form,

$$\Omega_{nd}(\zeta) = \frac{1}{2\beta(1-\delta)}\Big[|\zeta-(1-\beta)| + |\zeta-(1+\beta)| - |\zeta-(1-\beta\delta)| - |\zeta-(1+\beta\delta)|\Big] \tag{S15}$$

for the non-diffusive case, and

$$\Omega_d(\zeta) = \frac{\tilde{\sigma}}{\sqrt{2}\beta(1-\delta)}\left[\mathcal{E}\left(\frac{\zeta-(1-\beta)}{\sqrt{2}\tilde{\sigma}}\right) + \mathcal{E}\left(\frac{\zeta-(1+\beta)}{\sqrt{2}\tilde{\sigma}}\right) - \mathcal{E}\left(\frac{\zeta-(1-\beta\delta)}{\sqrt{2}\tilde{\sigma}}\right) - \mathcal{E}\left(\frac{\zeta-(1+\beta\delta)}{\sqrt{2}\tilde{\sigma}}\right)\right] \tag{S16}$$

for the diffusive case, where

$$\mathcal{E}(x) = x\mathrm{erf}(x) + \frac{\mathrm{e}^{-x^2}}{\sqrt{\pi}}$$

and $\mathrm{erf}(x)$ is the error function.

For the scanning DMA, one must account for variation of $\Delta\phi$ for particles that enter the DMA at different times. The instantaneous variation of ϕ with x for the SEMS is

$$\Delta\phi = \int_{x_i}^{x_e} r \frac{V_i}{r \ln \frac{R_2}{R_1}} \mathrm{e}^{\pm \frac{t}{t_s}} dx = \int_{t_i}^{t_e} \frac{V_i}{\ln \frac{R_2}{R_1}} \mathrm{e}^{\pm \frac{t}{t_s}} \frac{dx}{dt} dt = \int_{t_i}^{t_e} \frac{V_i}{\ln \frac{R_2}{R_1}} \mathrm{e}^{\pm \frac{t}{t_s}} \bar{U} \tilde{u}(\omega(t)) dt$$

so $\Delta\phi$ cannot be assumed to be constant during scanning. Nonetheless, the values of $\Delta\phi$ for any (ψ_i, ψ_e) pair can be computed numerically, enabling numerical evaluation of the transfer function. While this approach lacks the analytical solution attained for the steady-state DMA, it does afford an efficient approach for determination of the scanning DMA transfer function. Since the integral above involves a variable of time, which cannot be resolved with the streamline method, we show in the main manuscript that starting from dynamic trajectories one can derive the same conclusion as the streamline method and at the same time can be applied in the scanning case.

2 Estimation of $\tilde{\sigma}$

As shown in Section 1, the variance of streamline, σ_Γ^2, is scaled by $\left(\frac{Q_{\mathrm{sh}} + Q_{\mathrm{ex}}}{4\pi}\right)^2$ as $\tilde{\sigma}^2$ in Eq. (S12), which is the same as the dimensionless variance of flow fraction. In this section, we will show the derivation of the integral part in Eq. (S12) for both the static and scanning cases, and show that the integral of the scanning case is identical to that of the static case when the scanning time $\tau_s \to \infty$.

From Eq. (S12), $\tilde{\sigma}^2$ can be written in two forms:

$$\tilde{\sigma}^2 = 4\left(\frac{1+\beta}{1-\gamma}\right) \frac{1-\sqrt{\gamma}}{\mathrm{Pe}^*_{\mathrm{mig}}} \zeta \int_{\omega_i}^{\omega_e} \tilde{v}^2 \omega d\tau = 4\left(\frac{1+\beta}{1-\gamma}\right)^2 \frac{1-\sqrt{\gamma}}{\mathrm{Pe}^*_{\mathrm{mig}}} \lambda \int_0^{\theta_t} \tilde{v}^2 \omega d\theta \tag{S17}$$

where $\lambda = \zeta \frac{1-\gamma}{1+\beta} \tau_s$, $\theta_t = \frac{\tau_e - \tau_i}{\tau_s}$ and $\theta = \frac{\tau_e - \tau}{\tau_s}$. The expression on the right hand side will simplify the calculation for the scanning case.

2.1 Static DMA

To simplify the calculation, we use the $\tilde{\sigma}^*$ at the centroid streamline as the key calculation, the same strategy as that adopted by Stolzenburg (1988), which is within the precision since the diffusion range is small relative to ψ_{in} and ψ_{out}. To derive $\tilde{\sigma}^2$ in the context of this manuscript, first we can transform the integral variable from τ to ω.

From Eq. (11) in the main manuscript, we have $d\tau = -\dfrac{1}{\zeta^*}\dfrac{1+\beta}{1-\gamma}d\omega$, thus the mid-term in Eq. (S17) becomes

$$\tilde{\sigma}^2 = 4\left(\frac{1+\beta}{1-\gamma}\right)^2 \frac{1-\sqrt{\gamma}}{\mathrm{Pe}^*_{\mathrm{mig}}}\zeta \int_{\omega_e^*}^{\omega_i^*} \left(\tilde{v}_r^{*2} + \tilde{u}^2(\omega)\right)\omega d\omega = 4\left(\frac{1+\beta}{1-\gamma}\right)^2 \frac{1-\sqrt{\gamma}}{\mathrm{Pe}^*_{\mathrm{mig}}}\zeta\left(I_r + I_z\right) \tag{S18}$$

where we have used the fact that $\zeta^* = 1$.

Since

$$v_r^* = \left(\frac{dr}{dt}\right)^* = Z^*E = \zeta^*\left(\frac{Q_{\mathrm{sh}}+Q_{\mathrm{ex}}}{4\pi LV}\ln\frac{R_2}{R_1}\right)\left(\frac{-V}{R_2\ln\dfrac{R_2}{R_1}}\right)\frac{R_2}{r} = -\zeta^*\frac{Q_{\mathrm{sh}}+Q_{\mathrm{ex}}}{4\pi LR_2}\frac{R_2}{r} \tag{S19}$$

where we have used the definition of Z^* in Eq. (S6), so $\dfrac{v_r^*}{\bar{U}} = \tilde{v}_r^* = \zeta^*\dfrac{Q_{\mathrm{sh}}+Q_{\mathrm{ex}}}{4\pi R_2 L}\dfrac{R_2}{r}\Big/\dfrac{Q_{\mathrm{sh}}+Q_{\mathrm{ex}}+Q_{\mathrm{a}}+Q_{\mathrm{c}}}{2\pi(R_2^2-R_1^2)} = \dfrac{\zeta^*}{2}\dfrac{1-\gamma}{1+\beta}\dfrac{R_2}{L}\dfrac{R_2}{r}$ and $\dfrac{u_z}{\bar{U}} = \tilde{u}(\omega)$. Then we have

$$I_r = \int_{\omega_e^*}^{\omega_i^*}\frac{\zeta^{*2}}{4}\left(\frac{1-\gamma}{1+\beta}\right)^2\left(\frac{R_2}{L}\right)^2\frac{\omega}{\omega}d\omega = \frac{1}{4}\left(\frac{1-\gamma}{1+\beta}\right)^2\left(\frac{R_2}{L}\right)^2\left(\omega_i^* - \omega_e^*\right) \tag{S20}$$

where we have used the fact that $\zeta^* = 1$, and

$$I_z = \int_{\omega_e^*}^{\omega_i^*}\tilde{u}^2(\omega)\omega d\omega \tag{S21}$$

where $\tilde{u}(\omega) = \frac{\ln\gamma(1-\omega)-(1-\gamma)\ln\omega}{\frac{1+\gamma}{2}\ln\gamma+1-\gamma}$.

Note that Stolzenburg (1988) uses a different integral range of ω, which is from γ to 1, not the same range from ω_e^* to ω_i^* (the centroid point). The results from Eqs. (S20) and (S21) will be the same as Stolzenburg's expression if ω_i^* and ω_e^* are replaced by 1 and γ).

Instead of using the same $\tilde{\sigma}^*$ for all the flow fractions (or streamlines), we can calculate the $\tilde{\sigma}$ for every pair of (ω_i, ω_e). Starting from the right-hand-side of Eq. (S17) and $d\theta = \dfrac{1}{\lambda}d\omega$ in Eq. (11) in the main manuscript, we get

$$\tilde{\sigma}^2 = 4\left(\frac{1+\beta}{1-\gamma}\right)^2\frac{1-\sqrt{\gamma}}{\mathrm{Pe}^*_{\mathrm{mig}}}\int_{\omega_e}^{\omega_i}\tilde{v}^2\omega d\omega \tag{S22}$$

Thus for every pair of (ω_i, ω_e), there will be a dependent $\tilde{\sigma}$, i.e. $I_r = \dfrac{\lambda^2}{4\tau_s^2}\left(\dfrac{R_2}{L}\right)^2(\omega_i - \omega_e)$ and $I_z = \displaystyle\int_{\omega_e}^{\omega_i}\tilde{u}^2(\omega)\omega d\omega$.

2.2 Scanning DMA

Here, we use V_e as the reference static DMA working voltage. First, we still apply the centroid point method to assess if at the limit $\tau_s \to \infty$, the scanning method will approximate the static result. From Eq. (7) in the main manuscript, the scanning $v_r^\dagger$ is

$$v_r^\dagger = \left(\frac{dr}{dt}\right)^\dagger = -\zeta^\dagger \frac{1}{t_m}\frac{1-\gamma}{1+\beta}\mathrm{e}^{-\theta}\frac{R_2^2}{2r} \tag{S23}$$

so $\frac{v_r^\dagger}{\bar{U}} = \tilde{v}_r^\dagger = -\zeta^\dagger \frac{1}{t_m}\frac{1-\gamma}{1+\beta}\mathrm{e}^{-\theta}\frac{R_2^2}{2r}\Big/\frac{L}{t_m} = \frac{\zeta^\dagger}{2}\frac{1-\gamma}{1+\beta}\frac{R_2}{L}\frac{R_2}{r}\mathrm{e}^{-\theta}$ and $\frac{u_z}{\bar{U}} = \tilde{u}(\omega)$.

From Eq. (S17), we have

$$I_r = \int_0^{\theta_t^\dagger} \frac{\zeta^{\dagger 2}}{4}\left(\frac{1-\gamma}{1+\beta}\right)^2\left(\frac{R_2}{L}\right)^2\frac{\omega}{\omega}\mathrm{e}^{-2\theta}d\theta = \frac{1}{4}\left(\frac{1-\gamma}{1+\beta}\right)^2\left(\frac{R_2}{L}\right)^2\left(\frac{1}{2}\right)\left(1-\mathrm{e}^{-2\theta_t^\dagger}\right) \tag{S24}$$

We note that as $\tau_s \to \infty$ and substituting $\theta_t^\dagger = \frac{\omega_i^* - \omega_e^*}{\lambda^*}$ into Eq. (S24) gives the same $\tilde{\sigma}^2$ as Eq. (S20) does, and I_z is the same expression as Eq. (S21). If we want to calculate σ for every pair of (ω_i, ω_e), then $I_r = \frac{\lambda^2}{4\tau_s^2}\left(\frac{R_2}{L}\right)^2\left(\frac{1}{2}\right)\left(\mathrm{e}^{1-2\theta_t}\right)$ and $I_z = \int_0^{\theta_t}\tilde{u}^2(\omega)\omega d\theta$.

3 Arrival-Time of Monodisperse Particles

In this section, we show the results of the time that particles have experienced inside the scanning DMA. Particles of the same mobility are injected continuously and uniformly into the inlet. The arrival time instance τ_a of the particle that enters the scanning DMA at the position $\omega_i = 1$ and ends at $\omega_e = \omega_c$ (lower left corner of Fig. S1-A) is set to its transit time τ_t, thus the entering time instance of that particle is $\tau_i = 0$. The entering times τ_i of all other particles that can penetrate the scanning DMA are referred to that particle, so the contour lines in Fig. S1-A represent the particles that enter the DMA at the same time instant τ_i but at different position and successfully get through the DMA. Fig. S1-B shows the transit time of each particle, which is the same as in Fig. 5B in the main manuscript, indicating that the transit time is an inherent property of the trajectories in the scanning DMA, independent of the reference time. The summation of τ_i and τ_t is the arrival time of the particles, which can be used to calculate the arrival-time transfer function (ATF). The contour line distribution in Fig. S1-C is similar to that in Fig. 4. The calculation of the ATF is a 2D integral along the contour line, yielding a trapezoid-shaped ATF (Fig. S1-D), the same as the derived instantaneous transfer function in the main manuscript but different from those in Collins et al. (2004) and Dubey and Dhaniyala (2008), which could be a result of the choice of particle injection interval at the inlet and counting time interval at the outlet in the numerical simulation.

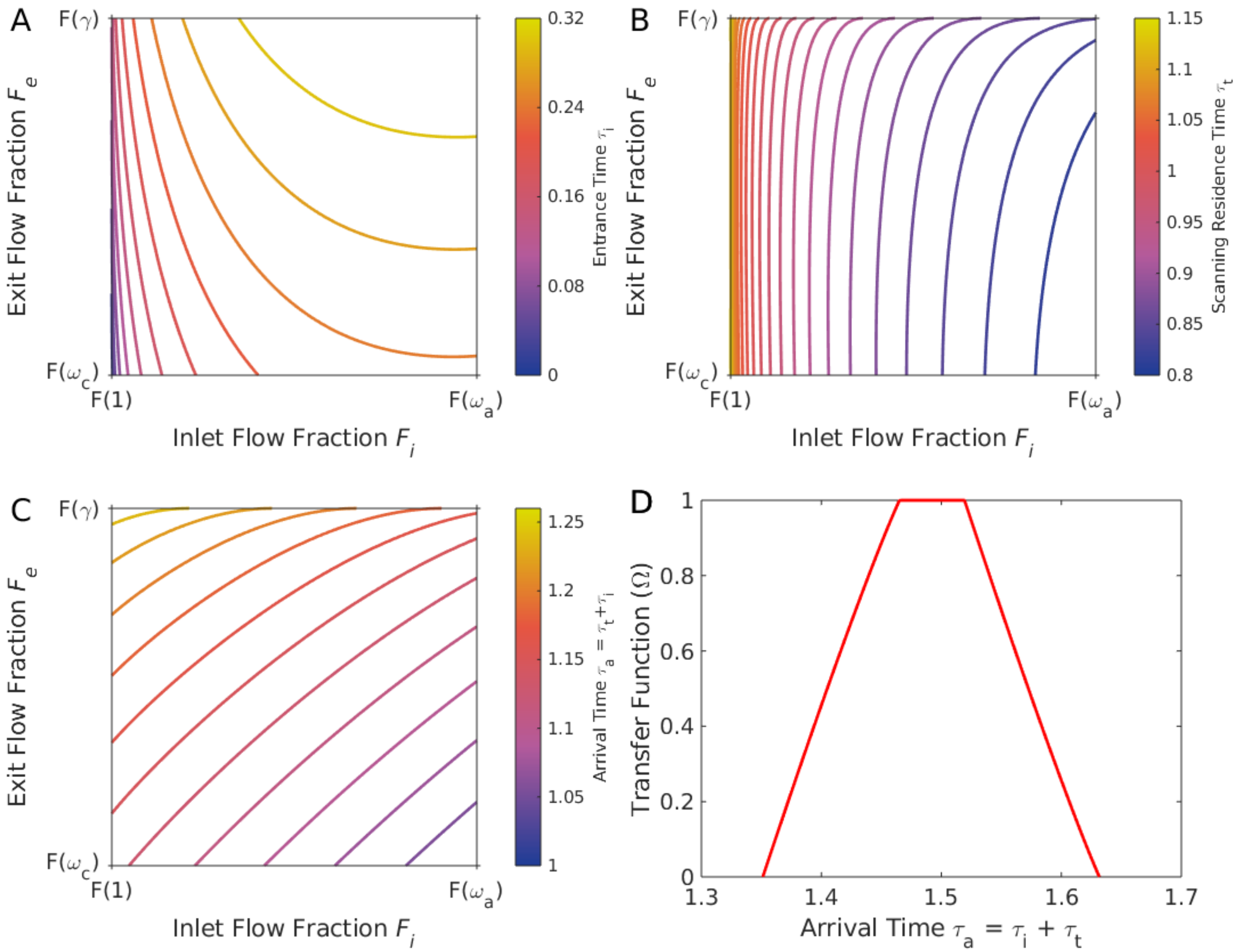

Figure S1: Contour plots of the entering time τ_i (at which time particles of the same mobility entering the DMA can transit the DMA, Panel A), the transit time τ_t (which time the particles have experienced inside the DMA, Panel B), and the arrival-time $\tau_t + \tau_i$ (the overall time that the particles have spent since the continuous injection at time $\tau_i = 0$, Panel C) for the scanning DMA ($\beta = \frac{1}{10}$, $\delta = 0$, and $\tau_s = 1$). Panel D: Arrival-time transfer function. See Section 3 for details.

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