

Open-source modelling of aerosol dynamics and computational fluid dynamics: bipolar and unipolar diffusion charging and photoelectric charging

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Abstract: Electric charging is one of the essential aerosol dynamic mechanisms and is harnessed for detection, capture and control of ultrafine aerosol particles in a range of devices. For simplicity, charging and transport mechanisms are commonly modelled with zero spatial dimensions (0-D) and averaged properties such as mean charge or mean particle diameter. These models often neglect localised effects of the flow distribution, diffusion, discrete charge states, and particle polydispersity, often proving inadequate to explain experimental data. This work aims to provide an open-source three-dimensional (3-D) aerosol charging and transport model including bipolar and unipolar diffusion charging, and photoelectric charging algorithms for use in detailed design and analyses of aerosol systems. The computational model consists of more than 200 particle transport equations for discrete charge states and polydisperse sizes coupled with ion conservation equations in the framework of OpenFOAM, an open-source computational fluid dynamics platform. Three test cases are introduced to verify implementation of three charging models by comparison with published literature: bipolar and unipolar diffusion charging, and photoelectric charging. Tutorial cases, which model three distinct aerosol sensors, are described and demonstrate the capabilities of the 3-D aerosol

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charging and transport models within the predetermined flow field. The openAerosolCharging code is available at <https://openaerosol.sourceforge.io> for widespread use and can be further modified under the GNU general public licence.

Program Summary

Program title: openAerosolCharging

CPC Library link to program files: (to be added by Technical Editor)

Developer's repository link: <https://openaerosol.sourceforge.io/>

Code Ocean capsule: (to be added by Technical Editor)

Licensing provisions: GNU General Public License 3 (GPL)

Programming language: C++

Nature of problem (approx. 50-250 words): openAerosolCharging solves generalised aerosol electrical charging and transport equations coupled with computational fluid dynamics using the open-source computational platform, OpenFOAM [1]. The electric charging algorithm including unipolar diffusion charging, bipolar diffusion charging, and photoelectric charging is verified with zero-dimensional test cases [2-4] and generalized to three-dimensional monodisperse or polydisperse particle distributions for more than 50 particle charge states. openAerosolCharging can be straightforwardly coupled with other existing solvers, which enables computations of complex multi-physics aerosol charging processes in practical conditions.

Solution method (approx. 50-250 words): openAerosolCharging employs an explicit time-stepping for the time-dependent source terms for generalised aerosol charging. The solution methods and schemes provided by OpenFOAM 9 are used for solving the spatial and time derivatives in the transport equations.

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1 Introduction

Aerosols can directly and indirectly affect the climate, environment and human health. The dynamics of aerosols are fundamental to problems such as air pollution formation (e.g. particulate matter from combustion and secondary organic aerosols), cloud formation (e.g. atmospheric aerosols), delivery of medicine/drugs in vaporizers (e.g. e-cigarettes, pharmaceuticals), disease transmission (e.g. via respiratory aerosols and droplets), and gas-phase production of engineered nano-materials (e.g. carbon nanotubes; catalysts and catalyst supports) [1, 2]. Electrical charging of particles plays a significant role in many natural processes and is commonly harnessed in a wide range of engineered systems for measurement, capture, and control of aerosol particles [1, 3].

Aerosols acquire charge through a range of mechanisms including flame charging, static electrification, electrolytic charging, field charging, and photoelectric charging. However, even in ambient air, transfer of charge is continuously occurring between gaseous ions and aerosol particles by diffusional and electrostatic forces, referred to as ‘diffusion charging’ [1, 3]. A range of measurement systems (typically for aerosol particles smaller than $\sim 1\ \mu\text{m}$ in diameter) include a charge conditioning stage which most commonly employs unipolar or bipolar diffusion charging (ions of one or both polarities, respectively) or photoelectric charging for subsequent measurement of the charged particles. For example, ISO standard methods for measuring aerosol size distributions rely on knowledge of the charge distribution which results from bipolar diffusion charging [4-6]. Many instruments employ unipolar diffusion charge conditioners often when high charge levels of one polarity are desired for electrometer measurements [7] including those with fine size resolution [8-10] and those which are lower in cost and portable [11-14]. Furthermore, instruments may use the photoelectric effect to charge aerosols for subsequent measurement, by irradiating them with ultraviolet light, and must

design for discharging by diffusional charge transfer with gaseous ions [15, 16]. Knowledge of the charging processes and behaviour of charged particles is important for the design of instruments for more accurate measurements or for optimization considering size, weight and cost. Therefore, this work focuses on modelling of bipolar and unipolar diffusion and photoelectric charging as well as particle transport in the context of aerosol measurement systems.

Charge conditioners yield charge distributions (i.e. the fraction of particles with different integer numbers of discrete charges, referred to as ‘charge state’) which are predictable by models for correct measurement inversion to yield aerosol properties, e.g. particle mobility diameters or concentrations. In the absence of predictable charge distributions, detailed experimental calibration is necessary. Or, if charge distributions are incorrectly predicted, systematic bias in measurements could occur [17]. Since charge conditioners are subject to localized flow and charging effects [18-20], spatial detail must be accurately modelled for design and optimization, and to accurately predict charge states. Advances in modelling of electrical charging either focus on detailed physics of ion-particle collisions [21] or, if the spatial distributions of ions or particles are considered for applications in charge conditioning [22], significant assumptions are made to simplify the transport equations. In low-cost sensors, the integrated measured signal is a direct function of particle polydispersity, yet models often assume monodisperse particle distributions, mean charge states (rather than distributions of charge states), and assume transport phenomena such as diffusion are negligible to simplify the solution algorithms [13, 23]. While these assumptions may be acceptable in simple systems, they may not be sufficient in other cases [18, 19, 24, 25]. For example, localized transport of ions due to electrostatic dispersion should be considered when high concentrations of charge are present [18, 19, 24, 25] and the expected charge distributions may not be reached in bipolar chargers in part due to localized effects of advection and diffusion of ions or particles [18, 19, 22, 24, 25]. Comprehensive modelling of the temporo-spatial dynamics of particle charging coupled with fluid dynamics represents an opportunity for improved design and optimization of aerosol instruments.

Although many numerical models for specific charging behaviour, e.g. diffusion charging [7, 26-29] or photoelectric charging [23], have been developed, the models for comprehensive aerosol charging in combination with computational fluid dynamics (CFD) simulations [25, 30] are rare. Open-source codes which combine aerosol and computational fluid dynamics

phenomena are limited to other applications in the areas of aerosol dynamics, transport or aerosol growth rather than charging [31, 32]. In this work, we present an open-source toolbox for generalized aerosol electrical charging and transport equations. The toolbox includes 0-D models up to three-dimensional (3-D) CFD models. Equations for steady-state particle and electric field transport including electrostatic effects are solved and modularized to selectively include or neglect charging and transport terms. The charging equations include unipolar diffusion, bipolar diffusion, and/or photoelectric charging. Sample cases are included for simple 0-D models which retain sufficient complexity in charging equations but neglect localized geometrical effects. Source terms for a range of mechanisms may be selected at run-time (immediately prior to solving): for example, particle diffusion loss to walls. The solution algorithm is generalized to monodisperse or polydisperse particle distributions and up to 50+ particle charge states. The codes are written using C++ in OpenFOAM v9, an open-source CFD platform [33]. By incorporating a CFD platform, users may easily adapt to different flow conditions and geometries, and the code may be operated natively in parallel if needed. Importantly, the computational method and its implementation are verified by comparison with existing literature to provide confidence in numerical results of other aerosol systems.

2 Governing equations

Figure 1 shows the driving forces for particle and ion transport under consideration and conceptual figures for three particle charging mechanisms used in this study. As shown in Figure 1a, charged particles are subject to electrostatic forces from an externally applied electric field and the interactions with gaseous molecules through advection and drag forces. The electric charging of particles is represented by a probability of collision and charge transfer between gaseous ions (ionized gaseous molecules) and particles considering diffusive (via Brownian motion) and electrostatic forces [34]. As shown in Figure 1b and c, bipolar and unipolar diffusion charging occurs when aerosol particles are exposed to gaseous ions of either dual or single polarities, respectively. For bipolar diffusion chargers (of Figure 1b), gaseous ions of both polarities are typically generated by exposing gaseous molecules to ionizing radiation from a radioactive source (e.g. Am-241 or Kr-85), though other ionizing sources such as X-ray are also common [35]. Ions are typically generated throughout the internal volume of a bipolar charge conditioner (i.e. the volume which is exposed to ionizing radiation). For unipolar diffusion chargers (of Figure 1c), a corona discharge process is typically used as the

ion source where ions are generated near the corona wire or needle. There are various methods to transport ions through a charging section, where particles are exposed to the ions, such as convection, diffusion, or electric field transport [36]. For either unipolar or bipolar diffusion charging, the gaseous ions subsequently transfer charge to particles resulting in a charge distribution which may be estimated. In a direct ultraviolet (UV) photoelectric charger, particles are exposed directly to UV light, absorb light energy and emit electrons which, in turn, quickly become gaseous ions (as seen in Figure 1d). An externally applied electric field, created by applying a potential between two electrodes, is typically used to capture highly mobile ions generated during particle photoelectric charging since those (negative) ions may neutralize the (positively) charged particles by diffusion charging. However, charged particles are captured less readily in an electric field than gaseous ions due to their significantly lower electrical mobility, leaving them to remain suspended in an advective flow for subsequent measurement [20, 37].

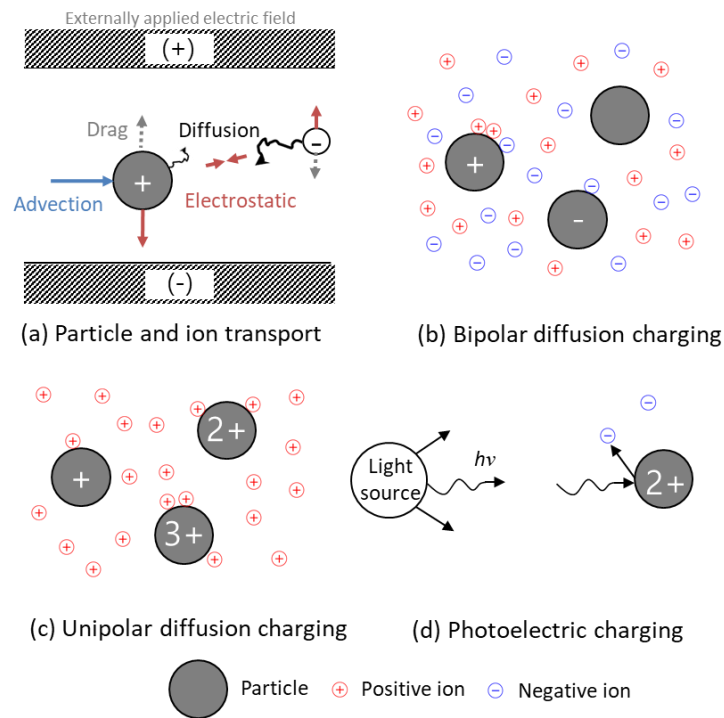


Figure 1: Schematics of (a) particle transport and charging mechanisms, (b) bipolar diffusion charging, (c) unipolar diffusion charging, and (d) photoelectric charging.

The air flow entraining particles is solved by a 3-D incompressible Navier-Stokes equation. Temperature is assumed to be constant, such that the temperature-dependent physical properties (i.e. density and viscosity of fluid) are constant. It is assumed there is negligible momentum transfer from particles or ions to the flow field, which allows decoupling solution procedures of flow from the transported ions or particles. Therefore, the velocity field dictating the particle advection is predetermined in the present model. Unsteady particle-fluid interaction is not considered in the present study. The general aerosol dynamics including nucleation, coagulation, and surface growth are not considered in this study, and interested readers may refer to the complementary open-source models of Woo et al. [31], released on the same *openAerosol* platform.

Considering the assumptions stated above, the general equations of aerosol dynamics and electric charging can be represented as the 3-D steady state conservation equations for the particle number concentration $N_{b,q}$ of a given charge level q and size within a bin b :

$$\nabla \cdot (\mathbf{u}N_{b,q}) = \nabla \cdot (D_{b,q} \nabla N_{b,q}) - \nabla \cdot (Z_{b,q} \mathbf{E} N_{b,q}) + S_{b,q,\alpha} + S_{b,q,\beta} \quad (1)$$

The left-hand side of the equation represents the advection of particles with the gas velocity $\mathbf{u}$ of the independently solved flow field. The first and second terms of right-hand side are particle transport by diffusion and electric field, respectively. D and Z represent the diffusivity and electrical mobility of particles, respectively. Diffusion charging in the free-molecular, transition and continuum regimes are captured by a range of theories which have been validated in many experimental studies [35, 38-42]. Equations based on Fuchs' limiting sphere theory [43] are widely accepted for charging applications in aerosol instrumentation and are therefore used in this work [4, 34, 36]. The fourth term in the RHS of Eq. (1) accounts for the ion particle collisions assuming that the ion transfers its charge to the particle, as follows.

$$S_{b,q,\beta} = \beta_b^{q+1 \rightarrow q} N_{b,q+1} n^- - \beta_b^{q \rightarrow q-1} N_{b,q} n^- + \beta_b^{q-1 \rightarrow q} N_{b,q-1} n^+ - \beta_b^{q \rightarrow q+1} N_{b,q} n^+ \quad (2)$$

where β is the attachment coefficient defined as [44]

$$\beta = \frac{\pi \delta^2 c \alpha_c(q)}{\exp\left(\frac{\varphi(i(q), \delta)}{k_B T}\right) + \frac{\delta c \alpha_c(q)}{4D_i} \int_0^1 \exp\left(\frac{\varphi(i(q), \delta/x)}{k_B T}\right) dx} \quad (3)$$

and where δ is the limiting sphere radius as a function of the mean free path of the ions [45], c is the mean thermal velocity of the ions, and α_c is the ion-particle collision probability

depending on charge and distance, T is the temperature, k_B is Boltzmann's constant, and integration is over the non-dimensional distance x from infinity to the limiting sphere, δ . The potential φ is a function of the particle diameter and its conductivity as follows

$$\varphi(i, r) = V_e \left(\frac{i}{r} - K \frac{(d_p/2)^3}{2r^2(r^2 - (d_p/2)^2)} \right) \quad (4)$$

where r is the distance from the centre of the particle, i is the number and polarity of particle charges such that i is positive if the ion and particle are of the same polarity. The non-dimensional conductivity parameter K is set to 1 for a perfectly conducting particle, and V_e is the elementary electron potential defined as:

$$V_e = \frac{e^2}{4\pi\epsilon_0} \quad (5)$$

where e is electron charge and ϵ_0 is vacuum permittivity.

The third term on the RHS, which may be neglected if photoelectric charging is not present, is a source term for photoelectric charging defined by

$$S_{b,q,\alpha} = \alpha_b^{q-1 \rightarrow q} N_{b,q-1} - \alpha_b^{q \rightarrow q+1} N_{b,q} \quad (6)$$

where α is the combination coefficient for photoelectric charging calculated by the extended form of Fowler-Nordheim equation. A combination coefficient $\alpha^{q \rightarrow q+1}$ [23, 46] accounting for the probability of photoelectric charging spherical particles from charge level q to $q+1$ is

$$\alpha^{q \rightarrow q+1} = K_c (h\nu - \Phi^{q \rightarrow q+1})^m \frac{I\pi d_p^2}{4h\nu} \quad (7)$$

where I is the intensity of UV irradiation (power per unit area), and $h\nu$ is the specific photon energy. K_c is a material dependent and empirically determined proportionality constant. The value of the exponent m is commonly 2 for metals and a few non-metals [47, 48]. Φ is the particle surface work function extracted from Michaelson [49] represented as

$$\Phi^{q \rightarrow q+1} = \Phi_\infty + \frac{2V_e}{d_p} \left(q + 1 - \frac{5}{8} \right) \quad (8)$$

where Φ_∞ is the work function of a flat surface of the same material. The value of $h\nu$ must be greater than Φ to enable a positive emission probability.

The steady transport equations for positive (+) ions, n^+ , and negative (-) ions, n^- , are given by

$$\begin{aligned} \nabla \cdot (\mathbf{u}n^+) &= \nabla \cdot (D^+ \nabla n^+) - \nabla \cdot (Z^+ \mathbf{E} n^+) \\ &+ \gamma^+ - \sum_{b=1}^m \sum_{q=q_{\min}}^{q_{\max}} (\beta_b^{q \rightarrow q+1} N_{b,q} n^+) - a n^+ n^-, \end{aligned} \quad (9)$$

$$\begin{aligned} \nabla \cdot (\mathbf{u}n^-) &= \nabla \cdot (D^- \nabla n^-) - \nabla \cdot (Z^- \mathbf{E} n^-) \\ &+ \gamma^- + \sum_{b=1}^m \sum_{q=q_{\min}}^{q_{\max}} (\alpha_b^{q \rightarrow q+1} N_{b,q} - \beta_b^{q \rightarrow q-1} N_{b,q} n^-) - a n^+ n^- \end{aligned} \quad (10)$$

where γ is the ion generation rate, and a is the ion-ion recombination coefficient. Note that the ion and particle charge conservation equations are coupled by the fact that both n and N appear in the source terms. In this study, the local charge density due to contributions from ions and charged particles is assumed not to affect the local electric field, $\mathbf{E}$, which is independently solved for from an externally applied potential. This assumption is valid for dilute particle and ion concentrations, e.g. less than 5×10^{13} ions m^{-3} [18, 50].

To comprehensively model aerosol charge conditioners, local advection, diffusion and electric field transport are considered along with detailed unipolar and bipolar diffusion charging and photoelectric charging theory. However, the implementation must be carefully verified against reference data as shown in the following sections.

3 Model verification

The implementation of the electric charging equations is verified by the limit cases in three 0-D test cases corresponding to unipolar diffusion charging, bipolar diffusion charging, and photoelectric charging mechanisms, respectively. The 3-D particle transport equation, Eq. (1), is simplified as

$$\frac{dN_{b,q}}{dt} = S_{b,q,\alpha} + S_{b,q,\beta} \quad (11)$$

Without spatial derivatives, Eq. (11) forms a homogeneous linear ordinary differential equation and was numerically solved by the first order Euler method. In the absence of transport

phenomena, the test cases focus on the verification of time-dependent particle behaviours dictated by the coupled source terms and ion transport equations. The 0-D computational domain in OpenFOAM can be setup by a $1 \times 1 \times 1$ cubic domain with cyclic boundary conditions for all pairs of opposite faces. The physical constants used in the three verification cases are as follows. The Boltzmann constant k_B is $1.38065 \times 10^{-23} \text{ m}^2 \text{ kg s}^{-2} \text{ K}^{-1}$, the elementary charge is $1.60217 \times 10^{-19} \text{ C}$, and the vacuum permittivity is $8.85418 \times 10^{-12} \text{ F m}^{-1}$, which are used in Eq. (3). The mass of one molecule of carrier gas is set to $4.65173 \times 10^{-26} \text{ kg}$, used in calculating the mean free path of ions for Eq. (3).

3.1 Bipolar diffusion charging

The bipolar diffusion charging process was verified by replicating an example case of Hoppel and Frick [26] in which an initially uncharged aerosol is exposed to a region initially free of ions and approaches a “steady-state”, bipolar charge distribution as ions are introduced. In the zero-dimensional domain, the last term in the particle transport equation, Eq. (1), and the last two terms in the ion transport equations, Eq. (9) and (10) are the only terms governing this problem, and the other terms are disabled prior to solving.

Table 1 summarises the physical properties used in this example. Note that the attachment coefficient was not calculated by Eq. (3) but the values given in Hoppel & Frick [26] were directly employed to replicate that work. Their attachment coefficients are overall slightly higher than those obtained by Eq. (3).

Table 1: Physical properties used in the bipolar diffusion charging example case from Hoppel and Frick [26].

Quantity	Value	Unit
Particle diameter	200	nm
Electric mobility for positive ions	1.3×10^{-4}	$\text{m}^2 \text{ V}^{-1} \text{ s}^{-1}$
Electric mobility for negative ions	1.8×10^{-4}	$\text{m}^2 \text{ V}^{-1} \text{ s}^{-1}$
Mass of positive ion	200	AMU*
Mass of negative ion	100	AMU*
Ionization rate, γ	10^{11}	$\text{m}^{-3} \text{ s}^{-1}$
Recombination coefficient, α	1.6×10^{-12}	$\text{m}^3 \text{ s}^{-1}$

* Atomic Mass Unit

The concentrations of monodisperse particles with diameter of 200 nm were solved for charge levels, q , between -5 and +5. A concentration $N_0 = 10^{10} \text{ m}^{-3}$ of the neutral particle (zero charge) was set as an initial particle number concentration. The calculations ran for 100 s simulation time with a time step of 10^{-2} s .

Figure 2 exhibits the results from the present numerical simulation compared to the corresponding results in Hoppel & Frick [26]. The number of neutral particles decreases, while the number of charges on the particles increases, as ions are introduced to the system. The number of positive and negative ions increase almost identically. Particles acquire charge until a “steady-state” charge distribution is reached, in which the rate of charge transfer to particles with q charges to reach $q + 1$ charges (by positive ions) is equal to the rate of charge transfer to particles with $q + 1$ charges (by negative ions) to reach q charges [43]. The time required to reach a steady-state charge distribution is approximately 10 s in this case. Negative ions have a higher diffusivity than positive ions and therefore transfer charge to particles at a faster rate. The resulting steady-state charge distribution is asymmetric, carrying a net amount of negative charges, proportional to the logarithm of the ratio of mobilities of positive ions to negative ions [51]. The results of the present model are in excellent agreement with the results from Hoppel & Frick [26] with the maximum difference of “steady-state” particle number concentration of 4.2%, though values from Hoppel & Frick [26] (and other verification cases) are extracted from a pdf of the publication using graph digitization software.

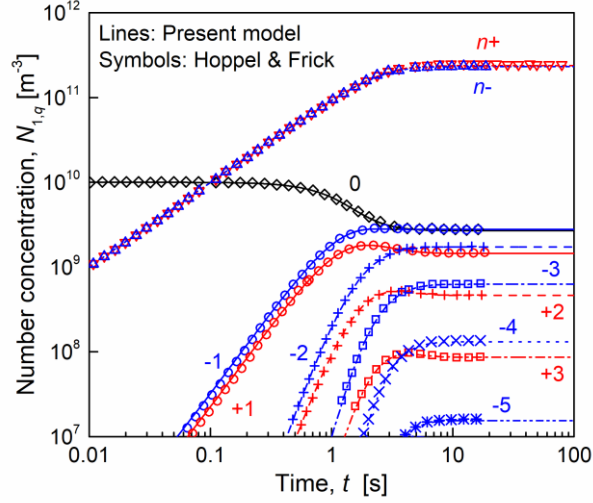


Figure 2: Bipolar diffusion charging of particles for diameter of 200 nm and ionization rate of $10^{11} \text{ m}^{-3}\text{s}^{-1}$. Comparison of the results of the present model and from Hoppel & Frick [26]. Black, red, and blue lines represent neutral, positive, and negative charges with the integer number representing q for each line.

3.2 Photoelectric charging

The implementation of photoelectric charging equations was verified by comparison with the case considered in Maisels et al. [23] that solves the charge levels acquired by monodisperse particles as a function of UV irradiation. Photoelectric charging model is modelled by the photoionization source term of Eq. (6) and the source term containing α in the negative ion transport equation in Eq. (10). The physical properties used in this example case are summarised in Table 2. The light energy $h\nu$ can be rewritten as hc/λ where c is the speed of light, and λ the corresponding wavelength, which yields the value of $h\nu$ as 7.20838 eV.

Table 2: Physical properties used in the photoelectric charging example case from Maisels et al. [23].

Quantity	Value	Unit	Ref.
Particle diameter	53	nm	[23]
Electric mobility for negative ions	1.35×10^{-4}	$\text{m}^2\text{V}^{-1}\text{s}^{-1}$	[23, 52]
Mass of positive ion	140	AMU	[53]
Mass of negative ion	101	AMU	[53]
Product of empirical constant and irradiation intensity, $K_c I$	4.38×10^{34}	$\text{J}^{-1}\text{m}^{-2}\text{s}^{-1}$	[23]

Potential barrier of the plane surface, Φ_{∞}	2.5	eV	[23]
Wavelength of UV irradiation, λ	172	nm	[23]

In this example case, residence time of particles within the irradiation region is 0.7 s, beyond which particles move into an irradiation-free region. For the zero-dimensional time-dependent calculation, this was represented by an initial run with photoelectric charging until 0.7 s, followed by a continuation without photoelectric charging that stops after 2 s. Four test cases with different total concentrations N_{tot} were considered in this example case. The initial concentrations were set to $N_{\text{tot}} = N_{1,0}$ with the values of 10^{14} , 10^{13} , 10^{12} , and 10^{11} m^{-3} , respectively.

As shown in Figure 3, the concentration of neutral particles decreases over time as particles are charged to increasingly positive charge levels by the photoelectric effect by Eq. (6). For the relatively low total particle concentration of $N_{\text{tot}} = 10^{11} \text{ m}^{-3}$, high, positive charge levels (e.g. +10 charges per particle) are reached as shown in Figure 3 (a). In comparison, for the relatively high total particle concentration of $N_{\text{tot}} = 10^{14} \text{ m}^{-3}$, lower charge levels are reached as shown in Figure 3 (b). At high particle concentrations, more negatively charged ions are generated by the photoelectric effect and diffusion (dis-)charging is more likely to occur for high particle and ion concentrations (of opposite sign) as shown in Eq. (12). Particles with more than 10 charges need to be calculated (using a separate transport equation for each charge level as in Eq. (1)), for accurate solutions of the case with $N_{\text{tot}} = 10^{11} \text{ m}^{-3}$, while they can be neglected for higher total concentrations. The modular code enables the user to select, at runtime, the range of charge states to solve, and therefore the number of transport equations.

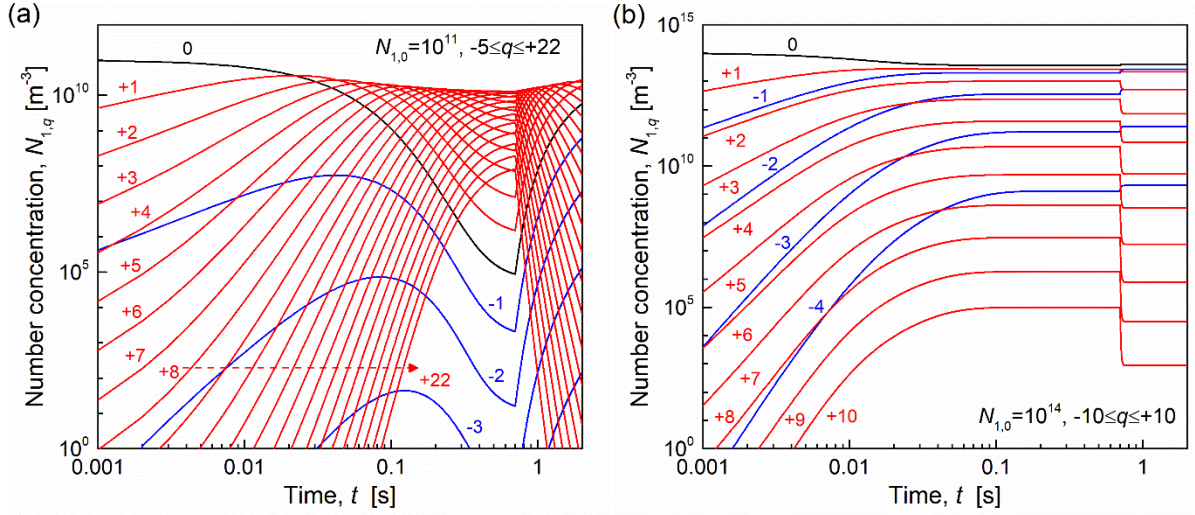


Figure 3: Photoelectric charging of 53 nm particles depending on the total number concentration N_{tot} and the number of charges q , (a) 10^{11} m^{-3} and $-5 \leq q \leq 22$, and (b) 10^{14} m^{-3} and $-10 \leq q \leq 10$. Particle concentrations outside the selected range of charge levels would have negligible contributions to the mean charge if calculated. Black, red, and blue lines represent neutral, positive, and negative charges with the number representing q for each line.

Figure 4 shows the mean charge per particle as a function of time for four different particle concentrations compared directly with Maisels et al. [23]. In this case, the range of charge levels, q , considered (i.e. a distinct transport equation is solved for each charge level, q) for $N_{\text{tot}} = 10^{11} \text{ m}^{-3}$ is -5 to $+22$, and for the other cases, q is -10 to $+10$. In the early stages of charging, all cases match an asymptotic solution [23] indicating that the mean charge is only a function of particle size and the intensity of the irradiation and does not depend on the particle number concentration. With fixed intensity of irradiation, the mean charge per particle reaches a steady-state condition (plateau) at which photoelectric charging is balanced by diffusional dis-charging which occurs at a higher rate for higher particle and ion concentrations. After the irradiation is turned off, the mean charge immediately decreases due to diffusional discharging and this occurs more rapidly for the case with higher particle concentration. The results of the present model are in very good agreement with those from Maisels et al. [23] and the maximum difference is 3.6%.

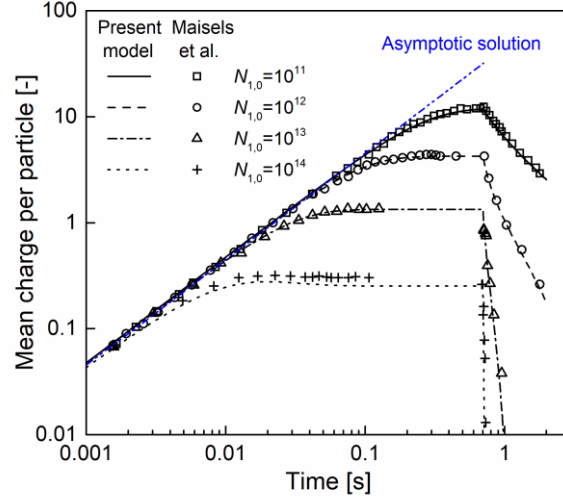


Figure 4: Mean charge per particle as a function of time for photoelectric charging of a monodisperse aerosol particles of 53 nm diameter at four different particle concentrations. The results of the present model are compared to the results from Maisels et al. [23].

3.3 Unipolar diffusion charging

Kaminski et al. [7] compare measured and predicted charge distributions of an opposed flow unipolar charge conditioner. Unipolar diffusion charging in the present model is verified by comparison with Kaminski et al. [7] using the same Fuchs diffusion charging model [43], properties and conditions as follows.

The mass and electrical mobility of the unipolar, positive ions are 290 AMU and $1.15 \times 10^{-4} \text{ m}^2 \text{V}^{-1} \text{s}^{-1}$, respectively, which follows approximately the Kilpatrick relationship between mass and mobility of ions [54]. Charge distributions are solved for distinct, monodisperse particle sizes between 10 and 500 nm. Thus, there is no interaction among particles of different sizes in this case (except particle-ion interactions), and the concentration of ions is fixed (at $7 \times 10^{12} \text{ m}^{-3}$). Therefore, the solution process was simplified in this tutorial case by setting parameters of a lognormal distribution: number of size bins, geometric standard deviation, and count median diameter to 100, 1.8, and 70 nm, respectively, to generate the files for the boundary and initial conditions for the particle transport equations, and then prescribing an initial concentration of $1 \times 10^5 \text{ m}^{-3}$ at each particle size to give a fixed concentration at each particle size bin considered. The considered charge range is $-2 \leq q \leq 20$. With the time step for calculation of 1 ms, the calculation was run for 1 s of physical time.

Figure 5 shows the charge fractions (fractions of particles at each discrete charge state) as a function of particle diameter during the unipolar charging process. Larger particles carry higher discrete charge levels when exposed to the same unipolar ion concentrations as smaller particles. The model predictions are in very good agreement with the Kaminski's Fuchs model by maximum relative error of 1.6% over a range of particle sizes and discrete charge levels from 0 to +8 charges per particle.

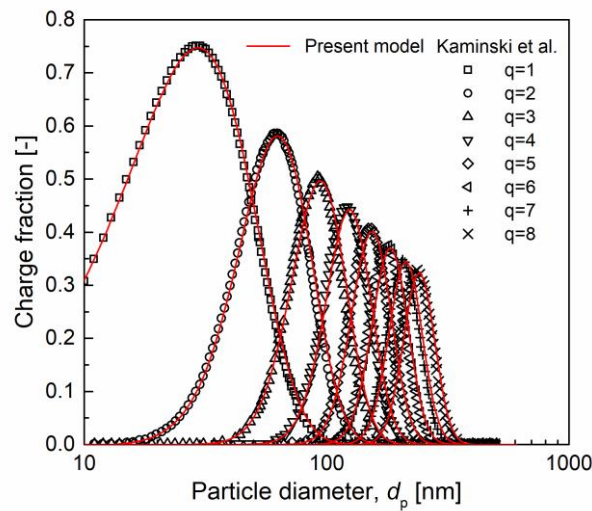


Figure 5: Unipolar charge distributions for 1 to 8 elementary charges obtained from the present model and a Fuchs model from Kaminski et al. [7].

Kaminski et al. [7] discuss that, due to some quantifiable disagreement with their experimental data, Fuchs' model may be used to predict average charge levels, but cannot deliver accurate charge distributions for the charger considered, and an empirical approach is required. This conclusion highlights that while bipolar charge distributions are well modelled and characterized, particularly for spherical particles [35], and considered to be known *a priori* for ISO standard measurements [4], more work is required for accurate modelling of unipolar diffusion chargers considering a range of possible charging and transport effects. Comprehensive modelling of the temporo-spatial dynamics of particle charging coupled with particle transport represents an opportunity for improved design and optimization of aerosol instruments. The models demonstrated in this work may provide a basis for validating unipolar chargers with experimental data.

4 Tutorials

Three tutorials for 3-D steady-state particle charger simulations are discussed to familiarise users with the implementation of the openAerosolCharging code in OpenFOAM and demonstrate the capabilities of the model. Figure 6 presents the file structure of openAerosolCharging code on top of OpenFOAM file structure. Three tutorials cases are described here: (1) a unipolar diffusion charger, (2) a bipolar diffusion charger, and (3) a photoelectric charger. In the solution procedure for these examples, the flow through the 3-D charger channel is predetermined, and the particles classified into discrete numbers of the size and charge are calculated. The following subsections introduce the test procedure, boundary conditions, and results in detail. To represent typical operating conditions for aerosol measurement devices, the properties of the carrier gas (i.e. $p = 1 \text{ atm}$, $T = 20^\circ\text{C}$, $\rho = 1.2 \text{ kg m}^{-3}$) are specified for dry air at normal temperature and pressure (NTP). Gas viscosity is calculated as $1.82 \times 10^{-5} \text{ kg}\cdot\text{m}^{-1}\text{s}^{-1}$ according to Kim et al. [55]. The carrier gas properties may be adjusted at runtime and standard OpenFOAM flow solvers may be used

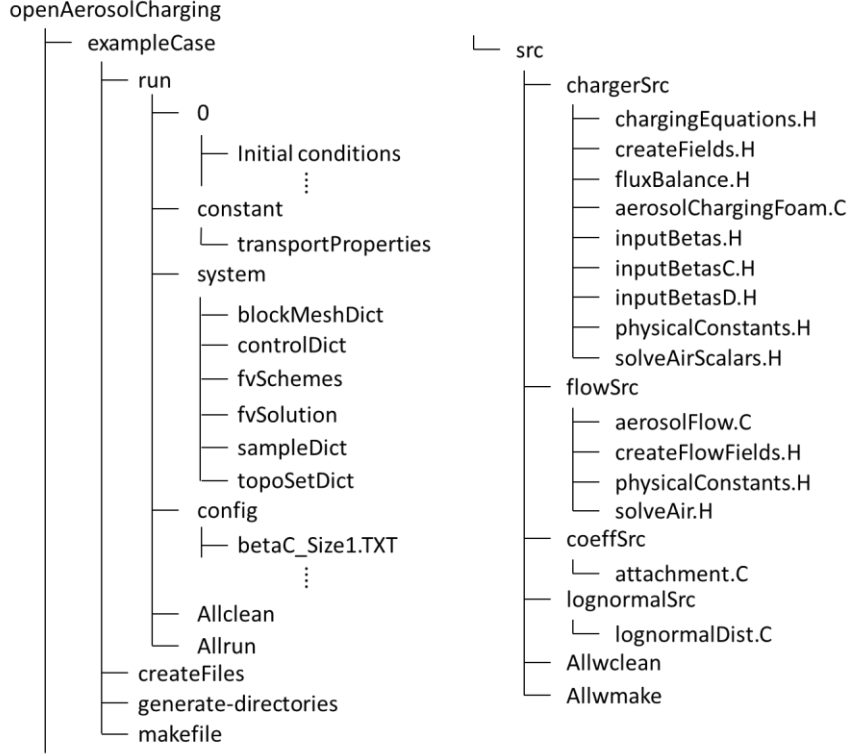


Figure 6: File structure of the aerosolChargingFoam solver (src) and tutorial cases (exampleCase).

4.1 Test procedure

The main procedure, which is common to all tutorial cases, is coded following the OpenFOAM style guide [56] where possible. In addition, several C++ and shell scripts are utilised for generating lookup tables for model parameters and evaluating solutions for visualisation. In the ‘makefile’ (located in the top level folder of each test case) commands are predefined for running the test cases as shown in Figure 7. The first step is generation of the computational mesh by ‘make mesh’ command. This generates computation mesh based on the information written in ‘blockMeshDict’ at ‘/run/system/’. Then, any regions of the computational geometry which require special consideration (e.g. defining an irradiated region vs. a non-irradiated region) are defined by running ‘topoSet’. After mesh and topology are defined, the flow that is not affected by particle motion can be precalculated by the command ‘make runFlow’. The flow solver based on icoFoam solves incompressible laminar Navier-Stokes equations without time derivatives using the PISO algorithm.

```

mesh:
    ( cd run && blockMesh | tee log.blockMesh)
    ( cd run && topoSet | tee log.topoSet)

runFlow:
    ( cd run && aerosolFlow | tee log.flow)

runCharging:
    (./createFiles)
    (./generate-directories )
    ( cd run && ./Allrun )
    ( cd run && aerosolChargingFoam | tee log.run)

clean:
    ( cd run && ./Allclean )

```

Figure 7: Generic test procedure composed in makefile.

The initial and boundary conditions for each variable are defined in ‘/run/0’ directory, and all model parameters including those used for generating lookup tables are defined in ‘transportProperties’ as shown in Figure 8. The charging and transport terms in the equations may either be considered or neglected by indicating in the ‘transportProperties’ file at runtime. For example, the ion-ion attachment during bipolar charging process is activated by setting the Boolean ‘useIonIonAtt’ to ‘true’. Photoelectric charging is activated by setting ‘usePhotoCharging’ as true. For unipolar charging, the keywords ‘useIonIonAtt’ and ‘usePhotoCharging’ are disabled.

For particles, the command ‘createFiles’ generates the files required for defining boundary and initial conditions for each size and charge level (e.g., nPart0Size1) based on a common file starting with underscore (e.g., _nPart). After that, ‘generate-directories’ populates each discrete particle size bin with a number concentration according to parameters defined for a log-normal particle size distribution: number of size bins, b (‘Nbin’), geometric standard deviation (‘stdev’), count median diameter (‘CMD’), and total number concentration (‘Ntot’). Then, ‘generate-directories’ generates lookup tables for the attachment coefficients β solved for the particle sizes and charge levels considered. The command ‘aerosolChargingFoam’ starts running the solver and the output is saved to a log file. Each command can be executed by ‘make’ command, e.g. ‘make runCharging’ for running the charging solver. For evaluation of the zero-dimensional verification cases, time evolutions of particle and ion number concentration were retrieved at the centre of the domain by using ‘probe’ function defined in ‘/run/system’. Prior to using openAerosol, and for details on other applications within the

OpenFOAM suite, it is recommended to consult the OpenFOAM User Guide and tutorial cases [33].

```

/*-----* C++ *-----*\
| ===== |
| \ \ / F i e l d | OpenFOAM: The Open Source CFD Toolbox |
| \ \ / O p e r a t i o n | Version: 9 |
| \ \ / A n d | Web: www.OpenFOAM.org |
| \ \ / M a n i p u l a t i o n |
\*-----*/

FoamFile
{
    version      9;
    format       ascii;
    class        dictionary;
    location     "constant";
    object       transportProperties;
}
// * * * * *
//General Simulation Settings
//useTransient      true; //use transient equations

//Physics used (terms in particle and ion equations)
usePhotoCharging    false; //photoelectric charging
usePartElecTrans    false; //particle electric field transport
usePartDiffTrans    false; //particle diffusion
useIonElecTrans     false; //ion electric field transport
useIonDiffTrans     false; //ion diffusion
useIonIonAtt        true; //ion-ion recombination
useFluxBalance      false; //use flux balance for 3D geometry

//Fluid Properties
rho    rho    [1 -3 0 0 0 0 0]    1.2041; //Constant gas density
mu     mu     [1 -1 -1 0 0 0 0]    1.82e-5; //Constant viscosity
TAir   TAir   [0 0 0 1 0 0 0]    293; //Constant gas temperature
mfp    mfp    [0 1 0 0 0 0 0]    66.5e-9; //Mean free path of gas

//Ion Properties
zIonsP  zIonsP  [-1 0 2 0 0 1 0]  1.35e-4; //Electrical mobility of +1 ions
zIonsN  zIonsN  [-1 0 2 0 0 1 0]  1.6e-4; //Electrical mobility of -1 ions
mIonsP  mIonsP  [0 0 0 0 0 0 0]    200; //mass of +1 ions
mIonsN  mIonsN  [0 0 0 0 0 0 0]    100; //mass of -1 ions

//Particle Properties
//Polydisperse, for monodisperse, use Nbin = 1 and stdev = 1
Nbin    Nbin    [0 0 0 0 0 0 0]    1; //Number of polydisperse size bins
CMD     CMD     [0 0 0 0 0 0 0]    100e-9; //Count median diameter
stdev   stdev   [0 0 0 0 0 0 0]    1; //For monodisperse, use stdev = 1
Ntot    Ntot    [0 -3 0 0 0 0 0]    1e10; //Particle number concentration
minQ     minQ     [0 0 0 0 0 0 0]    -5; //Minimum charge state to solve
maxQ     maxQ     [0 0 0 0 0 0 0]    5; //Maximum charge state to solve

//Diffusion Properties
ionsPGenRate  ionsPGenRate [0 -3 -1 0 0 0 0] 1e11; //Volume source for positive ions

```

```

ionsNGenRate ionsNGenRate [0 -3 -1 0 0 0 0] 1e11; //Volume source for negative ions
ionRec        ionRec [0 3 -1 0 0 0 0] 1.6e-12; //Volumetric +1/-1 ion recombination

//PhotoCharging Properties
KCI      KCI      [-1 -4 1 0 0 0 0] 4.38E36; //Empirical photoionization constant
exponent exponent [0 0 0 0 0 0 0] 2; // (hv-phi)^exponent
Rexponent Rexponent [0 0 0 0 0 0 0] 2; // d^Rexponent
distFromLight distFromLight [0 1 0 0 0 0 0] 0.02; //dist. from UV source to chamber
hv        hv        [0 0 0 0 0 0 0] 6; //light energy [eV]
Phi_inf    Phi_inf    [0 0 0 0 0 0 0] 4.5; //work function [eV]

//Solution output
units      fA; //electrical current units to display
iUnitConv iUnitConv [0 0 0 0 0 0 0] 1e15; //electrical current conversion to amps
minDisplay minDisplay [0 0 0 0 0 0 0] 1e-30; //minimum current to display in amps
fluxBalTol fluxBalTol [0 0 0 0 0 0 0] 0.001; //Solution tolerance for particle
                                              conservation (Number rate, #/s)
iBalTol    iBalTol    [0 0 0 0 0 0 0] 1e-19; //Solution tolerance for current
                                              conservation in Amps

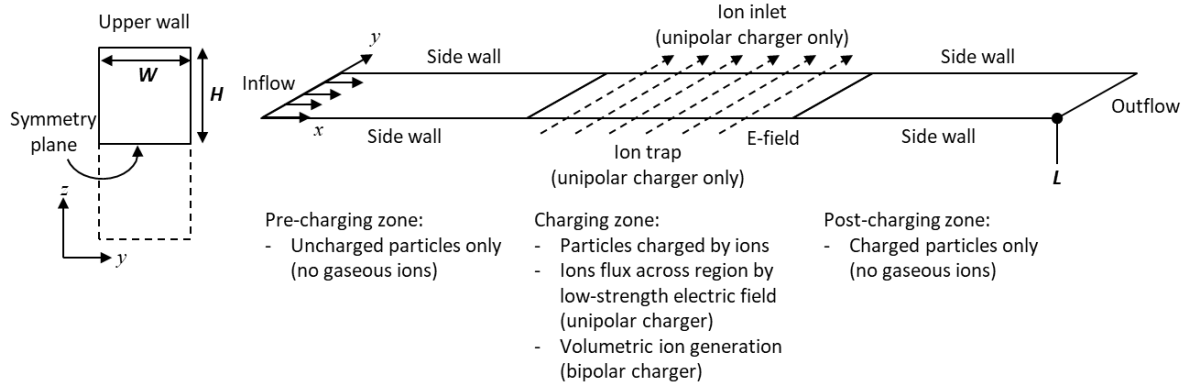
```

Figure 8: Generic example of ‘transportProperties’ to set the model parameters for the aerosolChargingFoam solver.

4.2 Geometry and boundary conditions

The geometry and boundary conditions for the three tutorial cases are depicted in Figure 9. The types and values of boundary conditions for each variable are summarised in Table S1-S3 in Supplement Information, SI 1.

(a) Unipolar or bipolar diffusion charger



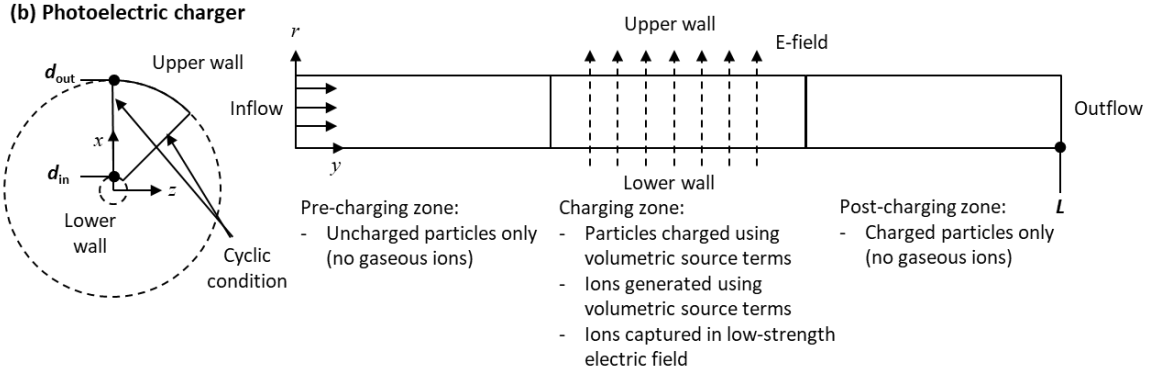


Figure 9: Geometry and boundary conditions for two tutorial cases corresponding to (a) square channel unipolar or bipolar diffusion chargers and (b) cylindrical geometry of a photoelectric charger.

4.2.1 Unipolar and bipolar diffusion charging

The example case of diffusion charging in this study is a charging channel wherein the particle flow is passed through an ion wind created by electric field transport of ions perpendicular to the flow direction. Figure 9 (a) shows the geometry and boundary conditions for the diffusion charger in the tutorial case. The geometry consists of three blocks as ‘Pre-charging zone’, ‘Charging zone’, and ‘Post-charging zone’ within a symmetrical rectangular flow channel with a length L of 60 mm and a cross section $H \times W$ of $4 \times 16 \text{ mm}^2$. The computational mesh of $70 \times 105 \times 70$ cells yields the total cell number of 514,500. Simple grading was applied such that the cell size near the wall is approximately four times smaller than the cell size in the middle of the channel. This charger geometry was used for both the unipolar charging and bipolar charging tutorial cases.

For both cases, the air flow entrains an uncharged, lognormal distribution of particles set by 8 particle size bins and geometric standard deviation of 1.7. The count median diameter and the total concentration were set to 50 nm and 10^{10} m^{-3} , respectively. In this test case, an indirect diffusion charger is employed where the ion source (e.g. corona wire) generates ions which pass through an ion inlet, pass through a charging region and are captured at an ion trap due to an externally applied electric field. The charging region is sufficiently shielded from the ion source such that field charging (which may occur near e.g. corona wire) is not significant. For this purpose, the ion trap voltage is set to -150 V applied for a distance of 4 mm between

electrodes such that only ions are significantly captured and particles remain in the advective flow. This ensures that most of mobile ions can be captured at the ion trap and do not contribute to the flux of charge at the outlet caused by the charged particles. Note that this flux of charged particles over time may be predicted by the model and represents sensor signals (i.e. electrical currents) of commercial aerosol sensors.

In the unipolar charging case, the ‘Ion inlet’ was used to represent a transmissive boundary condition for ions generated from the outside of the channel. The ion concentration at the ‘Ion inlet’ boundary is determined to match an averaged nt product, where n is the ion concentration and t is the characteristic time defined by the volume of the charging region divided by the volumetric flow rate. Positively charged ions are transferred from the ‘Ion inlet’ at a potential of 0 V and transferred to the oppositely located ‘Ion trap’ where a negative DC voltage is applied to capture excess ions. Thus, the positive ion concentration at the ‘Ion inlet’ was set to $n = 1.3 \times 10^{13} \text{ m}^{-3}$. The two boundary conditions for ions were not used in the bipolar charging tutorial. Instead, the positive and negative ions were generated by volumetric source (ionsPGenRate, ionsNGenRate in Figure 8) in the charging region, representative of volumetric ion generation by ionization of gas molecules by e.g. a radioactive source placed nearby within the charging volume. The ion generation rates for both positive and negative ions were set to $\dot{n} = 10^{15} \text{ m}^{-3}\text{s}^{-1}$. The recombination coefficient was $1.6 \times 10^{-12} \text{ m}^3\text{s}^{-1}$. For unipolar diffusion charger, the inlet velocity of the air flow was set to 0.26 ms^{-1} that corresponds to the flowrate of 1 L min^{-1} , while the flow rate for bipolar diffusion charger was 0.3 L min^{-1} .

4.2.2 Photoelectric charging

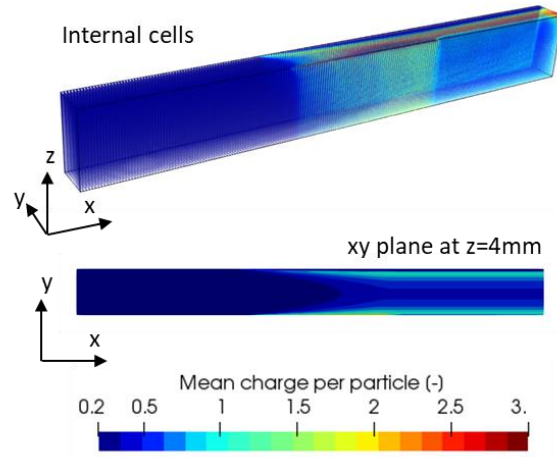
The geometry of the photoelectric charging tutorial case shown in Figure 9 (b) represents a typical photoelectric charger that consists of a cylindrical flow chamber including two concentric electrodes dictating electric field transport of ions and particles. The outer electrode is a cylinder d_{out} of 25 mm diameter and 200 mm length L , and the inner electrode is a concentrically located rod d_{in} of 1.5 mm diameter. A UV light source is placed at the particle inlet and light is directed towards the outlet of the chamber. To represent the non-collimated light intensity that decreases with the inverse square law in the flow direction, the distance from light source ‘distFromLight’ was set to 2 cm and the product $K_c I$ was fixed as $10^{34} \text{ J}^{-1}\text{m}^{-2}\text{s}^{-1}$. The computational domain corresponds to a 45° section of the tube with

symmetric boundary conditions in angular direction. The total number of cells is 6×10^4 cells with increasing cell density towards the walls where the highest concentration gradients occur. Since the transported particles and ions which reach the walls are assumed to be immediately captured, the charged particle and ion concentrations at the walls are set to zero.

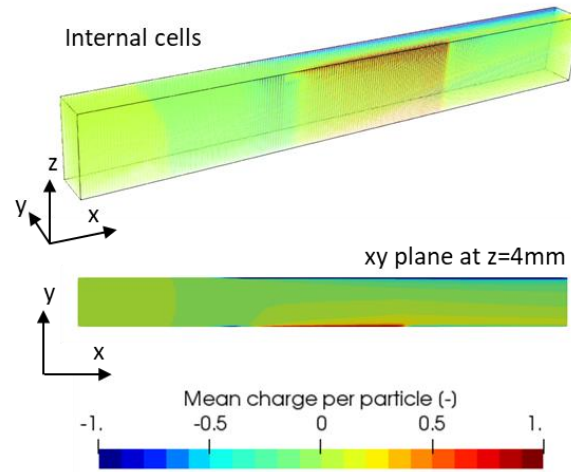
4.3 Results

The solver for the 3-D tutorial case captures the evolution of charge distributions over space and time, whereas the 0-D solver used for the verification cases described in Section 3 considered only the evolution over the corresponding convective time. For both diffusion charger cases, the calculation of flow for 100 PISO iterations (1 s with time step of 0.01 s in the ‘controlDict’ file) was preceded and the calculation of particle electric charging was conducted for 200 iterations (2 s with time step of 0.01 s). The conservation of particles and charges is confirmed after each solution iteration by performing an integral sum of the advective and diffusive fluxes through each boundary (e.g. walls, inlet and outlet) as well as summing number of particles and charge produced in volumetric source terms (if any). Conservation is confirmed when the summation of particles and charge through all boundaries and produced within the volume equals zero. The user specifies a tolerance for particle number and charge conservation and these tolerances are used as the convergence criteria for particle charging and transport. The ‘make view’ command generates the VTK files for visualisation. Figure 10 shows the distribution of mean charge per particle for the three tutorial cases. The calculation time depends on the number of discrete bins to represent the particle distribution. For the unipolar diffusion charging case, the calculation time for 8 bins and 16 bins is approximately 7 and 17 times that of monodisperse particle, respectively, while, for photoelectric charging, the calculation time for 8 bins and 16 bins is approximately 29 and 52 times than that for monodisperse particle, respectively [25]. The unipolar diffusion charger and photoelectric charger cases successfully replicated the test cases from Nishida et al. [25].

(a) Unipolar diffusion charger



(b) Bipolar diffusion charger



(c) Photoelectric charger

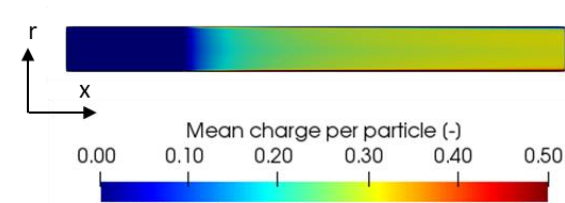


Figure 10: Distribution of mean charge per particle obtained from the (a) unipolar diffusion charger, (b) bipolar diffusion charger, and (c) photoelectric charger tutorial cases.

Moreover, the bipolar diffusion charger tutorial predicted physically reasonable results. Figure 11 shows the average charge fractions at the outlet obtained with four different monodisperse particle diameters. These charge fractions are extracted at the outlet of a 3-D

simulation, where a sufficiently long convective time enabled a “steady-state” charge distribution to be reached as was seen for a 0-D simulation in Figure 2 after a convective time of approximately 10 s. The results are very close to the theoretical steady-state charge fractions from Wiedensohler [57] and Gunn and Woessner [58], and deviations are attributed to small differences in ion-particle attachment behaviour compared with the model used ion-particle attachment model used (Eq (3)). Though there are complex spatiotemporal charging effects captured by this model, the resulting “steady-state” charge fractions are key to monitoring operation of charge conditioners. The series of tutorial cases shows that the validated 0-D model was successfully extended to the 3-D model of particle and ion conservation equations solved using flow-fields which were pre-determined by solving the Navier-Stokes’ equations. All equations were solved within the open-source platform, OpenFOAM.

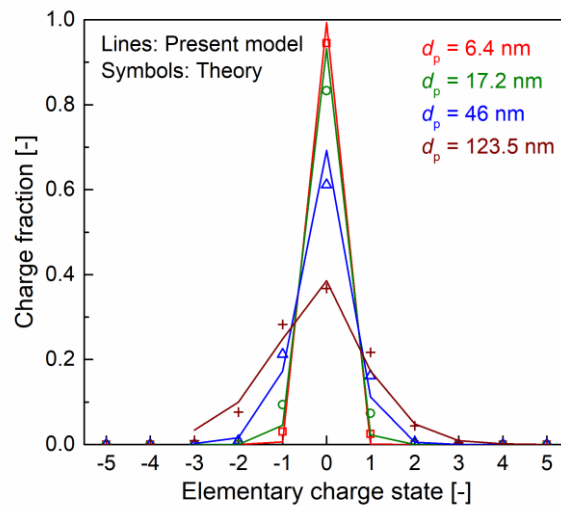


Figure 11: Average charge fraction at the outlet of a bipolar charger compared to the theoretical “steady-state” charge fraction from Wiedensohler [57] and Gunn and Woessner [58].

It is noted that while detailed verification of model implementation with the 0-D charging models available is appropriate and necessary, further verification and validation would provide more confidence in the models. The authors welcome other cases which include 3-D modelling of charging and transport for further verification of model implementation.

5 Conclusions

This study aimed to develop a comprehensive, open-source computer code for three-dimensional aerosol dynamics and electric charging including bipolar, unipolar, and photoelectric charging. The computer code includes more than 200 equations for transport and charging of polydisperse particle distributions in the CFD open source platform OpenFOAM. Three 0-D limit cases of charging models available in literature were implemented in the OpenFOAM platform and were verified by accurately replicating bipolar diffusion, unipolar diffusion and photoelectric charging processes. Comparison with the bipolar charging model of Hoppel & Frick [26] showed good agreement with a maximum difference of 4.2%, though values from Hoppel & Frick [26] (and other verification cases) are extracted from a pdf of the publication using graph digitization software. The photoelectric charging model of Maisels et al. [23] was replicated with a maximum difference of 3.6%. For unipolar diffusion charging, the model of Kaminski et al. [7] was implemented and showed a maximum difference of 1.6%. The detailed verification with 0-D charging models provides confidence in the correct implementation of the charging equations and provides a foundation for accurate modelling of charging phenomena. The implementation in OpenFOAM allows straightforward extension of the 0-D models for 3-D problems by coupling readily available transport equations. Three 3-D tutorial cases for bipolar diffusion, unipolar diffusion and photoelectric charging simulations coupled with computational fluid dynamics are used to describe the detailed implementation of the model and to demonstrate capabilities for comprehensively modelling practical aerosol systems. The source code and tutorial cases are released open-source on the openAerosol platform. This provides a framework for modelling of temporo-spatial dynamics of particle charging and transport for future developments of aerosol instrumentation.

6 Acknowledgements

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7 Nomenclature

a	Recombination coefficient, m^3s^{-1}
c	Mean thermal velocity of ions, m s^{-1}
D	Particle diffusivity, m^2s^{-1}
d_p	Particle diameter, m
$\mathbf{E}$	Electric field $\mathbf{E} = (e_1, e_2, e_3)^T$, V m^{-1}
e	Electron charge, C
$h\nu$	Specific photon energy, eV
k_B	Boltzmann constant, J K^{-1}
N	Particle number concentration, m^{-3}
n	Ion number concentrations, m^{-3}
p	Pressure, atm
t	Time, s
T	Temperature, K
$\mathbf{U}$	Velocity field $\mathbf{u} = (u_1, u_2, u_3)^T$, m s^{-1}
V_e	Elementary electron potential, V
Z	Particle electric mobility, $\text{m}^2\text{V}^{-1}\text{s}^{-1}$

Greek symbols

α	Combination coefficient for photoelectric charging, s^{-1}
α_c	Ion-particle collision probability, $-$
β	Attachment coefficient, m^3s^{-1}
γ	Ion generation rate, $\text{m}^{-3}\text{s}^{-1}$
ϵ_0	Vacuum permittivity, F m^{-1}
ρ	Density, kg m^{-3}
Φ	Particle surface work function, eV

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