nature portfolio

Peer Review File

Using a Resistor Network to Simulate Charge Transport in Organic Semiconductors through Injection Layers and Transport Layers

Corresponding Author: Professor Wolfgang Wenzel

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors of the manuscript have developed a resistor network-based computational scheme to determine current densities in organic semiconductor devices across various device architectures. The computational workflow has been shown to exhibit significant acceleration in determining the I-V characteristics of monolayer, bilayer, and doped systems. The authors claim that the method developed can be a suitable alternative to the kinetic Monte Carlo method for modeling multilayer organic devices. The manuscript is well-written, and the developed methodology will be of interest to both computational chemists and the device physics community. However, several points mentioned in the article need slightly more elaboration/justification, and I recommend publication of this article after the authors address the following points.

1. The resistor network model includes several parameters (occupational probabilities and Coulomb energies) that are extracted from a shorter KMC simulation until the system reaches a steady state (~10^4 steps). What is the variation in the current density calculated after the preliminary KMC simulation and the full KMC simulation (~10^6 steps)? Since the system already achieves a steady state after ~10^4 KMC steps, the average current density is not expected to change significantly afterwards. Please justify why the current density may change significantly over a longer KMC simulation run and why the resistor network method should be preferred over slightly longer KMC simulations, say with 25000 steps.

2. For monolayer systems, higher energetic disorders resulting in higher current is a surprising observation (even when the energetic disorder is increased by 10-fold), since detrimental effect of energetic disorder on charge transport in organic semiconductors is a well-known fact; screening of molecular materials has shown that on-site energy disorder has a negative correlation with charge mobility (Adv. Funct. Mater. 2020, 30, 2001906). The authors have described the effect resulting from the energy leveling between the system and the electrode. Please justify the validity of the constant current model, which does not differentiate between realistic materials with significantly different charge transport parameters.

3. Similar to comment 2, please justify the observation in Fig. 3, which shows similar current density even when the energy offset in the bilayer system has been tripled.

4. In the doped system, the change in current density going from 0.1% to 1% doping is much greater than the enhancement in the current density for higher doping (15%). Is it due to the Coulomb hole effect? Please comment on this.

5. The title of the article reads "… with high charge carrier densities", but no quantitative measure of this parameter is given in the article. Can the authors provide some numerical values for the charge carrier density in monolayer and bilayer devices?

6. Can the authors comment on how "…the prohibition of double occupation of sites is treated implicitly" in the resistor network model?

7. Can the authors comment on the use of occupation probability and Coulomb energy in Eq. 4 together, which affect each other in a concerted way? Can this be a reason for better agreement with the KMC results when the Coulomb interaction's

contribution in ΔE_{ij} is entirely dropped?

8. Please elaborate on the following statement in the manuscript – “These systems, commonly known as doped systems … into full-stack OLED simulations.” (page 2, line 75-78). How can high doping accelerate simulation speed?

9. Please justify the use of such a small value for electronic coupling ($j0 \sim 10^{-22}$ eV)? The value does not represent any realistic organic material to the reviewer's knowledge.

10. There are several minor typographical errors in the manuscript and in the reference section (e.g., page 2, line 58; page 5, line 100; page 10, line 260, 262). Please fix them.

Reviewer #2

(Remarks to the Author)
Referee Report on "Using a Resistor Network to Simulate Charge Transport in Organic Semiconductors with High Charge Carrier Densities" by Petkau, Özdemir, Symalla, and Wenzel:

In this manuscript, the authors present a Resistor Network (RN) model with time-independent resistances to simulate charge transport in organic semiconductors at high carrier densities. Their approach reproduces current densities for a variety of device architectures while reportedly reducing computational time by roughly an order of magnitude compared to conventional Kinetic Monte Carlo (KMC) simulations. While the topic is of potential interest, particularly as computationally efficient alternatives to KMC are always valuable for systems with large parameter spaces, I find the presented data too limited to support the general conclusions, and several conceptual and methodological aspects require clarification before the work can be considered for publication. My main concerns are outlined below.

1) High carrier density limit and relation to earlier work:
The authors emphasize the high carrier density regime and contrast their work with that of Cottaar et al., which was limited to lower carrier densities via a percolation-based approach. However, it remains unclear why the framework of Cottaar et al. could not be extended conceptually to the high-carrier limit by considering it as a low hole density limit. The authors briefly mention single-carrier simulations (line 50) and the heuristic neglect of Coulomb contributions (line 200), but a clear and detailed explanation of how the high-density limit is incorporated or approximated is missing. Since this is a central claim of the manuscript and appears already in the title, the physical and numerical treatment of this limit needs to be presented more transparently in the methods section.

2) Motivation and role of the KMC setup:
The proposed approach is motivated by the desire to reduce the computational cost of KMC simulations. However, the RN model itself is initialized using a short KMC run. This raises the question of whether the method merely accelerates the evaluation of observables that are already encoded early in the KMC trajectories, even if full convergence takes longer. The authors should clarify what information is extracted from the short KMC simulation, why it is sufficient, and under what assumptions this transfer of information remains valid.

3) Reported speed-up and scalability:
The claimed computational speed-up appears modest, approximately a factor of ten according to Fig. 5, with the KMC setup run representing the largest cost contribution. Given that KMC is readily parallelizable across many cores or machines, it is unclear how the proposed approach would enable simulations that are otherwise computationally prohibitive. A quantitative discussion of scalability on modern architectures, as well as an explanation of how the method can claim broader applicability than KMC given its dependence on KMC for initialization, would be necessary.

4) Sparsity and scope of the data:
I find the dataset presented too limited to justify broad conclusions. All results shown demonstrate good agreement between the RN and KMC methods, but no examples are given where bigger deviations occur. Including cases where the agreement deteriorates while analyzing the underlying reasons for this would help define the method's range of validity. Moreover, Figs. 2-4 contain only four data points per panel. Given the reported computational costs, I estimate that each data point, including the 20 replica runs used for error estimation, should require roughly a day or less of computation time, suggesting that a denser sampling, potentially sufficient to yield smooth trends, should be feasible and would considerably strengthen the manuscript.

5) Numerical details and reproducibility:
Since the manuscript is primarily methodological, full reproducibility is essential. The simulation parameters, including those used in Lightforge, should be reported comprehensively. While some are illustrated in Fig. 1 and I appreciate the illustration, a complete list should be provided either in the main text or in the Supporting Information.

Additional suggestions on presentation:

Line 58: Please correct the extra commas in numerical values.

Figure 1: Adding sub-panel labels (a), (b), (c) and referring to them in the text would improve readability.

Figures 2,3,4: It would be clearer if the titles or captions explicitly indicated the disorder parameter, e.g., by writing "σ = ...".

A short discussion of cases where the method is expected to perform well or fail would help readers assess its practical applicability.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have made the necessary changes in the revised version of the manuscript and have responded to all comments from both reviewers. The suitability of the resistor network-based computational framework for overall OLED-type device modelling is better explained in the current version of the manuscript, while the limitations of the method have also been outlined. I recommend publication of the article with minor revisions.

1. Use of the term ‘high charge carrier density’ in the title as well as in the abstract seems misleading. As the authors pointed out in the response to comment 5 of Reviewer 1, as well as in the response to comment 1 of Reviewer 2, the high-carrier density is used as a ‘targeted use case’ which is prevalent in the doped injection layers and ‘is not tied to the physical behavior of the system with a high carrier density’; the acceleration in calculation time results simply from the weeding out of state rattling during KMC simulations and frequent recalculations of Coulombic interactions. The calculated results for monolayer, bilayer, as well as the doped systems, highlight the transport phenomena across the interface between layers, rather than through the material itself (which is evident from the high output current with increasing disorder resulting from Fermi level alignment), and the usefulness of the RN method across the carrier density spectrum. Additionally, in the literature, high charge-carrier density usually corresponds to carrier density > 10^{20} cm^{-3} (e.g., J. Am. Chem. Soc. 2022, 144, 3005-3019). Therefore, it will be useful if the authors reformulate the term ‘high charge carrier density’ with something like ‘ at OLED injection layer’.

2. The revised manuscript contains some minor typographic errors (e.g., line 28 – ‘even though’ is misspelled); I request that the authors correct them for the final submission.

Reviewer #2

(Remarks to the Author)
In the revised manuscript "Using a Resistor Network to Simulate Charge Transport in Organic Semiconductors with High Charge Carrier Densities" by Petkau, Özdemir, Symalla, and Wenzel, the authors have addressed some of the previous points. While I appreciate the additional data points in Figs. 2–4, the sublabels a), b), c) in Fig. 1, and the inclusion of the input file, the manuscript edits appear minor, especially in comparison to the explanations provided in the rebuttal letter. Many points remain insufficiently clarified. My specific comments are as follows:

1) Inclusion of Rebuttal Figure: The rebuttal letter introduces a new Fig. 1, cited multiple times to respond to reviewer questions. It is unclear why this figure or a similar one, along with the associated discussion, was not incorporated into the manuscript itself.

2) Previous Question 1: My earlier concern regarding the physical and numerical treatment of high-density systems remains inadequately addressed. Minimal edits in the revised manuscript correspond to this point, and the rebuttal letter only partially clarifies the issue. While the authors explain why high carrier density matters, the manuscript still lacks a clear, quantitative description of how the model approximates or incorporates the high-density limit. Statements such as "high carrier density is not a limit but a targeted use case" are understandable but informal. The connection between the resistor network and the physical treatment of carrier interactions is not explicit. The heuristic neglect of Coulomb contributions is not discussed. Furthermore, the connection to Cottaar et al. is weak; mentions of high energetic disorder recovering percolation pathways are brief and the statement "We cannot see how this would translate into a high carrier density but low hole density limit" is unconvincing. These points must be addressed, also in the revised manuscript.

3) Previous Question 2: The revised manuscript does not properly clarify how the resistor network improves KMC simulations or why skipping calculations in the injection layer is justified. While the rebuttal letter claims this was clarified, I could not identify corresponding edits in the manuscript. The repeated use of Fig. 1 in the rebuttal letter suggests that the advantage of the resistor network is to bypass the injection layer calculation, but this should be explicitly discussed in the manuscript. Additionally, the frequent use of the word "could" in the rebuttal letter creates ambiguity: it is unclear what the authors are actually doing versus what is hypothetical.

4) Sparsity of Data and Applicability Limits: One of my main concerns from the previous review, regarding the sparsity of

data, was not addressed and was not included in the rebuttal letter. While additional data points are included which is appreciated, the manuscript still explores only a limited portion of the parameter space. There should at least be a discussion if not a demonstration of the limits or failure modes of the method. Such information is critical for the community to assess the reliability of the method and to avoid erroneous results and need at least be commented on in the revised manuscript.

All other points have been addressed satisfactorily. I am hopeful that I can recommended the manuscript for publication once these issues are fully resolved.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)
I have reviewed the revised manuscript and the authors' responses to my previous comments. The authors have addressed my concerns satisfactorily, in particular by incorporating additional discussion in the Methods and Discussion sections, clarifying the intended regime of applicability, and explicitly outlining the limitations of the resistor network approach. The inclusion of additional data and the new discussion of failure modes improve the manuscript and make its scope clearer for the reader.

Overall, I am satisfied with the revisions and believe the manuscript is now suitable for publication. I therefore recommend it for publication.

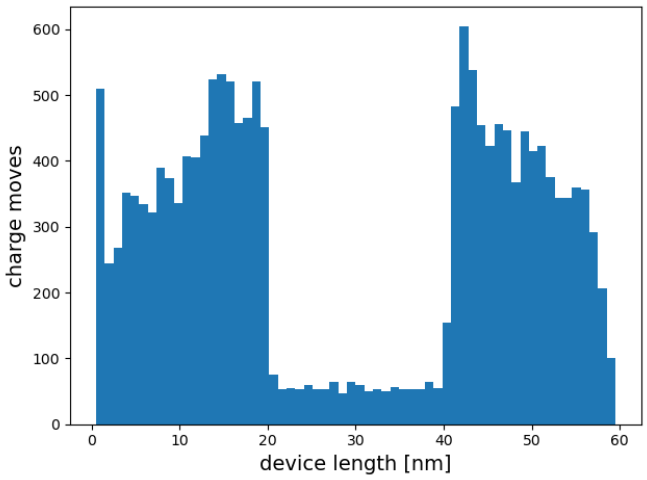


Figure 1. Move distribution in a system with two doped injection layers (doped at 15%) on the outside. The majority of charge moves are used in the injection layers.

1 Reviewer #1

1. The resistor network model includes several parameters (occupational probabilities and Coulomb energies) that are extracted from a shorter KMC simulation until the system reaches a steady state ($\sim 10^4$ steps). What is the variation in the current density calculated after the preliminary KMC simulation and the full KMC simulation ($\sim 10^6$ steps)? Since the system already achieves a steady state after $\sim 10^4$ KMC steps, the average current density is not expected to change significantly afterwards. Please justify why the current density may change significantly over a longer KMC simulation run and why the resistor network method should be preferred over slightly longer KMC simulations, say with 25000 steps.

Response: The use case of the resistor model is for doped injection layers, where KMC performs badly both for individual layers, but even more so in device simulations. This work was motivated by the problems we had in equilibrating device simulations with injection layers. As you recall, the rejection free KMC algorithm selects processes according to their rates. Since, essentially all charges in the device stem from Coulomb bound electron-hole pairs induced by doping these are so numerous, that essentially all numerical effort of the device simulation is spent on charges that oscillate back and forth in the injection layer[1]. This problem is relevant for KMC simulations with high carrier densities (i.e. many individual carriers to simulate). We have tried to make this point more clear in the revised manuscript.

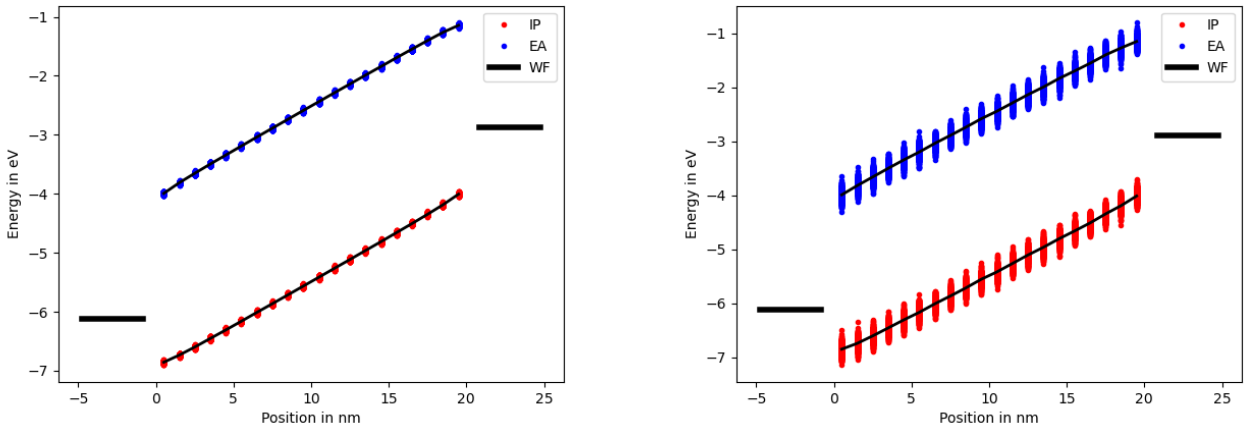
The advantage of the resistor network model is not strictly about simulation time of the individual layer, but we get rid of simulating this layer in the device simulation altogether. In such applications we calibrate the resistor network first for the doped injection layers until charge transport pathways are established (and hence included in the charge distribution). After this preliminary KMC simulation is finished, the occupational probabilities are extracted and put into the resistor network. Going forward, the full stack KMC simulation can use the resistor network to model the expensive layer at little to no extra computational cost. This will focus the performed KMC steps onto the other layers (by excluding expensive effects like the state rattling in the doped injection layer from the list of available KMC steps - leaving in its place only effective currents in and out of the injection layer).

While this coupling of the resistor network back into a KMC simulation was not yet done, the advantage (especially with these use-cases in mind) lies therefore not in any specific step count but rather in the static nature of the resistor network.

The problem and the solution is illustrated in Figure 1. The majority of the moves are used in the doped injection layers. This imbalance is due to the high carrier density fostering state rattling.

We agree with the reviewer that the KMC simulation should be equilibrated and we have just reported different simulation times to demonstrate this feature.

2. For monolayer systems, higher energetic disorders resulting in higher current is a surprising observation (even when the energetic disorder is increased by 10-fold), since detrimental effect of energetic disorder on charge transport in organic semiconductors is a well-known fact; screening of molecular materials has shown that on-site energy disorder has a negative correlation with charge mobility (Adv. Funct. Mater. 2020, 30, 2001906). The authors have described the effect resulting from



(a) $\sigma = 15\,\text{meV}$ **(b)** $\sigma = 100\,\text{meV}$

Figure 2. Energy alignment of the monolayer system at 3 V with energetic disorder $\sigma = 15\,\text{meV}$ and $\sigma = 100\,\text{meV}$. The IP and EA values are plotted as a scatterplot in red and blue for all sites in the system.

the energy leveling between the system and the electrode. Please justify the validity of the constant current model, which does not differentiate between realistic materials with significantly different charge transport parameters.

Response: We are grateful for this comment. We have a long history of publishing on this effect[2,3]. The point raised by the referee is unexpected but yields a nice illustration why the resistor model is needed. In many KMC stack simulations the injection layers are simply left out because it is argued that they can be replaced by metallic electrodes[4] because they are said to behave like metals.

Also, as we pointed out in earlier work on doping, the effect of doping is really intricate: additional disorder does not automatically lead to a decrease in the mobility[3].

To answer the specific question: While it is true that the disorder affects broadens the density of states it also changes the injection barriers. It turns out that for the model system the latter effect is stronger than the former.

The mentioned alignment can be seen in Figure 2. The higher disorder leads to better alignment between the energy level of the electrodes and the materials energy levels. This fosters injection into and ejection from the material.

3. Similar to comment 2, please justify the observation in Fig. 3, which shows similar current density even when the energy offset in the bilayer system has been tripled.

Response: We thank the referee for this observation. While we call this layer a Bilayer and emphasize the interface between the two organic layers, there are actually three interfaces in the system as there are also interfaces with barriers between the electrodes and the organic material. By altering the organic-organic interface also the interface of one organic layer with its electrode is altered. This alteration might compensate the alteration at the organic-organic interface to some extend making the overall current insensitive in the tested regime.

4. In the doped system, the change in current density going from 0.1% to 1% doping is much greater than the enhancement in the current density for higher doping (15%). Is it due to the Coulomb hole effect? Please comment on this.

Response: We appreciate the referees observation. Our doped system is similar to the one used by Özdemir et. al[3]. There, the same observation is reported and attributed to the Fermi level alignment between electrode and material. After increasing the doping concentration and achieving Fermi level alignment, a saturation is reached an further doping does not increase the current density further.

In our opinion the same explanation applies here.

5. The title of the article reads "… with high charge carrier densities", but no quantitative measure of this parameter is given in the article. Can the authors provide some numerical values for the charge carrier density in monolayer and bilayer devices?

Response: We agree that this prominent use of the high carrier density needs some clarification, even though reducing it to the mere number might not transport the application scenario fully. The charge densities are in the order of 10^{17} to 10^{18} carriers per cm^3 for the monolayer systems, in the order of 10^{16} to 10^{17} carriers per cm^3 for the bilayer systems and in the order of 10^{18} to 10^{20} carriers per cm^3 for the doped systems. With the fastest of our KMC simulations in the order of 10^{-4} seconds per KMC step and the slowest in the order of 10^{-2} seconds per KMC step, depending mainly on the number of carriers to simulate.

The large difference in KMCs calculation speed highlights the usefulness of the resistor network especially in these cases, where the high carrier density slows down the KMC simulation. The strength of the resistor network is not tied to the physical behavior of the system with a high carrier density but connected to the implications that result from it during simulation with KMC.

We made this focus to the application in these scenarios more clear in the revised manuscript.

6. Can the authors comment on how "... the prohibition of double occupation of sites is treated implicitly" in the resistor network model?

Response: We appreciate the referees question and will clarify where the prohibition of double occupation is treated explicitly and why we argue that in the resistor network it is only treated implicitly.

In the short preliminary KMC the Coulomb interaction is treated explicitly to find a set of self-consistent occupation probabilities. These occupation probabilities are used in the resistor formula to make hops more probable if the source site is occupied (high occupation probability) and the target site is unoccupied (low occupation probability). Hence, the occupation probability is a measure on *how forbidden* it is to hop onto this site. It is therefore forbidden in the resistor network to hop toward a site that is always occupied during the preliminary KMC and similarly for a site that is mostly occupied it is highly unlikely to hop onto it.

(Please note, that we use *hopping* here in reference to the hopping rates, even though no individual carriers hop in the resistor network and only time averaged currents are treated.)

We extended the explanation of the use of occupation probabilities in the revised manuscript.

7. Can the authors comment on the use of occupation probability and Coulomb energy in Eq. 4 together, which affect each other in a concerted way? Can this be a reason for better agreement with the KMC results when the Coulomb interaction's contribution in ΔEij is entirely dropped?

Response: We are grateful for the referees explanation and agree at least partially with it.

The Coulomb energy contribution in the energy term of the hopping rate is an important term in KMC simulation as it controls the charge distribution in the material and balances the use of disorder-driven energetically favorable pathways against clustering of like charges. This same balance is present in the use of site specific occupation probabilities. Any process that tries to produce these occupation probabilities will therefore have to consider the Coulomb interaction among charge carriers. In our case this consideration is explicitly present in the preliminary KMC simulation.

We therefore think, that since the charge distribution of carriers is already encoded self-consistently in the occupation probabilities, produced by the preliminary KMC simulation, it is sufficient to not consider Coulomb interaction explicitly in the energy term of the hopping rate. Doing so would lead to double counting as pointed out by the referee. Hence, the term was dropped.

Nevertheless, we do not think the double consideration of the Coulomb interaction is the problem responsible for the mentioned disagreement in the simulations using our Coulomb interaction distribution. We think, problematic are unaccounted for local correlations. While the occupation probabilities we computed are site specific, we were not able to produce site specific Coulomb interaction distributions. The distributions are therefore trying to model the entire device. This distribution is then not able to capture local correlation between neighboring sites (i.e. a trapped charge will have a permanent influence on neighboring sites). In our opinion, this explains the very high variance that makes the results unusable.

8. Please elaborate on the following statement in the manuscript – "These systems, commonly known as doped systems ... into full-stack OLED simulations." (page 2, line 75-78). How can high doping accelerate simulation speed?

Response: We apologize for the lack of clarity there and revised the manuscript accordingly. We will briefly elaborate on what was meant.

Due to the KMCs combinatorial calculation of interactions in systems with a high carrier concentration, the individual KMC step tends to become more computationally costly. Hence, by introducing a method without frequent recalculations there is a higher cost-saving potential in these systems. Hence, not the calculation itself is faster but the cost saving-potential is higher due to the higher initial cost with pure KMC simulation.

9. Please justify the use of such a small value for electronic coupling (j0 $\sim 10^{-22}$ eV)? The value does not represent any realistic organic material to the reviewer's knowledge.

Response: We are very grateful to the referee for pointing out what was presumably a false unit conversion during the writing of the manuscript. We corrected the value.

The value used for all simulations was 0.001 eV. For further clarification the input files for the KMC are directly attached. The correct value can also be seen as a default value in the resistor network code provided.

10. There are several minor typographical errors in the manuscript and in the reference section (e.g., page 2, line 58; page 5, line 100; page 10, line 260, 262). Please fix them.

Response: We thank the referee for pointing out these errors and fixed the LaTeX-formatting, the missing "-" and the DOI hyperlink formatting.

2 Reviewer #2

1) High carrier density limit and relation to earlier work: The authors emphasize the high carrier density regime and contrast their work with that of Cottaar et al., which was limited to lower carrier densities via a percolation-based approach. However, it remains unclear why the framework of Cottaar et al. could not be extended conceptually to the high-carrier limit by considering it as a low hole density limit. The authors briefly mention single-carrier simulations (line 50) and the heuristic neglect of Coulomb contributions (line 200), but a clear and detailed explanation of how the high-density limit is incorporated or approximated is missing. Since this is a central claim of the manuscript and appears already in the title, the physical and numerical treatment of this limit needs to be presented more transparently in the methods section.

Response: We thank the referee for raising this point, which highlights the need for a clearer discussion on why we are referring to high carrier density systems. In response, we will first highlight why the previous work mentioned is not applicable for these systems and further emphasize why we were referring to these systems in the first place.

Cottaar et al.[5] state themselves that their approximations during the calculations of the conductances is not valid for higher charge densities (see Eq. 1a and 1b in their paper). We can not see how this would translate into a high carrier density but low hole density limit.

If one wants to see the approach from Cottaar et al. as a special case of our resistor network, this would be the limit of high energetic disorder. In this case strong percolation pathways would be present, most of our sites would be never occupied during the preliminary KMC, effectively cutting off these parts from the network. The remaining effective network would correspond then to their percolation pathways.

In contrast to Cottaar et al. we do not use the carrier density explicitly during calculation or for some approximation. (Apart from that we assume that charge movement is sufficiently sampled and examined for a long enough time for time averaged currents to be meaningful and large enough to not run into numerical problems with very small values.) For our model the high carrier density is not a limit but a targeted use case. The project was motivated by the problems we had in equilibrating device simulations with doped injection layers. The high carrier density in these layers makes KMC computationally expensive due to the interaction between these carriers and the state rattling where essentially all numerical effort of the device simulation is spent on charges that oscillate back and forth in the injection layer[1].

The problem is illustrated in Figure 1. The majority of the moves are used in the doped injection layers. The illustrated imbalance between the layers is due to the high carrier density fostering state rattling. This problem is relevant for KMC simulations with high carrier densities (i.e. many individual carriers to simulate). We have tried to make this point more clear in the revised manuscript.

In the application we aimed for, the resistor network is first calibrated to the doped injection layers by a short preliminary KMC simulation. After this preliminary KMC simulation is finished, the occupational probabilities are extracted and put into the resistor network. Going forward, the full stack KMC simulation can use the resistor network to model the expensive layer at little to no extra computational cost. This will focus the performed KMC steps onto the other layers (by excluding expensive effects like the state rattling in the doped injection layer from the list of available KMC steps - leaving in its place only effective currents in and out of the injection layer).

While this coupling of the resistor network back into a KMC simulation was not yet done, high carrier density systems are not a limit to our model but the use case scenario, where KMC becomes slow and expensive.

2) Motivation and role of the KMC setup: The proposed approach is motivated by the desire to reduce the computational cost of KMC simulations. However, the RN model itself is initialized using a short KMC run. This raises the question of whether the method merely accelerates the evaluation of observables that are already encoded early in the KMC trajectories, even if full convergence takes longer. The authors should clarify what information is extracted from the short KMC simulation, why it is sufficient, and under what assumptions this transfer of information remains valid.

Response: We are grateful for this comment and the opportunity to further elaborate on the aim of our project.

Referring to the doped injection layers in Figure 1 the importance of the resistor network compared to longer simulation times becomes apparent. The behavior of the middle layer could be simulated more efficiently (and thus longer - which is important for systems with slow convergence behavior or in degradation studies) after the characteristics of the injection layers are known. Hence, a resistor network could be calibrated in a first step and could be later used instead of the KMC injection layers that are burdened by effects like the aforementioned state rattling.

Together with the input from the previous question we revised our manuscript to make the aim of our project clearer.

3) Reported speed-up and scalability: The claimed computational speed-up appears modest, approximately a factor of ten according to Fig. 5, with the KMC setup run representing the largest cost contribution. Given that KMC is readily parallelizable across many cores or machines, it is unclear how the proposed approach would enable simulations that are otherwise computationally prohibitive. A quantitative discussion of scalability on modern architectures, as well as an explanation of how the method can claim broader applicability than KMC given its dependence on KMC for initialization, would be necessary.

Response: We thank the referee for this important comment and agree that the acceleration due to not waiting for the full convergence of the KMC simulation appears modest. We want to clarify that the main advantage of the resistor network approach is not a simple acceleration during the characterization of a layer but that the resistor network can afterwards be used to calculate currents through the layer very fast. Future studies could therefore use the resistor network to replace expensive layers (as long as they only transport charges - as is the case for example in doped injection layers) and focus further KMC steps toward other layers. This is important for example in full device simulations - if these simulations include for example doped injection layer, they oftentimes are troublesome to converge due to how expensive each step is.

4) Sparsity and scope of the data: I find the dataset presented too limited to justify broad conclusions. All results shown demonstrate good agreement between the RN and KMC methods, but no examples are given where bigger deviations occur. Including cases where the agreement deteriorates while analyzing the underlying reasons for this would help define the method's range of validity. Moreover, Figs. 2-4 contain only four data points per panel. Given the reported computational costs, I estimate that each data point, including the 20 replica runs used for error estimation, should require roughly a day or less of computation time, suggesting that a denser sampling, potentially sufficient to yield smooth trends, should be feasible and would considerably strengthen the manuscript.

Response: We thank the referee for this thoughtful comment and are expanding our simulations to include more data points and hence have smoother trends.

5) Numerical details and reproducibility: Since the manuscript is primarily methodological, full reproducibility is essential. The simulation parameters, including those used in Lightforge, should be reported comprehensively. While some are illustrated in Fig. 1 and I appreciate the illustration, a complete list should be provided either in the main text or in the Supporting Information.

Response: We agree with the referee that reproducibility is very important for this study and are gladly appending our input files to the Supporting Information.

Line 58: Please correct the extra commas in numerical values.

Response: We thank the referee for the careful reading and fixed the formatting of these values in the manuscript.

Figure 1: Adding sub-panel labels (a), (b), (c) and referring to them in the text would improve readability.

Response: We thank the referee for this suggestion and introduced the labels into the figure and mentioned them in the text.

Figures 2,3,4: It would be clearer if the titles or captions explicitly indicated the disorder parameter, e.g., by writing "σ = ...".

Response: We appreciate the reviewers suggestion regarding the readability and added the information to the corresponding captions.

Applicability: A short discussion of cases where the method is expected to perform well or fail would help readers assess its practical applicability.

Response: We agree with the reviewer that the practical applicability should be highlighted in the manuscript. Hence, we altered the manuscript to make this clearer and also emphasize that not all layers in for example an OLED could be replaced by the resistor network. Emissive layers for example can not be modeled as the emissive behavior would rely on

1. explicit electrons and holes and their pairwise interaction
2. formation of excitons and treatment of exciton movement
3. decay of excitons

none of which can be modeled by the resistor network.

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Authors Answers to the Reviewers Remarks

1 Reviewer #1

The authors have made the necessary changes in the revised version of the manuscript and have responded to all comments from both reviewers. The suitability of the resistor network-based computational framework for overall OLED-type device modelling is better explained in the current version of the manuscript, while the limitations of the method have also been outlined. I recommend publication of the article with minor revisions.

1. Use of the term 'high charge carrier density' in the title as well as in the abstract seems misleading. As the authors pointed out in the response to comment 5 of Reviewer 1, as well as in the response to comment 1 of Reviewer 2, the high-carrier density is used as a 'targeted use case' which is prevalent in the doped injection layers and 'is not tied to the physical behavior of the system with a high carrier density'; the acceleration in calculation time results simply from the weeding out of state rattling during KMC simulations and frequent recalculations of Coulombic interactions. The calculated results for monolayer, bilayer, as well as the doped systems, highlight the transport phenomena across the interface between layers, rather than through the material itself (which is evident from the high output current with increasing disorder resulting from Fermi level alignment), and the usefulness of the RN method across the carrier density spectrum. Additionally, in the literature, high charge-carrier density usually corresponds to carrier density > 1020 cm-3 (e.g., J. Am. Chem. Soc. 2022, 144, 3005-3019). Therefore, it will be useful if the authors reformulate the term 'high charge carrier density' with something like ' at OLED injection layer'.

Response: We thank the reviewer for this suggestion and altered the corresponding sections in the manuscript, making it clearer for readers what kind of systems we aimed for.

2. The revised manuscript contains some minor typographic errors (e.g., line 28 – 'even though' is misspelled); I request that the authors correct them for the final submission.

Response: We are grateful for the reviewers thorough reading and corrected the spelling error in the manuscript.

2 Reviewer #2

In the revised manuscript "Using a Resistor Network to Simulate Charge Transport in Organic Semiconductors with High Charge Carrier Densities" by Petkau, Özdemir, Symalla, and Wenzel, the authors have addressed some of the previous points. While I appreciate the additional data points in Figs. 2–4, the sublabels a), b), c) in Fig. 1, and the inclusion of the input file, the manuscript edits appear minor, especially in comparison to the explanations provided in the rebuttal letter. Many points remain insufficiently clarified. My specific comments are as follows:

1) Inclusion of Rebuttal Figure: The rebuttal letter introduces a new Fig. 1, cited multiple times to respond to reviewer questions. It is unclear why this figure or a similar one, along with the associated discussion, was not incorporated into the manuscript itself.

Response: We thank the reviewer for this suggestion and agree that adding the figure strengthens the manuscript. Further, we used it to highlight current problems of KMC simulations with processes such as state rattling (present for example in doped injection layers), which we wanted to address with our study. A reader of the manuscript can now easier grasp, what kind of systems we were aiming for.

2) Previous Question 1: My earlier concern regarding the physical and numerical treatment of high-density systems remains inadequately addressed. Minimal edits in the revised manuscript correspond to this point, and the rebuttal letter only partially clarifies the issue. While the authors explain why high carrier density matters, the manuscript still lacks a clear, quantitative description of how the model approximates or incorporates the high-density limit. Statements such as "high carrier density is not a limit but a targeted use case" are understandable but informal. The connection between the resistor network and the physical treatment of carrier interactions is not explicit. The heuristic neglect of Coulomb contributions is not discussed. Furthermore, the connection to Cottaar et al. is weak; mentions of high energetic disorder recovering percolation pathways are brief and the statement "We cannot see how this would translate into a high carrier density but low hole density limit" is unconvincing. These points must be addressed, also in the revised manuscript.

Response: We appreciate the feedback and have incorporated a more thorough discussion of these points in the Methods section of the manuscript, drawing from our rebuttal explanations. We have added a new subsection to clarify the treatment of carrier interactions and high-density systems in the Resistor Network model. Further, we emphasized the connections and distinction to previous work, namely the work of Cottar et al.[1] We also extended the discussion of the Coulomb contributions neglect by drawing more explicitly conclusions from the supplementary material.

3) Previous Question 2: The revised manuscript does not properly clarify how the resistor network improves KMC simulations or why skipping calculations in the injection layer is justified. While the rebuttal letter claims this was clarified, I could not identify corresponding edits in the manuscript. The repeated use of Fig. 1 in the rebuttal letter suggests that the advantage of the resistor network is to bypass the injection layer calculation, but this should be explicitly discussed in the manuscript. Additionally, the frequent use of the word "could" in the rebuttal letter creates ambiguity: it is unclear what the authors are actually doing versus what is hypothetical.

Response: We thank the reviewer for this point and added a discussion in the introduction section of the manuscript. Together with the rebuttal figure the reader now gets introduced to the cost saving potential that comes with replacing the doped injection layer in the presented example device. We also marked more explicitly sections that are meant as an outlook.

4) Sparsity of Data and Applicability Limits: One of my main concerns from the previous review, regarding the sparsity of data, was not addressed and was not included in the rebuttal letter. While additional data points are included which is appreciated, the manuscript still explores only a limited portion of the parameter space. There should at least be a discussion if not a demonstration of the limits or failure modes of the method. Such information is critical for the community to assess the reliability of the method and to avoid erroneous results and need at least be commented on in the revised manuscript.

Response: We appreciate the suggestion and added a new subsection to the discussion section of the manuscript, regarding the restrictions of the RN model. Here we point out that the RN will fail for systems with strong time-dependent effects or systems where the performance is dependent on pairwise interaction of particles.

References

1. Cottaar, J., Koster, L. J. A., Coehoorn, R. & Bobbert, P. A. Scaling Theory for Percolative Charge Transport in Disordered Molecular Semiconductors. *Phys. Rev. Lett.* **107**, 136601, DOI: 10.1103/PhysRevLett.107.136601 (2011).

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Reviewer #1

No further remarks from Reviewer #1 are passed on by the editor.

Response: We thank the Reviewer for his time and efforts. We are grateful for the Reviewers thorough reading and the questions, which led us to clarify the context and scope of our work, improving the accessibility.

Reviewer #2

I have reviewed the revised manuscript and the authors' responses to my previous comments. The authors have addressed my concerns satisfactorily, in particular by incorporating additional discussion in the Methods and Discussion sections, clarifying the intended regime of applicability, and explicitly outlining the limitations of the resistor network approach. The inclusion of additional data and the new discussion of failure modes improve the manuscript and make its scope clearer for the reader.

Overall, I am satisfied with the revisions and believe the manuscript is now suitable for publication. I therefore recommend it for publication.

Response: We thank the Reviewer for his time and efforts. We are grateful for pointing out various vague points in our manuscript, which helped to elaborate on weak points and considerably strengthen our argument.