Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Multi-molar CO_2 capture beyond the direct Lewis acid-base interaction mechanism

Chenchen Li,[a] Dongmei Lu,*[b] and Chao Wu*[a]

[a] Frontier Institute of Science and Technology, Xi'an Jiaotong University, Xi'an, Shaanxi 710054, China.

[b] Department of Chemistry, School of Science, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China.

*corresponding. lvdongmei@mail.xjtu.edu.cn, chaowu@mail.xjtu.edu.cn

Supporting Information

Contents

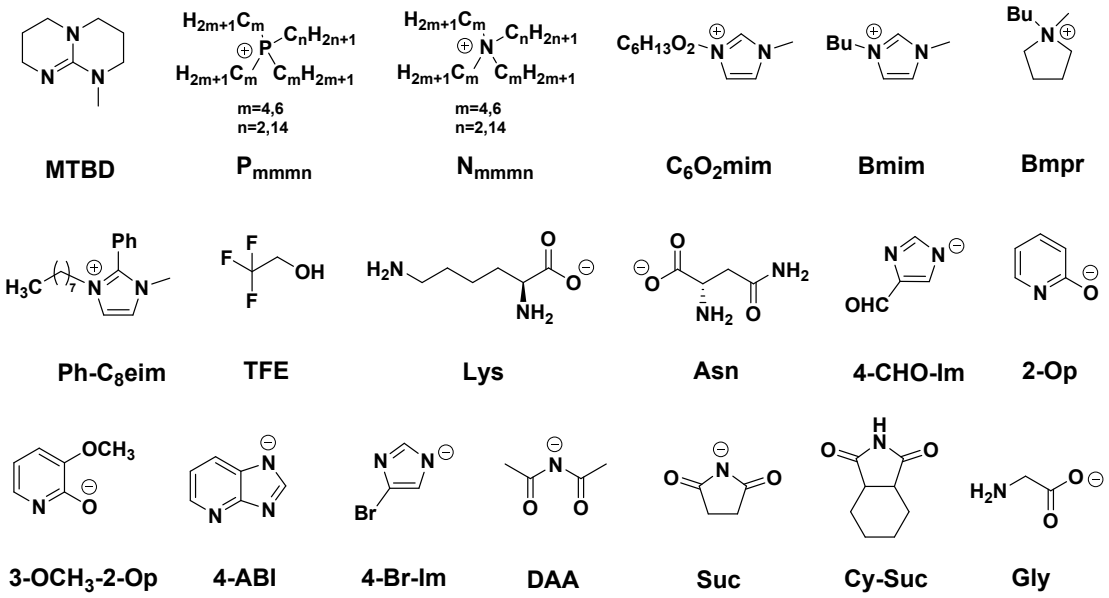


Figure S1. Structures of recently reported ionic liquids in Table 1 of manuscript.

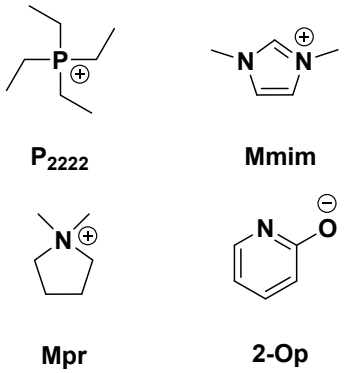


Figure S2. Cations and anions studied in this work.

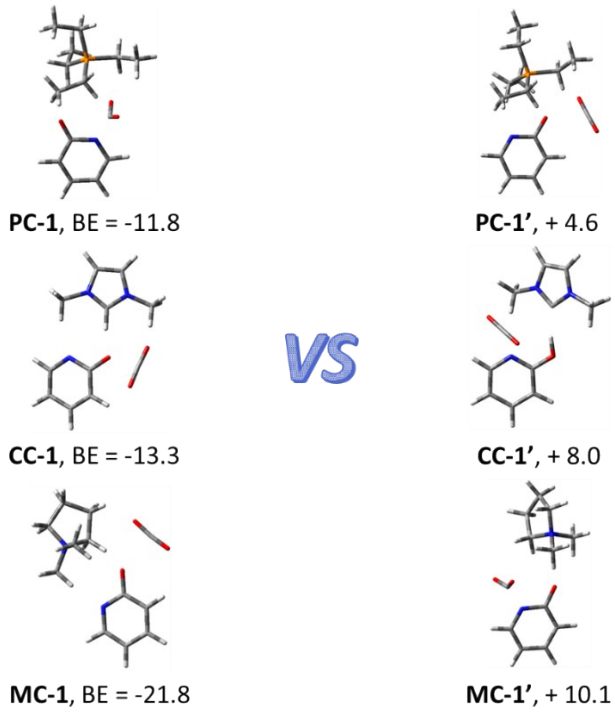


Figure S3. Stable structures of IL-CO_2 complexes identified by the configuration search program. The name and the binding energy or the relative binding energy with respect to the most stable one (in kJ/mol) are given below each configuration. Red is oxygen, blue is nitrogen, orange is phosphorous, gray is carbon and white is hydrogen.

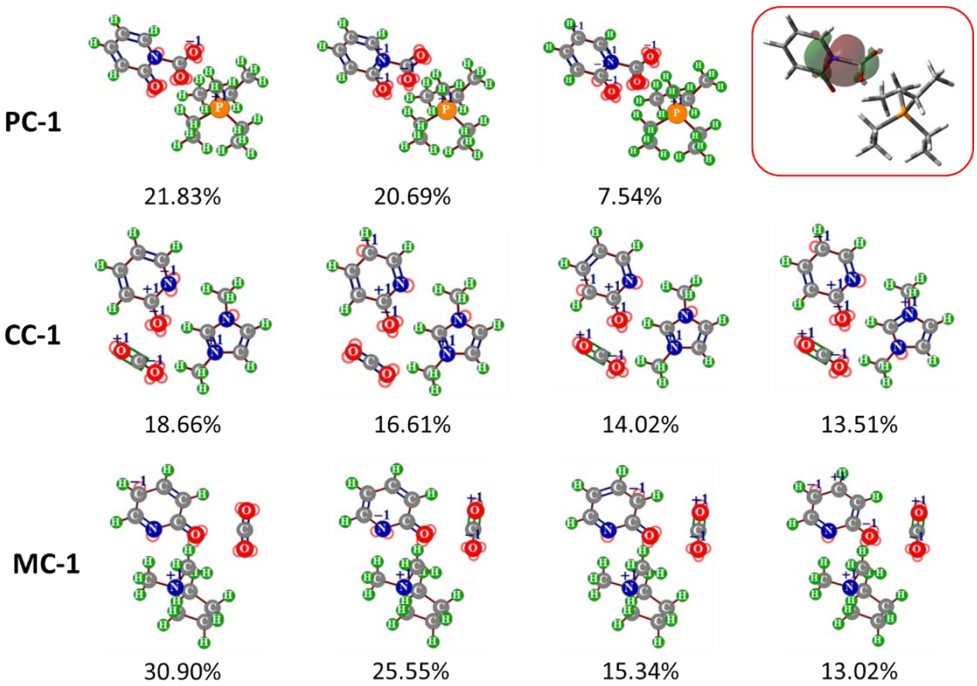


Figure S4. NBO and NRT analyses of IL-CO_2 complexes. Leading Lewis resonance structures and their abundance of the corresponding complexes are plotted. Each half circle represents a pair of electrons and formal charges are placed above the corresponding atoms. The NBO decomposition of the N-C bond of [P_{2222}][2-Op] is highlighted in red box.

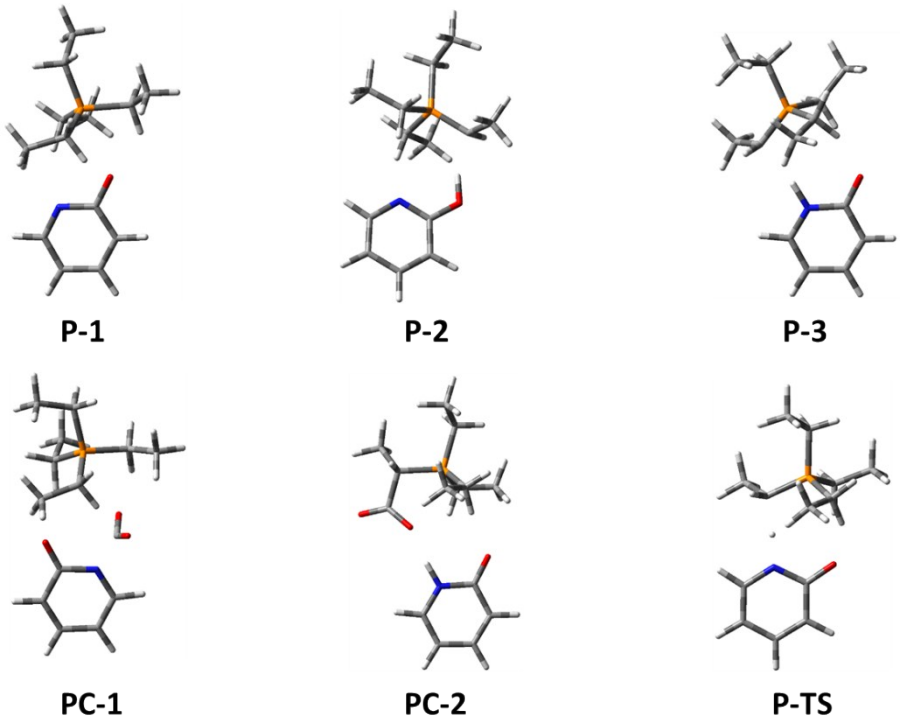


Figure S5. Configurations of the CO_2 absorptions over $[P_{2222}][2\text{-Op}]$.

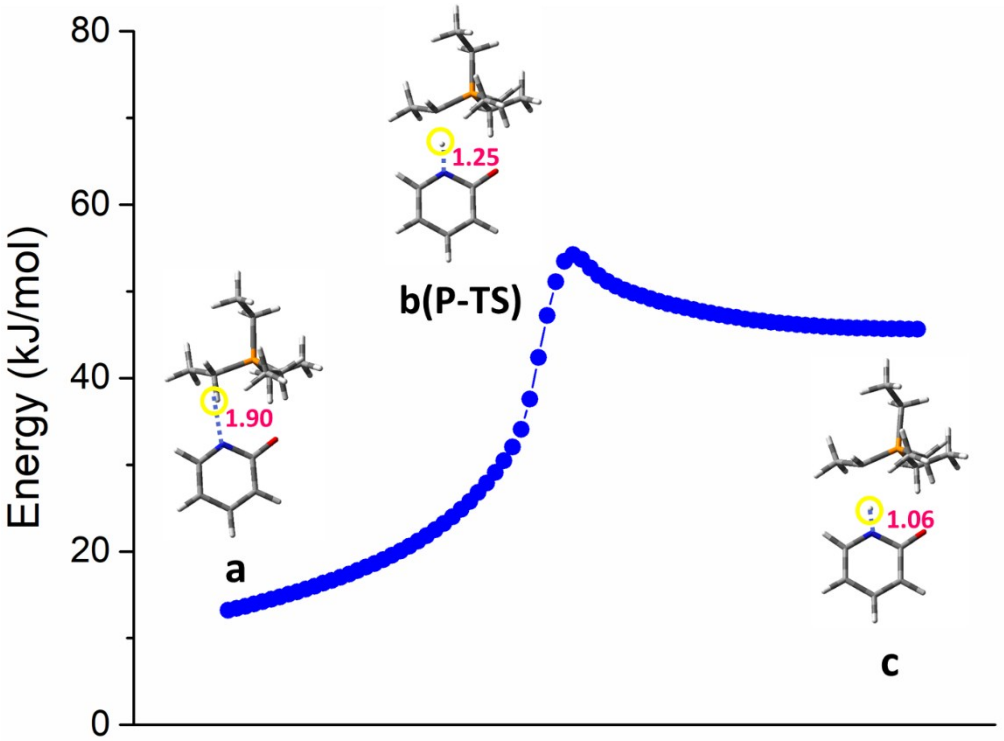


Figure S6. The interconversion energetics between ion pair and ylide intermediate of $[P_{2222}][2\text{-Op}]$ by the IRC method. Key structures are labeled and plotted. The electronic energy of ion pair $[P_{2222}][2\text{-Op}]$ is chosen as reference point. The transferring proton is highlighted.

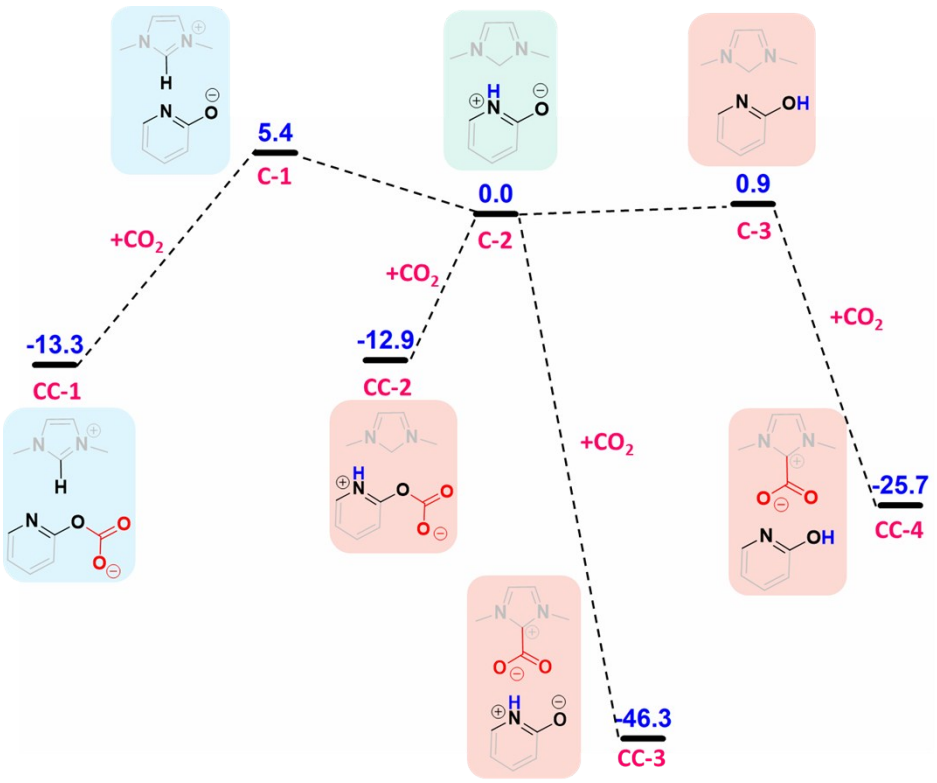


Figure S7. Comparison of conventional anion-mediated (in blue box) and proton transfer related (in orange boxes) reactions of CO_2 with [Mmim][2-Op]. The name and relative energies are given (in kJ/mol).

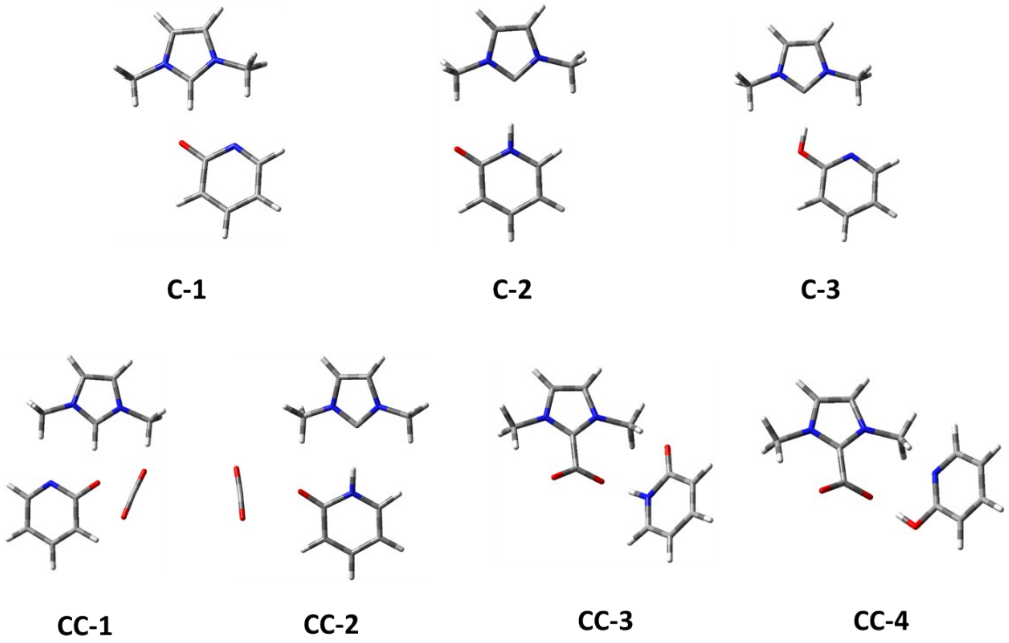


Figure S8. Configurations of the CO_2 absorptions over [Mmim][2-Op].

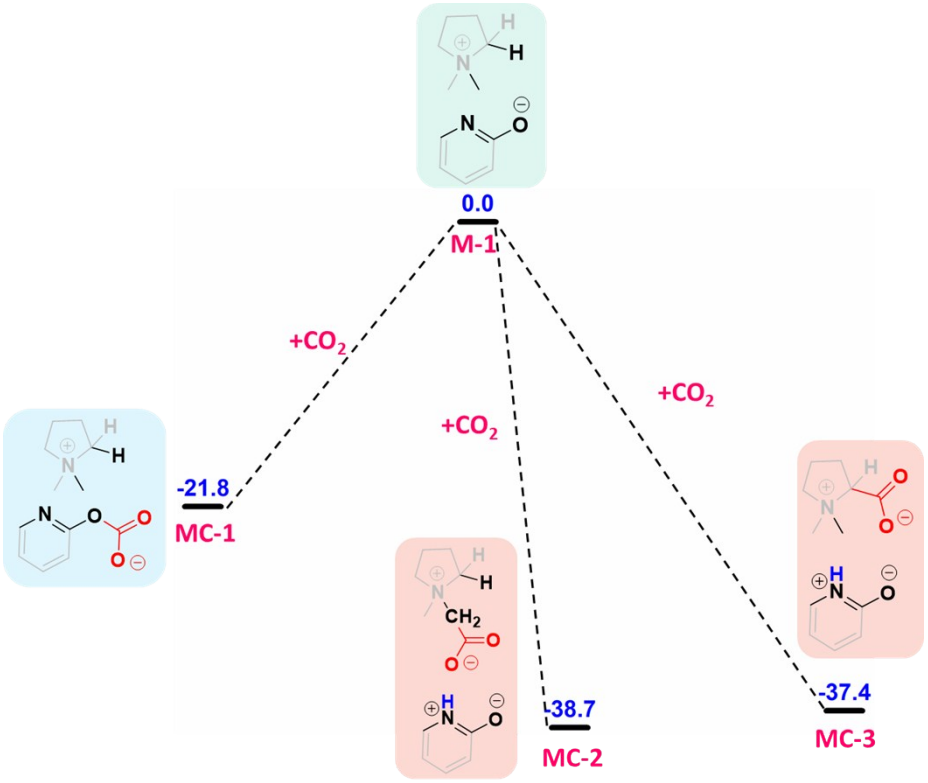


Figure S9. Comparison of conventional anion-mediated (in blue box) and proton transfer related (in orange boxes) reactions of CO_2 with [Mpr][2-Op]. The name and relative energies are given (in kJ/mol).

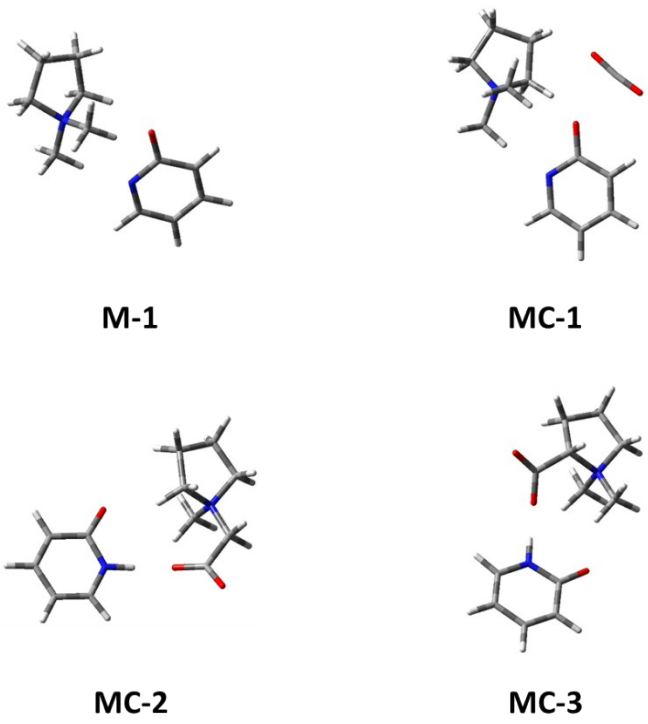


Figure S10. Configurations of the CO_2 absorptions over [Mpr][2-Op].

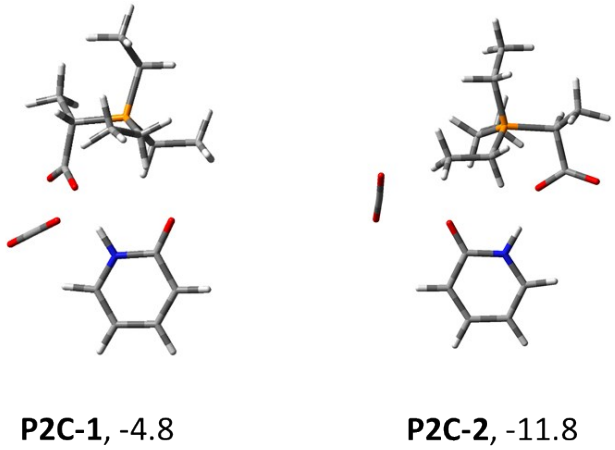


Figure S11. Configurations of $[P_{2222}][2\text{-Op}]\text{-}2CO_2$ complexes through two ways of post proton transfer routes. The name and sequential binding energies are given (in kJ/mol).

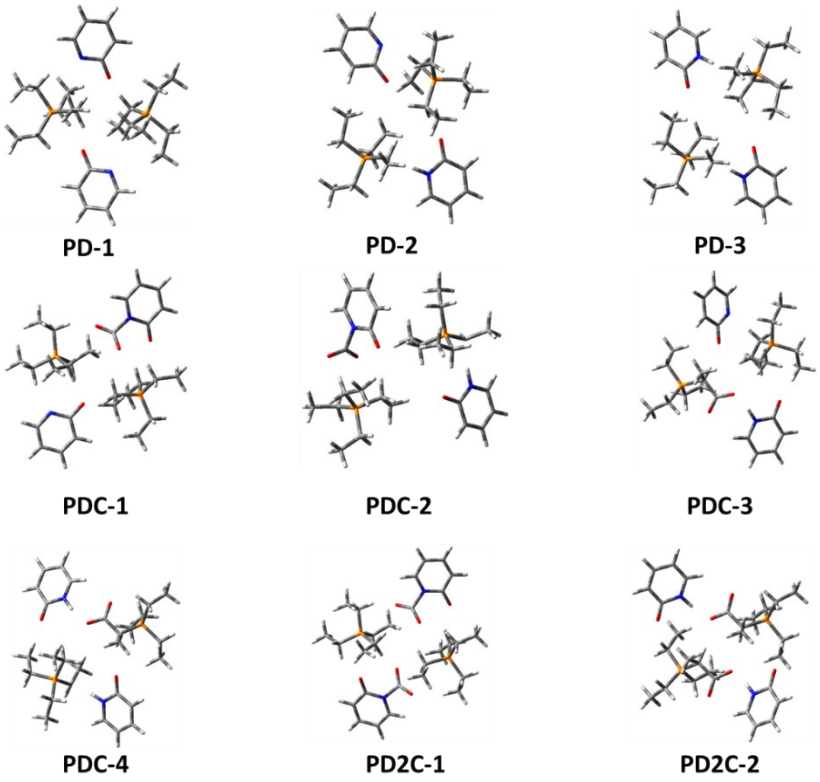


Figure S12. Configurations of the CO_2 absorptions over $[P_{2222}][2\text{-Op}]$ dimer.

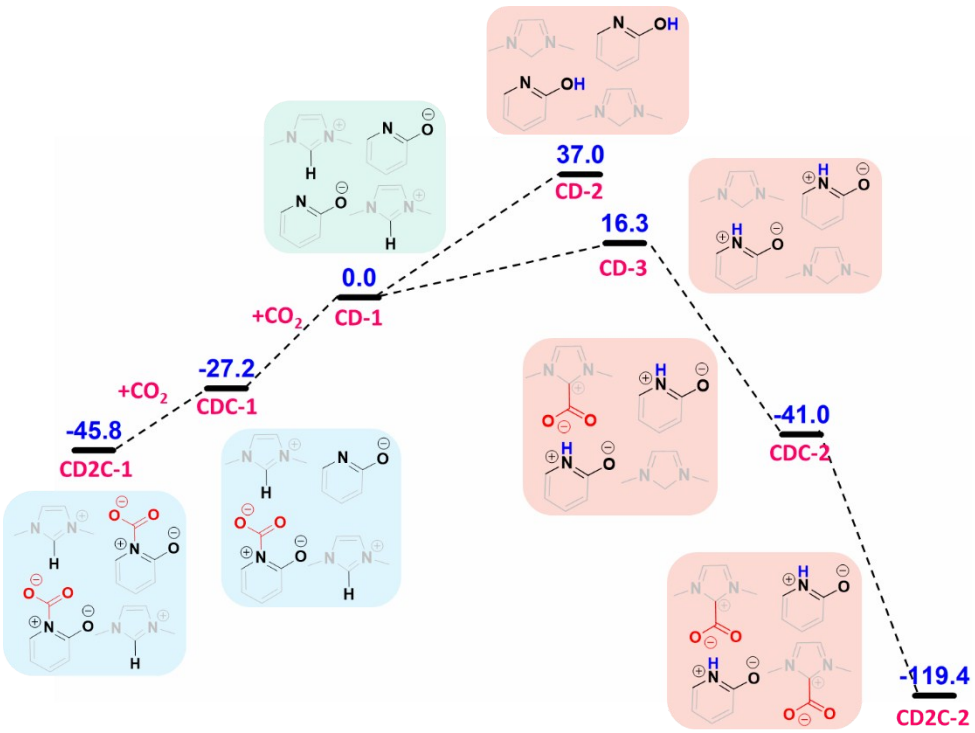


Figure S13. Comparison of conventional anion-mediated (in blue box) and proton transfer related (in orange boxes) reactions of CO_2 with [Mmim][2-Op] dimer. The name and relative energies are given (in kJ/mol).

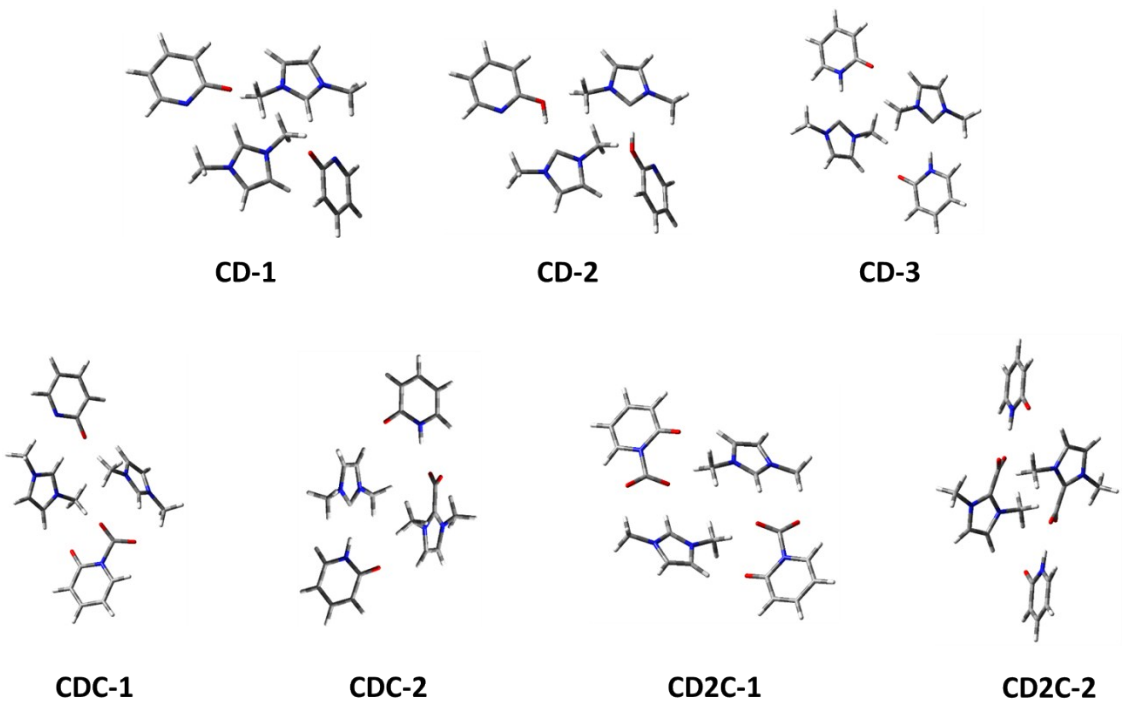
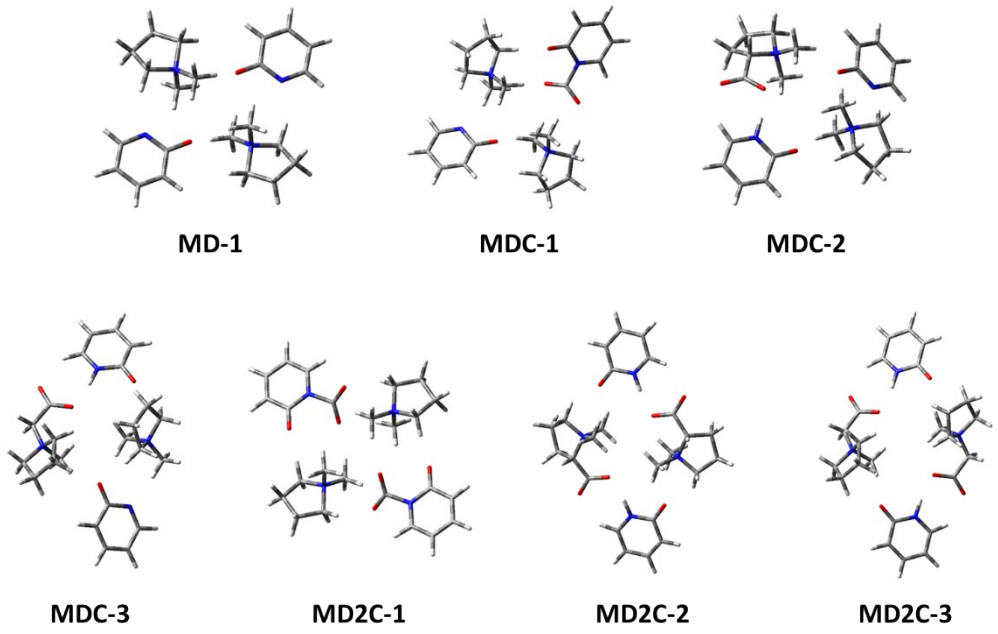


Figure S14. Configurations of the CO_2 absorptions over [Mmim][2-Op] dimer.



FigureS15. Configurations of the CO_2 absorptions over [Mpr][2-Op] dimer.

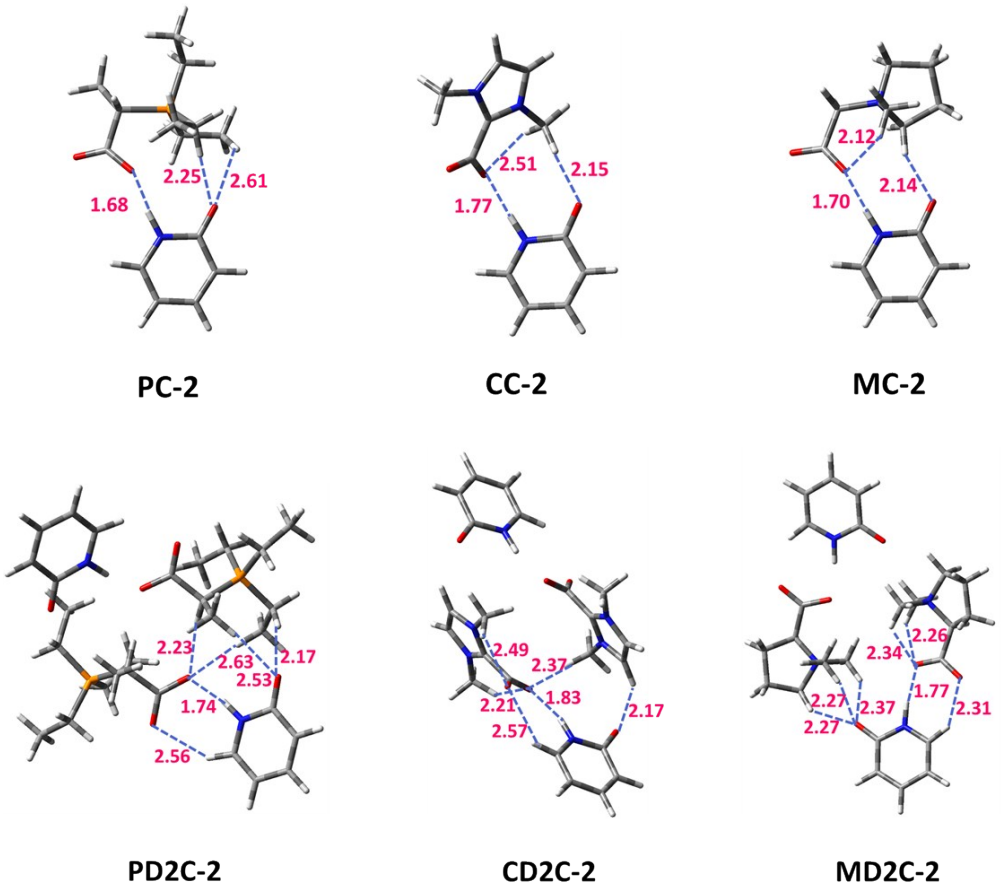


Figure S16. Comparison of most stable ylide-mediated configurations using the monomer and dimer models. Key intermolecular hydrogen bonds are labelled.