**Response to reviewers**

Reviewer #1:

The paper by Claridge, Kupce and co-workers builds on previous works from these authors on appending multiple pulse sequences to afford time savings by smart manipulation of the spin polarizations of different spins in molecules. In the current manuscript the focus lies on the concatenation of four experiments: 1H-15N se-hsqc, 1H-13C se-hsqc, 1H-1H COSY and 1H-1H TOCSY. As this approach generates so-called super sequences, there is a myriad of potential experiments that can be included, and in some cases also the order in which these appear. In the words of the authors "There exist many ways in which the new modules discussed above can be included in practical experiments ...". The work is ingenious, although the current approach is a refinement of more innovative contributions from this group and many others. The technical level is high, and the experiments have potential to be employed for applications in various areas of chemistry.

We thank the reviewer for their positive comments on the work.

I have one important suggestion, which I feel would require attention: The intertwining of multiple experiments will produce a time-saving, but has a significant potential to introduce artifacts in (a subset of) the experiments. In particular, when recording only very few scans per t1 increment, barring use of extensive phase cycling.

All experiments in the text were performed with only 2 scans per increment; thus very little phase cycling is possible (only enough for suppression of axial peaks). This is noted in the *Experimental* section of the manuscript.

Despite the substantial SI, this is still very difficult to evaluate. Ideally, the authors would make their data accessible in a repository (like Zenodo, github or other) to allow others to access both the new experimental pulse sequences and parameter sets, as well as gauge representative reference data from the current paper.

We agree with the reviewer that having access to raw data would make the spectral quality easier to evaluate. Following their suggestion, we have uploaded all the NMR data used in this paper, along with pulse programmes and associated processing scripts, to Zenodo (DOI: 10.5281/zenodo.4937946). The pulse programmes have also been uploaded to the Bruker User Library. The *Experimental* section of the manuscript has been updated to include these.

A few minor comments:

"We further demonstrate that these time savings can translate to increased detection sensitivity per unit time" is an open door as old as signal averaging

Resolution of the graphical abstract in my PDF was poor, but good in the PDF that was extracted from LaTeX - might be journal conversion problem?