

This contribution by Jelavic et al. concerns the evaluation of the adsorption capacity of soot and charcoal surfaces with respect to DNA as a function of physicochemical medium composition, including change in pH, solution ionic strength, presence/absence of divalent cations, eDNA length, under both equilibrium and non-equilibrium conditions. In addition to classical batch adsorption experiments, authors resort to a suite of spectroscopic techniques (XPS, XRD, Raman, BET) to characterize the surface of their adsorbing materials and they analyse the obtained adsorption data with help of classical models e.g. Langmuir, Sips, Freundlich isotherms, etc.

Overall, I think this paper deserves publication pending, however, major revisions listed below.

1. The reader misses a general discussion on the very mechanisms driving the adsorption of DNA on their tested soot and charcoal surfaces, with full integration of a consistent and complete cross-examination of the data derived from the various techniques and models they adopt. In the current form, the manuscript lacks this overall discussion, and the latter would help to support the statement by the authors "this study provides a fundamental basis for dsDNA-CM interactions". As far as I can judge, most of the elements advanced by the authors to explain the adsorption patterns they conclude on (i.e. relative importance of electrostatics and hydrophobic interactions, heterogeneities of adsorbing surfaces, etc) are already mentioned in the introduction section (p.2) and are known from classical physicochemical literature on e.g. protein adsorption (see e.g. the work by Norde W. et al, etc). In turn, the authors should better clarify the new rationale they provide as compared to that from existing literature on a wider perspective (protein/macromolecules adsorption on heterogeneous surfaces).

2. The reader is missing a quantitative assessment of the hydrophobicity level of the soot and charcoal surfaces (quite central in the manuscript), e.g. via macroscopic contact angle measurements or (better) via Atomic Force Microscopy (see e.g. Francius et al. ACS Applied Bio Materials 2021, 4, 2614-2627 or Nanoscale 2021, 13, 1257-1272). Related to this latter point, the reader is generally missing a physicochemical evaluation of the sorbing surface properties at the proper molecular scale as e.g. achieved with help of Atomic Force Microscopy operating in force spectroscopy mode with use of functionalized probes (e.g. decorated by -CH₃ or charged functional groups) or even a direct probing of the surface-DNA interaction upon functionalization of the AFM tip with tested DNA macromolecules (see e.g. Beaussart et al. Nanoscale 2018, 10, 3181 for the proof of principle). Such experiments would have the advantage to probe directly, at the molecular level, the operational interactions and the way these are impacted by surface heterogeneity (elaboration of spatially-resolved interaction maps) and possibly by changes in DNA conformation during the very interaction processes. These elements are missing from the literature quoted by the authors (see work on protein adsorption in literature).

3. Authors should mention in the very body of the main text the Table summarizing the models they used to analyse their equilibrium and kinetic adsorption data. I doubt that the DNA-sorbing surface interactions are generally included in the classical models the authors refer to. Such model extension (which exist in literature) would help the reader to compare the pristine adsorption capacities of soot and charcoal and the relative contributions of the interactions (i.e. deconvolution between non-specific and specific (chemical) components of the adsorption isotherms and adsorption kinetics). For the sake of illustration, non-DLVO behavior is expected for surface-macromolecule interactions in the presence of multivalent ions and/or in concentrated electrolyte as the result of ion-ion correlations. Do this apply to the systems of interest in the manuscript ?

4. The authors should discuss the relevance and applicability of their findings within a more general environmental context. More specifically, the authors focus here on well-defined DNA macromolecules (from salmon sperm, motivations of this choice ?) whereas eDNA involves macromolecules differing with respect to conformation, chemical composition, origin, degradation, etc. This opens the debate on how the very heterogeneity of the macromolecules in terms of their defining physicochemical properties affects the adsorption capacities of soot and charcoal (themselves defined by physical and chemical heterogeneities). On the basis of their own data and of known sorption-mechanisms reported in literature (related to competitive adsorption features, etc), the authors could discuss, at least qualitatively, these various points.

5. The authors refer to inner and outer particle surfaces, to shift of IEP and PZC with changing the concentration of 2:1 electrolyte. Authors should rather refer to inner and outer Helmholtz planes (accepted nomenclature in colloidal physical chemistry) and comment on the aforementioned shifts in relation to positioning of divalent cations at iHp or oHp and their extent of specific adsorption. Authors are referred to H. Lyklema, Fundamentals of Interface and Colloid Science, Vol. II, for that purpose. In addition, authors mention their measurement of zeta-potential. As a matter of fact, they do not explicitly state that they resort to (I think) DC electrophoresis measurements (measuring electrophoresis conditions should be specified, e.g. value of applied field ? etc), and they measure an electrophoretic mobility (not a zeta potential) that is subsequently converted into a "zeta-potential value". Authors should specify the nature of the equation used for the conversion (Smoluchowski, Debye-Onsager, etc ?) from mobility to zeta-potential and the extent of applicability of the adopted equation with regards to the very size and charge of the analyzed particles. It would have been further valuable to actually measure the dependence of the electrophoretic mobility of soot and charcoal on electrolyte concentration prior to AND after DNA adsorption. Upon application of soft particle and soft surface electrokinetic concepts (see e.g. Maurya et al. Journal of Colloid and Interface Science 2020, 558, 280 and Duval et al. Current Opinion in Colloid and Interface Science 2010, 15, 184-195), the authors could have then possibly probed the way surface electrostatics comes into play (or not) in controlling macromolecule adsorption process.