Comparison between 1D radial and 0D global models for low-pressure oxygen DC glow discharges

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Abstract
This work focuses on the comparison between a zero-dimensional (0D) global model (LoKI) and a one-dimensional (1D) radial fluid model for the positive column of oxygen DC glow discharges in a tube of 1 cm inner radius at pressures between 0.5 Torr and 10 Torr. The data used in the two models are the same, so that the difference between the models is reduced to dimensionality. A good agreement is found between the two models on the main discharge parameters (gas temperature, electron density, reduced electric field and dissociation fraction), with relative differences below 5%. The agreement on other species average number densities, charged and neutral, is slightly worse, with relative differences increasing with pressure from 11% at 0.5 Torr to 57% at 10 Torr. The success of the 0D global model in describing these plasmas through volume averaged quantities decreases with pressure, due to pressure-driven narrowing of radial profiles. Hence, in the studied conditions, we recommend the use of volume-averaged models only in the pressure range up to 10 Torr.

Keywords: oxygen kinetics, discharge spatial resolution, global model, fluid model

(Some figures may appear in colour only in the online journal)

1. Introduction

Low-temperature plasmas and gas discharges are non-equilibrium systems, where species densities can be out of equilibrium, temperatures of electrons, ions and internal states can differ from the translational gas temperature, and electrons can follow non-Maxwellian velocity distribution functions. In these media, neutral and charged particles undergo collisional and radiative processes, energy transfers, transport and the effect of electromagnetic fields. Furthermore, low-temperature plasmas are multicomponent media, where phenomena from several branches of physics take place at different temporal and spatial scales. This makes it difficult to accurately characterize discharges and their output for applications either by measuring all the physical quantities of interest or by analytical models. As such, numerical models based on solid physical grounds and making use of increasingly
advanced computational capabilities emerge as complement-
tary tools to experimental diagnostics. They are necessary for
discharge characterization and understanding and for predict-
ing the behaviour of important physical and chemical quanti-
ties in different plasma systems. Given the wide diversity of
plasma sources and conditions (pressures, excitation sources,
power densities, spatial and temporal scales, working gases)
in the low-temperature plasma field, different formulations
and algorithms are used for numerical models. A broad over-
view and description of such models is provided in the top-
cical review ‘Foundations of modelling of nonequilibrium low-
temperature plasmas’ by Alves et al (2018).

One widely developed and employed class of plasma mod-
els that provides a compromise between accuracy, versatil-
ity and computational effort is that of fluid models. These
are based on the solution of the spatially and temporally
dependent hydrodynamic equations for the multiple species
that compose the plasma. As such, the plasma is seen from
a macroscopic point of view as a multi-fluid system of elec-
trons, ions and neutrals in a gaseous phase. Fluid models
for non-equilibrium gases have been developed for approxi-
ately a century, by deriving hydrodynamic equations as
moments of the Boltzmann equation, i.e. macroscopic con-
servation laws (Self 1966, Self and Ewald 1966, Meyyappan
and Kreskovsky 1990), together with the employment of
Chapman-Enskog’s methods (Chapman 1917, Enskog
1917, Ferziger and Kaper 1972). Concerning plasmas, these
macroscopic conservation laws typically apply to the follow-
ing quantities associated to each species: mass, momentum
and energy. The conservation equations are usually coupled
to Maxwell’s equations or Poisson’s equation, that describes
not only the electrostatic field externally applied but also the
one generated by charge separation (Self and Ewald 1966,
Forrest and Franklin 1968, Ingold 1972, Alves 2007). Plasma
fluid modelling requires considering multiple components
(Hirschfelder et al 1964, Giovangigli 1990, 2012, Giovangigli
et al 2010, Peerenboom et al 2014) and multiple tempera-
tures (Braginskii 1958, Chnielenski and Ferziger 1967, Graille
et al 2009, Orlac’h et al 2018). In low-temperature plasma
models, the three conservation laws (mass, momentum and
energy) are not always necessary for all species. Often the
ions are considered thermalized with the neutrals and, depend-
ing on discharge parameters, the charged particle flux may be
defined by the drift-diffusion approximation instead of the full
equation for momentum conservation (Babaeva and Naidis
1997, Kulikovsky 1997, Alves et al 2018). Nevertheless, at
low pressures, it may be important to account for momentum
balance for both electrons and ions (Alvarez-Laguna et al
2020). While non-local effects on electron energy are gener-
ally included via the equation for the mean electron energy
(Abbas and Bayle 1981, Meyyappan and Kreskovsky 1990,
Park and Economou 1990, Feoktistov et al 1995, Braginskii
et al 2005, Volynets et al 2020), in cases when electron colli-
sion frequencies are sufficiently high as to justify a local equi-
librium between the energy gained by the electrons from the
applied field and the energy lost in electron collisions, the local
field approximation can be used (Boeuf 1987, Kulikovsky
1997, Grubert et al 2009, Markosyan et al 2015, Černák et al
2020, Dhali 2022). In order to solve the equations for elec-
tron properties, accurate electron transport parameters and
electron-neutral rate coefficients are needed in fluid models.
These are often obtained from calculations of the electron
energy distribution function (EEDF) by coupling the fluid
equations to the numerical resolution of the kinetic electron
Boltzmann equation (EBE) (Hagelaar and Pitchford 2005).
Fluid models can be solved in the whole three-dimensional
space, or they can take physical assumptions on some dimen-
sions and then be solved in only one or two remaining dimen-
sions. For instance, for some cylindrical discharges, azimuthal
symmetry may be assumed, and the plasma may be con-
sidered quasi-homogeneous in the axial direction, while hav-
ing steeper gradients in the radial direction. That can leave the
spatially-resolved fluid model resolution to the radial direction
alone.

Another class of plasma models is that of global models,
also called zero-dimensional (0D) chemical kinetics models.
As far as the authors know, the concept of global models
has been introduced by Lee et al (1994) and has since been
used in many works through the development and use of dif-
f erent codes, e.g. (Dorai and Kushner 2003, Monahan and
Turner 2008, Pancheshnyi et al 2008, van Dijk et al 2009,
Guerra et al 2019, Viegas et al 2020). Nevertheless, plasma
chemical kinetic models, in some contexts called collisional-
radiative models, were already being employed as early as in
1962 (Bates et al 1962, Gousset and Boulmer-Leborgne 1984,
Gousset et al 1989). An extensive review of global models is
provided in Hurlbatt et al (2017). Global models are spatially-
averaged models, that attempt to describe the plasma as a
whole. As such, they do not describe spatial gradients within
the plasma system, although they can use them as assump-
tions. Global models solve the same conservation equations
as fluid models, for volume-averaged quantities. In particular,
global models usually solve particle and energy rate-balance
equations. As these models are free from the computational
costs of increased dimensionality, they can often include a
comprehensive reaction set for dozens or hundreds of differ-
sent species. The balance equations in global models are based
on source and loss terms defined by chemical reactions and
by approximations to include effects of transport. The source
terms associated with electron impact reactions are usually
dependent on the EEDF, and thus global models are often
coupled to EBE solvers. With these features, global models are
very useful to reproduce trends and identify the most import-
ant source and loss mechanisms of the species of interest
for applications in different conditions. It should be pointed
out that zero-dimensional chemical kinetics models can also
be used as local models, rather than volume-averaged global
models. This is a valid approach when local phenomena that
determine species densities evolution, such as chemical reac-
tions, are expected to have a much shorter characteristic time-
scale than non-local transport phenomena, which is typically
the case at near-atmospheric pressures (Liu et al 2010, Lietz
and Kushner 2016, He et al 2021, Passaras et al 2021).

When addressing plasma simulation results, namely from
fluid and global models, it is important to take into account the
degree of dimensionality implicit in the model and distinguish
spatially-resolved from spatially-averaged quantities. This is particularly important when comparing simulation results with experimental measurements, that can also be obtained as averages or spatially-resolved quantities, depending on the diagnostics (Hofmans et al 2020, Viegas et al 2021). Moreover, modellers need to weight whether the insight provided by increased dimensionality compensates the higher computational cost, or potentially the lower insight into plasma kinetics, associated to it. While fluid models can provide spatial resolution to some important quantities in the plasma system, the spatially-averaged approach of global models may be precise enough to identify the main energy transfer pathways and when studying so-called homogeneous discharges. These are plasmas where gradients are smooth or where spatial profiles are well known, allowing to define spatial averages as representative of the plasma as a whole. An example of discharges where this should be the case are low pressure cylindrical discharges controlled by free diffusion or by ambipolar diffusion, where the radial distribution of electron density ($n_e$) follows a paraboloidal or a zero-order Bessel function of the first kind, with zero at the cylinder wall (Schottky 1924, Parker 1963, Ikegami 1968, Durandet et al 1989, Fridman and Kennedy 2004, Lieberman and Lichtenberg 2005, Moisan and Pelletier 2012).

In this work, we study the positive column of low-pressure cylindrical DC glow discharges, to assess under which conditions are global models precise enough, in comparison with spatially-resolved fluid models. These discharges are chosen as test-case for being widely studied and modelled homogeneous plasma configurations (Schottky 1924, Gousset and Boullmer-Leborgne 1984, Gousset et al 1989, Meyyapan and Kreskovsky 1990, Raizer 1991, Cenian et al 1994, Gordiets et al 1995, Ivanov et al 1999, Fridman and Kennedy 2004, Lieberman and Lichtenberg 2005, Alves 2007, Dyatko et al 2008, Golubovskii et al 2011, Gudmundsson and Heicmovic 2017, Silva et al 2018, 2021, Volynets et al 2018, Booth et al 2019, Morillo-Candas et al 2019, Naïdis and Babaeva 2021). We further choose oxygen glow discharges as a case of interest, due to its suitability to study the kinetics of oxygen species, which undergo all the important processes taking place in other molecular gases, such as dissociation, attachment, electronic excitation and vibrational excitation. Moreover, oxygen species are frequently cited as being important for applications. One example is the interest in the kinetics of atomic oxygen, which, among other applications, is crucial for product separation in plasmas for electrochemical conversion (Rohrke et al 2004, Meiss et al 2008, Patel et al 2019, Chen et al 2020, Pandiyan et al 2022, Zheng et al 2022). In fact, kinetics in low-pressure cylindrical oxygen DC glow discharges have been recently extensively studied through both diagnostics and simulations in the pressure range 0.2–10 Torr and the discharge current range 10–40 mA (Booth et al 2019, 2020, 2022). The model employed in Booth et al (2019, 2022) is a 1D fluid model radially-resolved, developed at Moscow State University, that solves the equations for particle conservation for the different species in the plasma, together with Poisson’s equation and one equation for energy conservation.

In the current work, the discharge configuration studied in Booth et al (2019, 2020, 2022) is examined, for a current of 30 mA and pressures varying between 0.5 and 10 Torr, as a test-case to compare different plasma models. The LoKI global model (Guerra et al 2019, Tejero-del Caz et al 2019, Silva et al 2021) is employed together with the 1D-radial fluid model in Booth et al (2019, 2022), considering the same plasma kinetics. In section 2 the two models are presented, together with a description of the discharge conditions under study. In section 3, the simulation results from both models are compared, providing a benchmark of the global model against the more accurate 1D model: firstly, radial profiles of important physical quantities obtained by the 1D model are compared with the radial profiles assumed in the global model; then, the benchmark proceeds by comparing the average values of several species densities, gas temperature and reduced electric field obtained by both models. Finally, an overview of the differences between the modelling results is given.

2. Methods and conditions

2.1. Discharge set-up under study

The discharge conditions examined in Booth et al (2019, 2020, 2022) are addressed in this work. The set-up consists of a DC glow discharge in O₂, ignited in a Pyrex tube of 1.0 cm inner radius and 56 cm length. The discharge current is kept at 30 mA and the pressure values are varied within the interval between 0.5 Torr and 10 Torr. The cylindrical tube outer surface is kept at a constant temperature of 50 °C. This is guaranteed by a water/ethanol mixture flowing through an outer envelope and connected to a thermostatic bath. The temperature drop across the Pyrex tube wall is considered to be negligible, of less than 2 K (Booth et al 2019). The distance between the hollow cathode electrodes (in side-arms) is around 50.0 cm. The anode is connected to a positive polarity high voltage power supply and the cathode is connected to the ground. The gas flow rate is kept low, between 3 sccm for pressures below 1 Torr and 10 sccm for 2 Torr and pressures above, as to ensure that the gas residence time ($\tau$) is longer than the lifetime of all the active species in the discharge. The leak rate of air into the system is small, below 0.015 sccm, corresponding to less than 0.4% N₂ in the mixture in the worst case. Such a N₂ concentration in O₂ gas is not expected to have any noticeable effect neither on the discharge parameters nor on the kinetics of oxygen plasma species. Hence an axially uniform plasma column is created with constant gas composition (almost pure O₂) and cylindrical symmetry in the cylindrical vessel.

2.2. Modelling data

The same reactions, cross sections and collisional probabilities are considered in both models (1D and 0D). These are based on the works by Ivanov et al (1999), Vasiljeva et al (2004), Braginskiy et al (2005), Kovalev et al (2005), Booth et al (2019, 2022). 7 charged species and 8 neutral species are described by the kinetic scheme: e, O⁻, O₂⁻, O₃⁻, O⁺,
O₂⁺, O³⁺, O(1P), O(1D), O(1S), O₂(X), O₂(a′Δg), O₂(b′Σ⁺), O₂(Δg) (an effective sum of the O₂(A₃Σ⁺, A₃Σ⁺, cΣ⁺, A₁Σ⁺) Herzberg states) and O₃. The table of reactions is presented in Braginskiy et al (2005). To these, the two following reactions are added, with corresponding rate coefficients:

\[ O_2^+ + O_2 \rightarrow O_2^+ + O_2 + O_2, \]  

(1)

\[ k_{O_2^+ + O_2} = 3.3 \times 10^{-12} \times \left( \frac{300 \text{K}}{T_g} \right)^4 \times \exp \left( \frac{-5030 \text{K}}{T_g} \right) \text{m}^3\text{·s}^{-1}, \]  

(2)

\[ k_{O_2(b) + O(1P)} = 10^{-16} \times \exp \left( \frac{-3700 \text{K}}{T_g} \right) \text{m}^3\text{·s}^{-1}, \]  

(4)

where \( T_g \) is the gas temperature expressed in units of K. The rate coefficients \( k_{O_2^+ + O_2} \) and \( k_{O_2(b) + O(1P)} \) are obtained from Kozlov et al (1988) and Booth et al (2022), respectively. Vibrations are considered in the solution of the EBE, by taking into account electron impact collisions of excitation of vibrational states of the ground electronic state \( O_2(X) \). However, no detailed vibrational kinetics of \( O_2(X,v) \) is considered in this work, since it is expected to have a negligible influence on the main discharge parameters assessed here: electron density, reduced electric field and dissociation degree. It has been checked that the rates stemming from vibrational kinetics are sufficiently small to not affect the main parameters. It should be noticed that the benchmark can proceed as long as the two models are coherent, as is the case.

Electron kinetics is described in both models by stationary homogeneous two-term EBE solvers. The cross sections for electron impact are mostly taken from Lawton and Phelps (1978) for \( O_2 \) collision partner and from Laher and Gilmore (1990) for O, as in Booth et al (2022). However, the momentum-transfer cross section for electron scattering with O has been used, as in Alves et al (2016). Having the same electron impact cross sections and the same type of EBE solver, it has been verified that the same EEDFs and electron impact rate coefficients are obtained in the two models. The verification has been obtained for a wide range of reduced electric field \( E/n_f \) for pure \( O_2 \) and for 20% O–80% \( O_2 \) mixtures. Moreover, the widely used EBE solver BOLSIG+ (Hagelaar and Pitchford 2005) has also been employed to confirm that the same results are obtained.

In both models, positive ions recombine at the surface with unit probability. The loss of neutral particles due to quenching or recombination at the wall surface is considered in both models by taking the wall loss probability \( \gamma_j \) of each species \( j \) as input parameter. \( \gamma_j \) can be related to the effective wall loss frequency for species \( j \) \( \nu_{j,\text{wall}} \), described by the following equation (Booth et al 2022):

\[ \nu_{j,\text{wall}} = \gamma_j \frac{n_j,\text{nw} \nu_{j,\text{th}}}{m_j} \frac{2}{4 \pi R}. \]  

(5)

\[ v_j,\text{th} = \sqrt{\frac{8k_BT_{\text{nw}}}{\pi m_j}}, \]  

(6)

where \( n_j, \text{nw}, n_j, \text{av}, \nu_{j,\text{th}} \) and \( m_j \) are the near-wall density, the radially-averaged density, the thermal velocity and the atomic mass of species \( j \). \( k_B \) and \( T_{\text{nw}} \) are the Boltzmann constant and the near-wall temperature and \( 2/R \) is the surface-to-volume ratio in cylindrical geometry. It should be noticed that, in the absence of significant gradients in species number densities, \( n_j, \text{nw} \approx n_j, \text{av} \), and thus equation (5) is reduced to \( \nu_{j,\text{wall}} \approx \nu_{j,\text{th}} \). The values of wall loss probabilities \( \gamma_j \) used in this work are outlined in tables 1 and 2.

The values of wall loss probability of O(1P) have been obtained from pressure-dependent loss frequency measurements in Booth et al (2019) and equation (5). The wall loss probabilities of \( O_2(a) \) and \( O_2(b) \) have also been calculated from experimental measurements, reported in Booth et al (2022) in the case of \( O_2(b) \) and not yet published in the case of \( O_2(a) \). Concerning excited states \( O_2(\Delta g) \) and \( O(1D) \), these are assumed to fully quench on the wall surface.

Both models consider that heat exchanges in the plasma volume take place via Joule heating and heat loss by the radiation from Herzberg states \( O_2(\Delta g) \rightarrow O_2(X) + h\nu \), together with heat conduction to the wall. Furthermore, they take as thermal data the gas heat capacity at constant pressure \( c_p \) and the gas thermal conductivity \( \lambda_p \). The thermal conductivity is assumed to be \( \lambda_p = \frac{33 \times 10^{-5}}{T_g^{0.78}} \text{J(s·m·K)^{-1}} \), based on the \( O_2 \) experimental data from Westenberg and Haas (1963), as in Booth et al (2022). The heat capacities of each component
2.3. 1D radial fluid model

A one-dimensional 1D(r) discharge and chemical kinetic model with the local electric field approximation for the EEDF is used. The standard continuity equations are used to calculate the spatial distributions of all charged and neutral gas species in the radial direction:

\[ \frac{\partial n_j(r,t)}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \left( D_j n_j \frac{\partial X_j}{\partial r} - q_j \mu_j E_r n_j \right) \right) + S_j(r,t) = 0, \]  

where \( n_j \) is the total gas number density and \( n_j \) and \( X_j \) are the concentration and mole fraction of a species indexed by \( j \). \( D_j \) and \( \mu_j \) are the diffusion and mobility coefficients of the corresponding species and \( S_j \) is the total rate of production-loss of the \( j \)th species through different reactions. \( E_r \) is the radial component of electric field, calculated from the solution of Poisson’s equation:

\[ \frac{1}{r} \frac{\partial}{\partial r} \left( r E_r \right) = \frac{1}{\varepsilon_0} \sum_j q_j n_j, \]

where \( q_j \) is the charge of each \( j \)th particle and \( \varepsilon_0 \) is the dielectric permittivity of free space. The pressure and temperature dependences of the diffusion coefficients for neutral and ionic components are assumed to be: \( D_j \propto T^{3/2}/P \) and \( D_+ \propto T^2/P \), following Raizer (1991). The values of the diffusion coefficients for neutrals in normal conditions are taken from Morgan and Schiff (1964) and Winter (1951) or can be calculated from the Lennard-Jones coefficients (Kee et al. 1986), giving close results. Diffusion coefficient values for ionic components are taken from Snuggs et al. (1971).

The EEDF is determined as a function of the local reduced electric field by solving the stationary homogeneous EBE for the EEDF (Tejero-del Caz et al. 2015). The EEDF is recalculated as necessary, to account changes in the local gas composition. The time-dependent equations for the particle number densities, the EEDF \( \phi(r,t) \), the gas temperature \( T_g(r,t) \) and the axial electric field \( E_z(r,t) \) are solved self-consistently until steady-state is reached and the input discharge current value is matched. The calculation time to obtain the steady state solution for each case/pressure can reach several hours, depending on the relative accuracy. However, the code has not been optimized for computational times. The use of implicit schemes for equation (7) and exponential schemes in the discretization of the EBE for EEDF calculations can reduce computational time if necessary.

2.4. 0D global model LoKI

The software used to assess the validity of global models in describing DC oxygen glow discharges is the Lisbon Kinetics (LoKI) model. LoKI comprises two modules, that in this work run self-consistently coupled: a Boltzmann solver (LoKI-B) for the EBE (Tejero-del Caz et al. 2019, 2021) and a Chemical solver (LoKI-C) for the global kinetic modelling of gases (Guerra et al. 2019, Silva et al. 2021). LoKI-B is an open-source tool, licensed under the GNU general public license and freely available at https://github.com/IST-Lisbon/LoKI. LoKI-C is not yet freely available.

Overall, LoKI provides a chemical and transport description of plasma species in zero dimensions for user-defined working conditions: mixture compositions, pressure, reactor dimensions, flow rate and excitation features. LoKI-B...
solves a space independent form of the two-term EBE for non-magnetised non-equilibrium low-temperature plasmas, excited by DC or HF electric fields (Tejero-del Caz et al. 2019) or time-dependent (non-oscillatory) electric fields (Tejero-del Caz et al. 2021), in different gases or gas mixtures. As such, it calculates the EEDF and macroscopic electron parameters, including collisional rate coefficients. LoKI-C solves the system of zero-dimensional rate balance equations for all the assigned charged and neutral species in the plasma, each with volume-averaged number density $n_j$, considering collisional, radiative and transport processes:

$$\frac{\partial n_j}{\partial t} = S^\text{chem}_j + S^\text{conv}_j + S^\text{diff}_j.$$  

(12)

$S^\text{chem}_j$ is the sum of the chemical source and loss terms of species $j$, given in this work by the collisional and radiative processes presented in section 2.2, and $S^\text{conv}_j$ and $S^\text{diff}_j$ are the transport loss terms, considering axial convective transport and radial and axial diffusive transport, respectively.

The axion convection term considered in equation (12) supposes the input of O$_2$ and the output of all species $j$ in a spatially-averaged way at the same frequency for all species, such that the number of atoms is conserved. The convective term is dependent on the flow rate $\Gamma_{\text{in}}$ (given in particles per second by $\Gamma_{\text{scm}}$×4.47797×10$^{17}$) and the cylindrical chamber volume $V$ (Silva et al. 2021):

$$S^\text{conv}_j = \frac{\Gamma_{\text{in}}}{V},$$  

(13)

$$S^\text{diff}_j = \frac{\sqrt{n_j \Gamma_{\text{in}}}}{n_{\text{g,0}} V},$$  

(14)

where $n_{\text{g,0}}$ is the gas density at the beginning of the simulation. For the low $\Gamma$ considered here (of a few sccm), the axial convection term has a negligible influence on the results.

The diffusion of neutral species is taken as in Guerra et al. (2019), based on the formula of Chantry (1987), that takes into account partial losses of species $j$ to the wall with a deactivation/recombination probability $\gamma_j$, such that:

$$S^\text{diff}_j = -\frac{n_j}{\tau^\text{diff}_j},$$  

(15)

$$\tau^\text{diff}_j = \frac{1}{D_j} \left[ \left( \frac{\pi}{L} \right)^2 + \left( \frac{J_0}{R} \right)^2 \right]^{-1} + \frac{2h \sqrt{\pi}}{v_j \gamma_j \tau_{\text{th}}},$$  

(16)

where $\tau^\text{diff}_j$ is the characteristic diffusion time and $v_j \tau_{\text{th}}$ is the thermal velocity of species $j$. $R$ and $L$ are the radius and the length of the discharge tube, respectively, $J_0 = 2.405$ is the first zero of the zero order Bessel function and $D_j$ is the diffusion coefficient of species $j$. $D_j$ is given by the simplified Wilke’s formula for multicomponent mixtures (Cheng et al. 2006, Guerra et al. 2019), with binary diffusion coefficients calculated as in Hirschfelder et al. (1964) (section 8.2, equation (8.2-44)) and Guerra et al. (2019) (section 4.1). Based on Lennard-Jones binary interaction potential parameters. Under the approach taken for the diffusion of neutrals, radial profiles of molar fractions of species $j$ tend towards uniformity when $\gamma_j < 1$, and to Bessel radial profiles when $\gamma_j \approx 1$ (Chantry 1987).

The diffusion of positive ions is described by classical ambipolar diffusion, considering the high-pressure limit of transport theories (Phelps 1990, Guerra et al. 2019). Indeed, in the conditions of the glow discharge positive column under study, the radial diffusion length $\lambda$ is about an order of magnitude higher than the positive ion mean-free-path $\lambda_+$. A condition for the use of classical ambipolar diffusion. However, a deviation from classical ambipolar diffusion is induced by the presence of negative ions. This influence is considered in this work, taking into account the effect of several negative ions on the electron density radial profiles. The approach taken is based on the work in Guerra and Loureiro (1999) and is further detailed in Dias et al. (2023).

LoKI-C includes also a gas/plasma thermal model, for the self-consistent calculation of the gas temperature. The model considers a 1D parabolic profile of gas temperature in the radial direction, assuming a nearly uniform heating source term and following the experimental measurements and 1D simulation results in Booth et al. (2019):

$$T_g(r) = T_0 - \left( T_0 - T_{\text{nw}} \right) \frac{r^2}{R^2}, 0 \leq r \leq R,$$  

(17)

where $R$ is the tube inner radius of 1 cm and $T_0$ is the peak temperature at $r = 0$. $T_{\text{nw}}$ is the near-wall temperature at $r = R$ and its calculation is detailed in equation (21). The average gas temperature used in the rate coefficient calculation in the 0D model is defined as $T_{\text{g,av}} = \frac{1}{2} (T_0 + T_{\text{nw}})$. Its temporal evolution is given by the gas thermal balance equation at constant pressure (Pintassilgo et al. 2014):

$$\sum_k c_{p,k} \frac{\partial T_g}{\partial t} = Q_{\text{in}} - 8 \lambda_g (T_g - T_{\text{nw}}).$$  

(18)

In the previous equation, $c_{p,k}$ is the heat capacity at constant pressure of each component ‘$k’$ and $\lambda_g$ is the gas thermal conductivity, defined in section 2.2. $n_k$ is the density of each component ‘$k’’ and $Q_{\text{in}}$ is the input power transferred to gas heating per unit volume. In this work, for coherence with the 1D model described in section 2.3, $Q_{\text{in}}$ is given by a Joule heating term ($Q_{\text{in}} = J_\text{e} \cdot \mathbf{E}$, where $J_\text{e}$ is the current density) and a heat loss by the radiation from Herzberg states: $\text{O}_2(\text{H}z) \rightarrow \text{O}_2(\text{X}) + \nu_\text{h}$. By neglecting the temperature drop inside the Pyrex tube, a fixed inner-wall temperature $T_{\text{nw}}$ is considered as equal to the outer-wall temperature of 50 °C or 323,15 K. It is assumed that there is a heat transfer between the gas and the wall, near its inner surface, defined by a flux:

$$\Gamma_{\text{nw}} = h_{\text{gas-wall}} (T_{\text{nw}} - T_{\text{w}}),$$  

(19)

where $h_{\text{gas-wall}}$ is the heat transfer coefficient between the gas and the wall. Simultaneously, the conductive heat flux in
the gas near the wall, taking into account equation (17), is defined as:

\[ \Gamma_{nw} = \lambda_g |\nabla T_g|_{nw} = \frac{4\lambda_g}{R} (T_g - T_{nw}). \]  \hspace{1cm} (20)

Together, equations (19) and (20) define:

\[ T_{nw} = \frac{4\lambda_g T_g}{4\lambda_g + h_{gas-wall} T_w}. \]  \hspace{1cm} (21)

Equations (18) and (21) are solved together to find the temporal evolution of \( T_g \). A \( h_{gas-wall} \) of 120 J \cdot s\(^{-1}\) \cdot m\(^{-2}\) \cdot K\(^{-1}\) has been used, as providing a good agreement with the gas temperature experimentally measured in Booth et al. (2019) for the whole pressure range considered. Further details about the thermal model solution are provided in Dias et al. (2023).

The working conditions and the plasma species described by the model are those outlined in sections 2.1 and 2.2. The rate balance equation (equation (12)) is not solved for electrons, as in each iteration the electron density \( n_e \) is given to the model and kept constant. LoKI solves the system of equations (equations (12), (18) and (21)) iteratively to guarantee that the steady-state solution satisfies the total input pressure (calculated via the ideal gas law) and quasi-neutrality (same number density of negative charges and positive charges), which determines the self-consistent reduced electric field \( E/n_e \) for the used value of \( n_e \). Moreover, it is guaranteed that the electron parameters employed are retrieved from the solution of the EBE obtained for the same gas mixture and reduced electric field \( E/n_e \) of the final steady-state quasi-neutral solution. Finally, the system of equations is solved iteratively for different input values of \( n_e \) to guarantee that, at steady-state, \( I = \pi R^2 q_0 n_e v_e \) provides the experimental discharge current of 30 mA; where \( q_e \) is the elementary charge of electrons and \( v_e \) is the electron drift velocity, calculated from the mobility and electric field magnitude simulated by LoKI. The relative tolerances for the pressure, quasi neutrality, mixture composition and discharge current cycles are, respectively, \( 10^{-2} \), \( 10^{-2} \), \( 10^{-4} \) and \( 10^{-3} \). Each of these cycles typically needs about 10 iterations to achieve such relative errors and each iteration takes a few seconds of computational time. As such, the final solution for each case/pressure is usually found within a few minutes. A schematic figure of the workflow of LoKI is provided in Silva et al. (2021) (figure 1).

3. Results

3.1. Benchmarking of radial profiles

The benchmarking of the 0D model starts by verifying that the radial profiles of the main physical quantities assumed in the 0D model match those obtained in 1D for the conditions under study. One of the main parameters determining the physics in the glow discharge is the translational temperature of neutral atoms and molecules \( T_g \), also called gas temperature. This is assumed in the 0D formulation to follow a parabolic profile (equation (17)). Figure 1 shows that this assumption is valid for a lower-pressure case (1 Torr) and for a higher-pressure case (10 Torr). This figure represents \( T_g(r) \) obtained from the 1D simulation results and from the 0D assumption, that uses \( T_{g,av} \) (also represented in figure 1) and \( T_{nw} \) calculated by LoKI.

It can be noticed in figure 1 that the 1D result of \( T_g(r) \), resulting from the 1D resolution of the heat equation, stands slightly below the 0D profile and is slightly flatter. However, the difference is lower than 10% and the 1D profile is very close to a parabola. Another feature that can be noticed is that \( T_g(r) \) is more centre-peaked (concave) for higher pressure, with a larger difference between \( T_{nw} \) and the peak \( T_0 \). This is determined by the increased collisionality with pressure (higher \( Q_m \) in equation (18)) that increases \( T_{g,av} \) and \( T_0 \), while \( T_{nw} \) is kept low due to wall cooling and finite thermal conductivity. Finally, it should be taken into account that, even though the 0D model assumes a parabola, only the average value \( T_{g,av} \) is actually used in rate coefficient calculations for volume reactions. Therefore, gas heating appears here as a dominant cause of radial gradients that are not fully described in the 0D model and increase with pressure. It could be more accurate for 0D models to consider the whole parabolic evolution of \( T_g(r) \) in the calculation of average rate coefficients (see also equation (34)). However, it is shown in this work that the use of \( T_{g,av} \) provides reasonable agreement with 1D simulations, which has not been significantly improved by a preliminary implementation of the alternative approach.

The profile of \( T_g(r) \) is directly related to that of the gas density \( n_g(r) \) via the ideal gas law. As such, it is determinant also for the main parameter accelerating electrons and determining electron impact rate coefficients, which is \( E/n_g(r) \). If the axial electric field magnitude \( E \) is assumed to be radially...
uniform and $T_g(r)$ is assumed to be parabolic, then $E/n_g(r)$ should also follow a parabolic profile. This assumption is verified in figure 2, for the same pressures studied in the previous figure, by comparing the assumed parabolic profiles of the 0D model with the 1D simulation results. Once more, the 1D radial profiles match closely the parabolic assumption and a more centre-peak profile of $E/n_g(r)$ is noticeable for higher pressure. Moreover, it should be noticed again that the average value $E/n_{g,av}$ is used to compute average rate coefficients in the 0D model, and not the parabolic profile $E/n_g(r)$.

Another main discharge parameter is the electron density $n_e$. In homogeneous low pressure cylindrical electropositive discharges where charged particle balance is controlled by electron impact ionization and ambipolar diffusion, $n_e$ follows a zero-order Bessel function of the first kind, with zero at the cylinder wall (Schottky 1924, Parker 1963, Ikegami 1968, Durand et al 1989, Fridman and Kennedy 2004, Lieberman and Lichtenberg 2005, Moisan and Pelletier 2012). This implies that the non uniformity of particle loss processes such as wall losses is a second factor inducing radial gradients in the plasma. The presence of negative ions induces a deviation from the Bessel profile (Gousset et al 1989, Guerra and Loureiro 1999, Dias et al 2023). In this work, for the sake of simplicity, we test the 1D simulation results against a Bessel assumption in figure 3. While the agreement is good for the 1 Torr case, it is degraded with pressure, and relative differences of 17% can be observed for the peak value of $n_e(r)$ at 10 Torr.

The increasing deviation of $n_e(r)$ from the Bessel profile with a change in plasma parameters (pressure, current) can be caused by the development of instabilities. The fact that $T_g(r)$, $E/n_g(r)$, $n_e(r)$ and other species densities all have centred profiles can produce non-linear effects whose intensity increases with pressure leading to discharge stratification and further contraction. The radial contraction of plasmas has been reported in many works (Kenty 1962, Petrov and Ferreira 1999, Kabouzi et al 2002, Dyatko et al 2008, Shkurenkov et al 2009, Golubovskii et al 2011, Wolf et al 2019, Viegas et al 2020, 2021, Vialetto et al 2022). The narrowing of the $n_e$ profile observed in figure 3 is not due to a departure from the ionization-diffusion regime, since in the current conditions and considering the chosen reaction scheme, diffusive losses are still the dominant charge loss process. In addition, we emphasize once more that all the studied discharge regimes in the present paper correspond to experimental conditions with a visible uniform radial discharge distribution, without striations (Booth et al 2019, 2020, 2022). Furthermore, the contraction is not caused by electronegativity, since according to the 1D simulation results, an increase of attachment in the centre of the plasma, on its own, leads to a flattening of $n_e(r)$ instead of a contraction. We attribute the pressure-driven change of $n_e$ radial profile in the positive column of the oxygen glow discharge to inhomogeneous gas temperature, which can lead to the so-called thermal-ionization instability with increasing pressure and current, as in other studies of plasma contraction (Martinez et al 2004, Moisan and Pelletier 2012, Shneider et al 2012, 2014, Ridenti et al 2018, Zhong et al 2019). Indeed, we have observed in figure 2 that $E/n_g(r)$ is centre-peaked and therefore the electron impact ionization rate coefficient $k_i$ ($k_i = \frac{1}{n_e} k_{O_2^- \rightarrow O_2^+} + \frac{1}{n_e} k_{O_2^- \rightarrow O^+}$) also has a centre-peaked profile. As a result, the assumption of radial homogeneity leading to a Bessel $n_e(r)$ profile is not generally satisfied. For the lower-pressure case of 1 Torr, the deviation of $E/n_g(r)$ from uniformity is small ($E/n_g$ varies only between 53 and 65 Td in the 1D simulation results) and hence the $n_e(r)$ profile is very close to a Bessel profile. Nevertheless, the inhomogeneous gas temperature observed in figure 1 increases with pressure and leads to a sharper gradient of $E/n_g(r)$ at 10 Torr, with values from the 1D results in figure 2 varying between 26 and 51 Td. Indeed, according to the 1D simulation results, while at 1 Torr $k_i$ varies only between around $2.4 \times 10^{-18}$ m$^3$·s$^{-1}$ at $r = 0$ and $5.4 \times 10^{-19}$ m$^3$·s$^{-1}$ at $r = R$, at 10 Torr the variation is much wider, between around $2.3 \times 10^{-19}$ m$^3$·s$^{-1}$ and
It has been shown in Booth et al. (2019) that the radial contraction (Zhong et al. 2019) can finally lead to the radial contraction (Zhong et al. 2019). The radial contraction cannot be captured by the 0D model, that always considers $n_e(r)$, based on the low radial variation of $n_e(r)$ under the Bessel assumption.

In the study of oxygen glow discharges, the spatial distribution of the number density of atomic oxygen is also of paramount importance, due to the high reactivity of this species. The radial profiles of $O^3(P)$ density are represented in figure 4, for 1 Torr (figure 4(a)) and for 10 Torr (figure 4(b)). The 1D simulation result is presented, together with the 0D-calculated average density and the profile obtained from it by assuming proportionality with the total gas density, which, according to the ideal gas law, gives: $[O^3(P)](r) = [O^3(P)]_{av} \times T_{g,av}/T_g(r)$. This assumption is based on an assumed radial uniformity of source (electron-impact dissociation) and loss (diffusion and recombination at the surface and in volume) processes of atomic oxygen, consistent with $\gamma_i \ll 1$ (see tables 1 and 2) in equation (16).

Figure 4 shows that, for both pressures, the 1D and 0D models provide $O^3(P)$ densities within the same order of magnitude but with slightly different spatial evolutions. For 1 Torr, the 1D simulation result approximately follows the ideal gas law, that provides a difference between minimum and maximum values below a factor 2. The 0D parabolic profile provides a good agreement near the wall but slightly lower values in the bulk of the plasma. However, for 10 Torr, the 0D parabolic profile provides a similar result to 1D in the bulk but a factor 2 higher near the wall. Indeed, at 10 Torr, the 1D radial profile of $O^3(P)$ density is almost flat, rather than parabolic. The reasons for not following the ideal gas law are related with diffusivity, O atom wall losses and the radial non-uniformity of $E/n_e(r)$ (see figure 2) and thus of the electron impact dissociation rate coefficient. It has been shown in Booth et al. (2019) that oxygen atoms in a similar DC discharge conditions are predominantly lost by recombination on the tube surface and hence the assumption of radially uniform losses is not fulfilled.

As a result of the previous analysis, we consider that, when using a 0D model without access to the resolved spatial distribution of $O^3(P)$ density, it is generally more accurate to consider flat profiles than parabolic ones. This consideration has implications on the calculation of source and loss terms in the rate balance equations (equation (12)) and on the calculation of wall loss probabilities from wall loss frequency measurements (equation (5)) by considering $[O^3(P)]_{nw} = [O^3(P)]_{av}$.

In this section, it has been shown that the radial profiles of some of the main discharge parameters obtained through 1D simulation results in the studied conditions are not far from those assumed in 0D models: $T_g(r)$ and $E/n_e(r)$ are approximately parabolic, $n_e(r)$ are near-Bessel and $[O^3(P)](r)$ are almost flat. The deviation from these assumptions generally increases with pressure, due to the pressure-driven narrowing of radial profiles, but is still rather small (below a factor 2) up to 10 Torr in the positive column of oxygen glow discharges. This shows that, even though the 1D model takes the full radially-resolved profiles and the 0D model uses only averaged quantities, it is valid to compare the averaged values from both models, as they refer to the same type of radial profiles. That type of comparison is the subject of the next section.

### 3.2. Benchmark of average values

To proceed with the benchmarking, $T_{g,av}$, $T_{nw}$ and the average values of $E/n_e$ are compared between the 0D and 1D simulation results for the whole range of pressures between 0.5 Torr and 10 Torr, in figure 5. It can be noticed that all the values are very close, with differences below 5%. $T_{g,av}$ obtained from the 1D model is always slightly lower than the one from the 0D calculations, while $T_{nw}$ is slightly higher in the 1D simulations, which reflects the flatter radial profiles of $T_g(r)$ in 1D results observed in figure 1. $E/n_e(p)$ follows the same evolution with pressure in both models, with maxima below 75 Td at 0.5 Torr.
and minima above 35 Td at 10 Torr. The electron temperature \( T_e(p) \) (not represented here) presents a similar profile, with maxima at around 2.7 eV and minima near 2.1 eV.

The comparison proceeds with the dissociation degree. Figure 6 shows the average molar fractions of O and \( O_2 \) for the several pressures. The calculation of the molar fractions includes all the \( O \) and \( O_2 \) neutral and ionized species considered in the models. The agreement between 0D and 1D models is good but decreases with pressure. Indeed, at higher pressures, the plasma is slightly (a few percent) more dissociated in the 0D model than in the 1D case, as already suggested by the results in figure 4. This small difference is justified mostly by the particle balance of \( O(3P) \), where electron impact dissociation of \( O_2(X) \) and \( O_2(a) \) are the main source terms, and volume and wall recombination are the main loss terms. As the \( E/n_e \) profile is centre-peaked and the \( O(3P) \), \( O_2(X) \) and \( O_2(a) \) profiles are convex (lower in the centre and higher on the edges) in the 1D simulation results at 10 Torr (see figures 2, 4 and 10), dissociation is hindered and recombination is promoted, relatively to the 0D results that only consider average values in rate calculations.

The reasonable agreement between 0D and 1D simulation results is also observed when analysing the average number densities of charged species. Figure 7(a) shows only the main charged species: electrons, \( O^- \) and \( O_2^+ \); since the densities of the remaining ions \( (O^-_2, O^-_3, O^+ \) and \( O_2^+ \) \) are negligible. While the average electron density always presents a good agreement between 0D and 1D simulation results, the plasma contains more (up to 25%) negative and positive ion densities in the 1D model than in 0D. The difference increases with pressure and is attributed to the centre-peaked profiles of \( n_e \) and \( E/n_e \) (see figures 2 and 3) that promote the production of negative and positive ions via electron impact reactions (attachment and ionization) in 1D simulations with respect to the 0D model, that considers only averages. Concerning the destruction of \( O^- \) and \( O_2^+ \), it takes place mostly via detachment by collisions with species with convex (lower in the centre and higher on the edges) radial profiles (\( O^- \) case) and via wall recombination (\( O_2^+ \) case). As these species have centre-peaked radial profiles (see figure 7(b)), their destruction is relatively decreased in 1D, which also contributes to the observed higher average densities.

Concerning excited state average densities, these are represented from 0D and 1D simulation results in figures 8 and 9. Despite the logarithmic vertical scale in the figures, it can be noticed that the agreement between the two models is quite good for most species. However, there is significantly more (a factor 3 at 10 Torr) \( O_3 \) average density in the 1D simulation results than in the 0D case. The deviation increases with pressure. It is important to assess this difference.

The main reactions of production of \( O_3 \) at the higher pressures, e.g. 10 Torr, and their rate coefficients (Braginskii et al. 2005) are:

\[ O(3P) + O_2(X) + O_2(X) \rightarrow O_3 + O_2(X), \]  
(22)

\[ k_{O(3P)+2O_2(X)} = 5.6 \times 10^{-41} \times T_e^{-2} \text{ m}^6 \cdot \text{s}^{-1}, \]  
(23)

\[ O^- + O_2(a) \rightarrow e + O_3, \]  
(24)
where only inference. Nevertheless, we should notice that the rate coefficients is inconclusive in terms of justifying the \( O \rightarrow \) reaction. Therefore, an analysis of the average densities of reactants is necessary for understanding the behavior of the reaction.

Conversely, the 1D results contain slightly less \( O \) than the 0D results, which promotes ozone production. Conversely, the 1D results contain slightly less \( O_2(a) \) and \( O(3P) \) averaged densities, which hinders ozone production. Therefore, an analysis of the average densities of reactants is inconclusive in terms of justifying the \( O_3 \) density difference. Nevertheless, we should notice that the rate coefficients for \( O_3 \) production decrease with \( T_g \). Despite similar \( T_{g,av} \), \( T_g \) is lower on the edges in the 1D model than in 0D, where only \( T_{g,av} \) is used (see figure 1). This is also the region where the reactants have higher number densities, as is shown in figure 10, where 1D radial profiles are compared with the averages used in the 0D model at 10 Torr. Together, these two factors determine higher source of \( O_3 \) in the 1D model than in 0D.

Moreover, regarding the losses of \( O_3 \), we should notice that at 10 Torr the main reactions of destruction of \( O_3 \) and their rate coefficients (Braginsky et al. 2005) are:

\[
O_2(a) + O_3 \rightarrow O(3P) + O_2(X) + O_2(X),
\]

\[
k_{O_2(a)+O_3} = 5.2 \times 10^{-17} \times \exp(-2840 \text{ K}/T_g) \text{ m}^3 \cdot \text{s}^{-1},
\]

\[
O_2(b) + O_3 \rightarrow O(3P) + O_2(X) + O_2(X),
\]

\[
k_{O_2(b)+O_3} = 1.5 \times 10^{-17} \text{ m}^3 \cdot \text{s}^{-1},
\]

\[
O(3P) + O_3 \rightarrow O_2(X) + O_2(X),
\]

\[
k_{O(3P)+O_3} = 7.68 \times 10^{-18} \times \exp(-2060 \text{ K}/T_g) \text{ m}^3 \cdot \text{s}^{-1},
\]

where \( k_{O_2(a)+O_3} \) and \( k_{O(3P)+O_3} \) are obtained from Baulch et al. (1984) and \( k_{O_2(b)+O_3} \) comes from Slinger and Black (1984).

The reactants for these loss processes (except \( O_3 \) itself), \( O_2(a), O_2(b) \) and \( O(3P) \), have slightly lower average densities at high pressures in the 1D simulation results than in the 0D case (see figures 8 and 9). Furthermore, the rate coefficients for \( O_3 \) destruction increase with \( T_g \), and thus are lower in the plasma edges, where most reactants, \( O_2(a), O(3P) \) and especially \( O_3 \), have higher densities (see figure 10). These two factors contribute to a relatively lower destruction of \( O_3 \) in the 1D model. We can conclude that the subtle differences in \( T_g \) and species densities induced by dimensionality lead to both higher production and relatively lower destruction of \( O_3 \) in the 1D model than in the 0D case. The difference could potentially be decreased by using parabolic profiles of \( T_g(r) \),

\[
k_{O_2(a)+O_3} = 1.9 \times 10^{-16} \text{ m}^3 \cdot \text{s}^{-1},
\]

\[
O_2(X) + O(3P) + O(3P) \rightarrow O_3 + O(3P),
\]

\[
k_{O_2(X)+2O(3P)} = 2.15 \times 10^{-46} \times \exp(345 \text{ K}/T_g) \text{ m}^6 \cdot \text{s}^{-1},
\]

where \( k_{O_2(a)+O_3} \) and \( k_{O_2(X)+2O(3P)} \) are respectively obtained from Rawlins et al. (1987), Belostotsky et al. (2005) and Steinfeld et al. (1987).

On average, the 1D results at 10 Torr contain slightly more \( O^- \) and \( O_2(X) \) than the 0D results, which promotes ozone production. Conversely, the 1D results contain slightly less \( O_2(a) \) and \( O(3P) \) averaged densities, which hinders ozone production. Therefore, an analysis of the average densities of reactants is inconclusive in terms of justifying the \( O_3 \) density difference. Nevertheless, we should notice that the rate coefficients for \( O_3 \) production decrease with \( T_g \). Despite similar \( T_{g,av} \), \( T_g \) is lower on the edges in the 1D model than in 0D, where only \( T_{g,av} \) is used (see figure 1). This is also the region

Figure 7. (a) Average number densities of main charged particles as \( f(p) \), from 1D and 0D models: \( e, O^- \) and \( O_2^+ \). (b) Radial profiles of the same densities, at \( p = 10 \text{ Torr} \).

Figure 8. Average atomic excited state densities as \( f(p) \), from 1D and 0D models: \( O^*(P), O^*(D) \) and \( O^*(S) \).
Figure 9. Average molecular excited state number densities as \( f(p) \), from 1D and 0D models: \( \text{O}_2(X) \), \( \text{O}_2(\text{a}) \) and \( \text{O}_2(\text{b}) \) (a); \( \text{O}_3(\text{Hz}) \) and \( \text{O}_3(\text{b}) \).

Figure 10. Radial profiles of the number densities of the reactants taking part in the main source reactions and loss reactions of \( \text{O}_3 \) at 10 Torr. Profiles from the 1D model (full line with \( \times \) marker) and uniform values used in the 0D model (dashed lines).

Figure 11 shows that, in the whole pressure range between 0.5 Torr and 10 Torr, there is a very good agreement (with differences below 5%) on average temperatures and reduced electric field obtained from 0D and 1D models. The disagreement on average species densities is also relatively low, but it increases with pressure. Indeed, the relative differences lie below 20% at 1 Torr and between 50% and 60% at 10 Torr. The fundamental reason for this increase appears to be the pressure-driven narrowing of radial profiles. As pressure increases, radial profiles become more centre-peaked and radial gradients become sharper in the 1D description. As a result, the success of 0D models in describing oxygen glow discharges through volume averaged quantities decreases with pressure. In the case under study, with 1 cm tube inner radius, in light of the results, we would recommend the use of 0D global models to describe this plasma in the pressure range up to 10 Torr, but not above, since the deviation with respect to radially-resolved results is expected to increase.

Different tube radii of 0.5 cm and 2 cm have been used for this study for a few pressure values. This has been done both keeping \( I = 30 \) mA, thus changing the current density, and keeping the average current density, thus changing \( I \). It
has been found that the upper limit of pressure of validity of global models to describe the positive column of oxygen glow discharges is mostly independent of the tube radius, within the \( R \) limits considered (\( R = 0.5 - 2 \) cm).

### 4. Conclusions

This work has focused on the comparison between different models for the positive column of low-pressure oxygen DC glow discharges. The models compared are a zero-dimensional (0D) global model, LoKI (Guerra et al 2019, Tejero-del Caz et al 2019, Silva et al 2021), and a one-dimensional (1D) radial fluid model (Booth et al 2019, 2022). In the cylindrical low-pressure glow discharges under study, azimuthal symmetry is assumed and the plasma is considered quasi-homogeneous in the axial direction, leaving the need for its description to the radial direction alone. In this manuscript we have assessed under which conditions is the spatially-averaged approach of global models precise enough in comparison with the radially-resolved fluid model. The discharge configuration examined is the one in Booth et al (2019), for a tube radius of 1 cm, a current value of 30 mA and pressures varying between 0.5 and 10 Torr. The data used in the two models (reaction scheme, electron impact cross sections, rate coefficients and thermal data) and the EBE solvers employed are the same, so that the difference between the models is reduced to dimensionality.

A good agreement has been found between the two models on the values and radial profiles of the main discharge parameters. Indeed, the gas temperature profiles \( T_g(r) \) obtained in the 1D simulation results are parabolic, as supposed in the global model. The same applies to the parabolic profile of reduced electric field \( E/n_g(r) \) and to the Bessel profile of electron density \( n_e(r) \). These profiles become more centre-peaked as pressure increases, and \( n_e(r) \) starts deviating from a Bessel profile at 10 Torr. Although the shape of these profiles is assumed in the global model formulation, only the average values \( T_{g,av} \), \( E/n_{g,av} \) and \( n_{e,av} \) are used in the source term calculations in 0D. This is a main difference of dimensionality that is aggravated as pressure increases.

The average values of \( T_g \), \( n_e \), \( E/n_g \), electron temperature \( T_e \), near-wall temperature \( T_m \) and dissociation fraction found with the two models are very similar for the whole pressure range considered and 1 cm tube radius, with relative differences below 5%. The agreement on species average number densities, charged and neutral, is slightly worse, with relative differences increasing with pressure from 11% at 0.5 Torr to 57% at 10 Torr. However, the results always have the same order of magnitude, with the only case of visible discrepancy (up to a factor 3) being the average number density of \( O \). That deviation has been attributed mostly to the centre-peaked profile of \( T_g(r) \), that is not considered in the 0D model.

The results analysed show that the success of 0D global models in describing the positive column of oxygen glow discharges through volume averaged quantities decreases with pressure, due to pressure-driven narrowing of radial profiles. Hence, for the studied conditions with tube inner radius between 0.5 cm and 2 cm, we would recommend their use only in the pressure range up to 10 Torr. Nevertheless, we point out that 0D models can be used as local models at higher pressures, in conditions where the characteristic time-scales of local reactive phenomena and species densities evolution are much shorter than those of non-local transport.

### Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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