We thank Andrea Chlebikova for the very thorough work and the positive and helpful feedback. We adjusted our manuscript accordingly. In particular we corrected all typos, rephrased unclear or misleading sentences, corrected wrong cross-references and units as pointed out in the technical corrections of section 3. In the following, general and specific comments are addressed.

p6, line 4/5: mistakenly referring to $R_2$ value as correlation coefficient
ESM, p15, p18, p20, p21, p22, p23: throughout (including the labels of Tables S3, S5, S6, S7), $R_2$ is mistakenly referred to as the correlation coefficient
We have corrected the mis-used term ‘correlation coefficient’ to ‘coefficient of determination’.

p21, line 7: change “has also” to “also has” and add comma before “decreasing”
Inconsistent use of hyphenation of gas-phase/aqueous-phase throughout this section – standardise
Concerning the seemingly inconsistent use of ‘gas-phase’ and ‘gas phase’, we have used the hyphenated form for the use as adjective as in ‘gas-phase compounds’ and the unhyphenated form to address the gas phase as a noun. Therefore, we believe that the hyphenation is correct in this context. Mechanism self-construction was consistently renamed to mechanism auto-generation.

While I have found the written text (despite the reservations above) for the most part easy to follow, I believe some of the figures used are quite difficult to read and interpret, and would benefit from reploting. This applies in particular to Figure 1, where I am struggling to see what the different boxplots represent. Figures 3, 4, 5, 7, S1, S3, S4, S5, S6 and S7 would benefit from using different symbols as markers rather than just different colours, as some of the colours are quite difficult to distinguish. Figure 8 could be made clearer by using more significantly different markers (rather than crosses with different orientations).
We have replotted Figures 1, 3-5, 7, S1-S7 with a revised colour and marker scheme. The marker scheme compromises between the distinctness of shapes and shapes that don’t cover other data points, when many data are close together in a graph. Moreover, we have expanded on the axis labels in Fig. 1 and introduced an additional legend. Moreover, alternate grey and white shading was used to mark boxplots of corresponding compound classes.
While reploting these figures, some mistakes were correct in the old graphs. This includes some mislabelled data as well as an update of some data from an earlier version of the kinetics database. Correlations in Table S3 and S7 were updated in this context and the tables slightly reformatted.

I would try to move more of the detail into the ESM, as the main text is currently very long. For example, at present, sections discussed at length in the ESM are still discussed in a lot of detail in the main text (e.g page 2 and page 7), when referencing the ESM should be enough. The text also feels unnecessarily long when sections are introduced carefully, and then go on to repeat the context mentioned previously.
To shorten the paper, the introductory part of section 2 was removed and partly moved to subsections 2.1 and 2.2. The analysis of the simple correlations in section 2.2 was shortened. Furthermore, we have introduced a list of abbreviations in a new appendix A, which is split into different topics/sections. In this context, we have removed Table 2 and moved the contents to appendix A.2.

ESM, p25, Table S9, p27, Table S10, p28 Tables S11 and S12: please check the units; I can understand those in Table S8, even though it would perhaps make more sense
to say C itself is dimensionless (etc.) unless the division by its units is also added into the equation; for these other tables, the volume part of the units does not seem to add up correctly.

In Tables S8 to S12, correlations are used to derive a second order aqueous-phase rate constant in $M^{-1}$ $s^{-1}$ from the BDE in $kJ$ $mol^{-1}$. Therefore, constants in the regression have the same unit as $k_{2nd}$ and are in in $M^{-1}$ $s^{-1}$. For terms with the BDE, the units are $M^{-1}$ $s^{-1}$ $kJ^{-1}$ $mol$ to yield the unit of $k_{2nd}$. With $M = mol$ $l^{-1}$, the unit can be simplified to $l$ $mol^{-1}$ $s^{-1}$ $kJ^{-1}$ $mol = l$ $kJ^{-1}$ $s^{-1}$. For terms with a quadratic BDE, $kJ$ $mol^{-1}$ has to be squared, which yields $mol$ $l$ $kJ^{-2}$ $s^{-1}$ after simplification. To unify all notations, units in Table S8 have been changed to the simplified form. In Tables S10 and S12, $l^2$ was correct to $l$ in the units of the quadratic term.

The figures showing simulation data can only be understood in conjunction with careful reading of the main text, and would benefit from more detailed descriptions in their labels.

We have expanded on the conditions in all figures of the sensitivity runs and explained the shading of cloud and night-time periods in the plots in the figure captions.

The new “advanced” Evans-Polanyi correlation strikes me as unusual, in that it can no doubt usefully scale with molecular size – but it would intuitively seem to me that we are losing most of the useful information that the individual bond enthalpies provide by lumping them into one sum. There ought to be a “better” way of accounting for more than the weakest bond than using the sum of BDEs, and perhaps this will be something that can be further improved in the future.

We thank Andrea Chlebikova for the useful feedback provided. Clearly, there is potential for further improvement of chemical mechanism development on both the experimental and theoretical side. To illustrate this fact, we added the below sentences in section 4.1 about the CAPRAM/GECKO-A protocol on page 15, ll. 28–30:

“There is a need for further development of a prediction method for kinetic and mechanistic data of nitrate radical reactions with organic compounds. However, for more advanced predication methods such as SARs, a more comprehensive experimental database is needed."

Moreover, we have re-phrased the following sentence

“A certain disadvantage of this method is that information about branching ratios and reaction products is lost."

to “... cannot be provided.” The previous description implied that the original Evans-Polanyi method was able to provide branching ratios, however, it can only suggest the major reaction products. This information is still available from the individual BDEs, but a quantitative analysis of the branching ratios is not straight-forward.