Dear Editors,

Hereby we would like to resubmit our article (Manuscript No. ES10751):

The Error Estimate of Short-Range Force Calculation in the Inhomogeneous Molecular Systems ,

by H. Wang, C. Schüette and P. Zhang to:

Physical Review E, as a regular article.

We would like to thank the referees for carefully reading our manuscript and the comments helping us to finally provide a clearer and more valuable article. Some changes to the manuscript have been done, as suggested by the referees, and are explained in the following.

Referee 2

I also did not understand how the adaptive cutoff method results were plotted as a function of cutoff distance in Figs 5 & 6. A variety of cutoffs are being used, yet it is plotted against a single cutoff distance?

Throughout the paper, when we mention that "the cut-off of the adaptive cut-off method is r_c^*", we mean that its control error is the maximum error of a uniform cut-off simulation using that cut-off r_c^*. Under this setting, the maximum cut-off radius of the adaptive cut-off method is the same as, or sometimes marginally larger than r_c^* (no more than r_c^{step}). We have made this point clear in the captions of Figures 5 and 6.

I also suggest you make brief mention of the following issues, and explain how your methods deal with them:

(1) Does the ARC method (adaptive cut-off) make sense for long-range Coulombic calculations where there is a short-range part with a cutoff coupled to a long-range Kspace calculation (e.g. particle-mesh Ewald). This is a commonly used approach for charged molecular systems (e.g. solvated biomolecules). In that case, I believe the Kspace formulation depends on their being a single cutoff on the short-range portion.

We have submitted a paper, titled "The Numerical Accuracy of Computing Electrostatic Interaction in the Inhomogeneous and Correlated Molecular Systems: for the Ewald Summation, SPME and Staggered Mesh Ewald Methods", dealing with the Ewald type long-range algorithms. In that paper, we concluded that the locally neutral systems do not have the error anomaly at the interfacial regions. Since most practical charged systems are locally neutral, we are safe and do not need ARC (adaptive cut-off) or LFC (long-range force correction) for the electrostatic calculation.

(2) Can the spatial error estimation technique and 2 correction methods be used for either periodic or non-periodic systems?

As far as we concerned, the error estimate technique and proposed corrections can only be used to periodic systems. We have added the following sentence to stress this point (line 5, page 26): **It should be noticed that the**

porposed error estimate and correction methods can only be used to periodic systems.

(3) You say that the additional cost of the methods is small even though they involve FFTs that scale as $N \log N$, compared to the $O(N)$ scaling of short-range MD. Your timing results bear this out, however these examples were all fairly small systems. If large simulations are performed (e.g. multi-million atoms), is the extra cost still small?

This is a very important comment, because the cost of FFTs $O(N \log N)$ will overcome the cost of short-range interaction $O(N)$ at some point when the system is very big. Eventually the computationaly cost will be overwhelmed by the FFTs as the size of the system grows to infinity. This point should be noticed. Fortunately, for the systems also calculate long-range interactions, the short-range error estimate scales the same as the Ewald type long-range algorithms. We have added the following remarks to the paper: (line 5 from the bottom, page 26): **In the systems tested by the present paper, the computational load of the error estimate is small compared to that of the short-range interaction. However, it should be noticed that the cost of the error estimate scales as $\mathcal{O}(N \log N)$, which may become intractable when the $\mathcal{O}(N)$ scaling short-range system is extremely large, and eventually overwhelms the total computational cost as the system size grows to infinity. If the system also needs long-range electrostatic calculation that is usually treated by the $\mathcal{O}(N \log N)$ scaling Ewald type algorithms [22-24], the extra load of the error estimate is likely to be acceptable.**