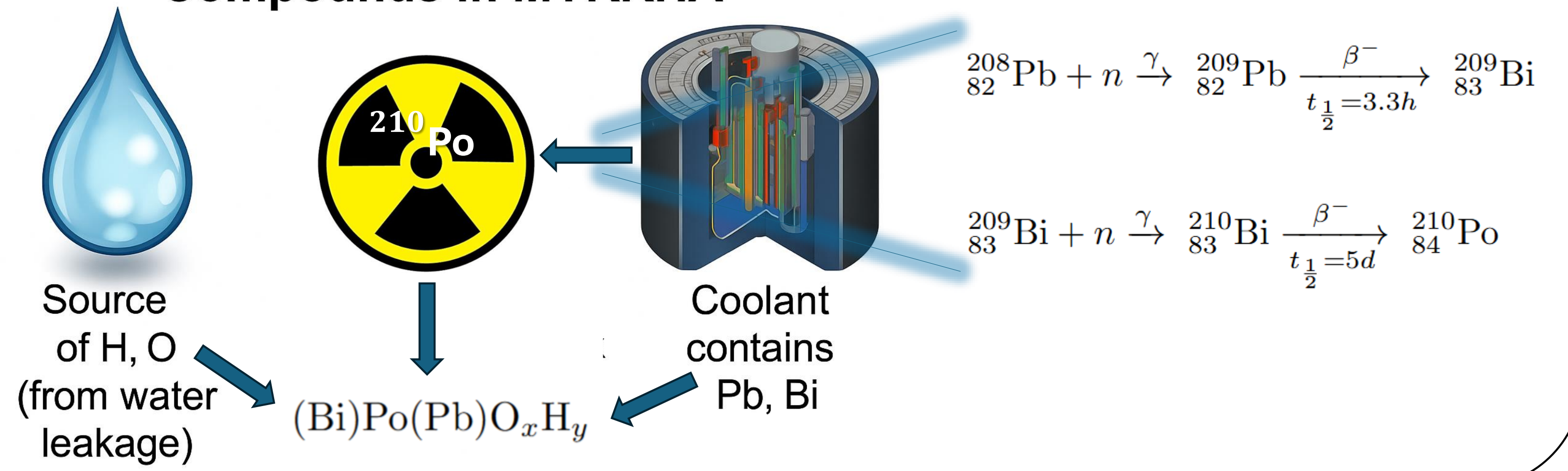


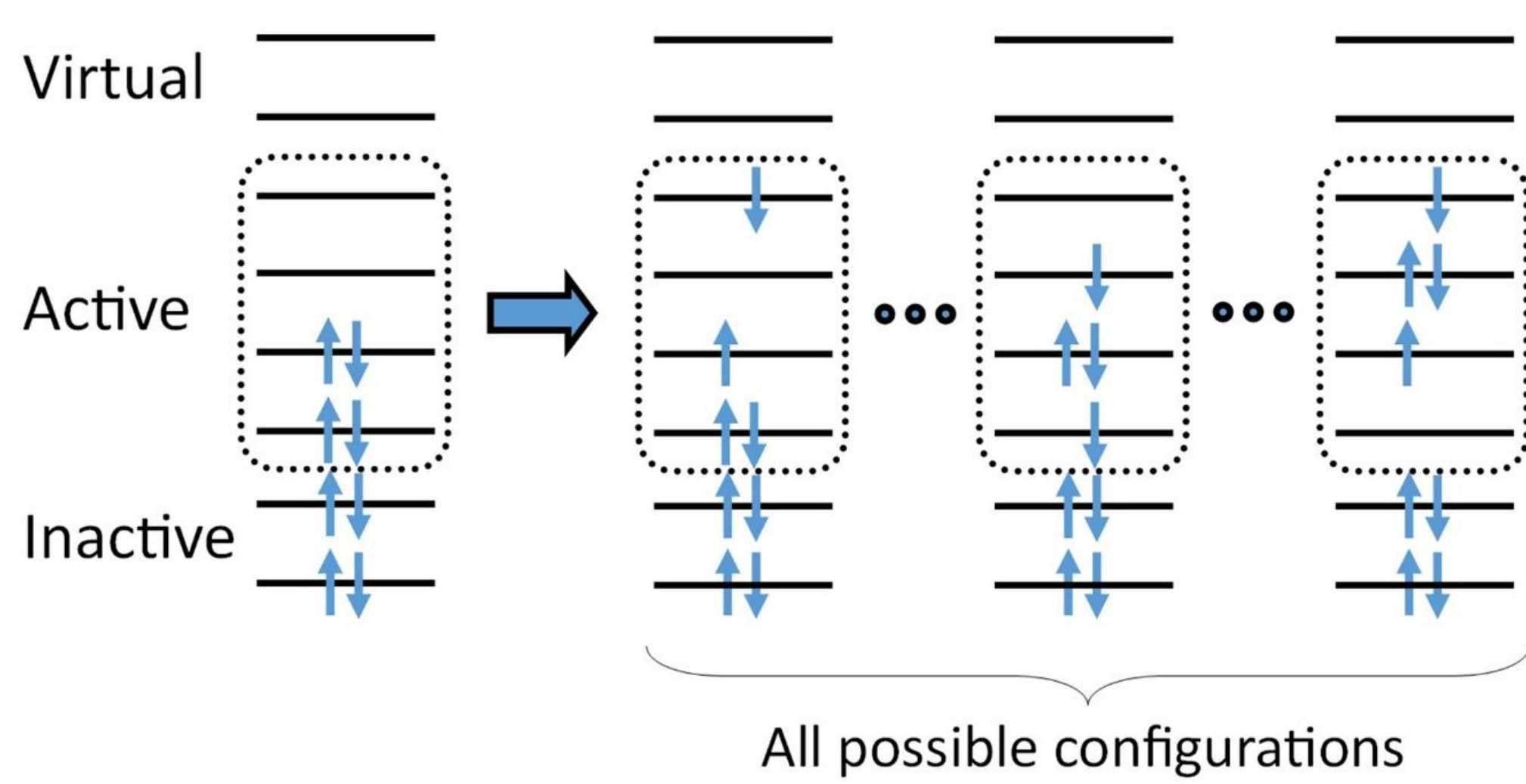
background and motivation

MYRRHA, a next-generation nuclear fission reactor, will produce, as an inevitable by-product, some amount of ^{210}Po . It stems from nuclear reactions that involve the Pb and Bi nuclei that are part of the liquid metal coolant. In the presence of the coolant and perhaps some moist, Po may form volatile molecules of the type $(\text{Bi})\text{Po}(\text{Pb})\text{O}_x\text{H}_y$. As ^{210}Po is radiotoxic, developing chemistry-specific filters to capture these molecules is essential. Yet, the (experimental) identification of relevant molecules remains limited.

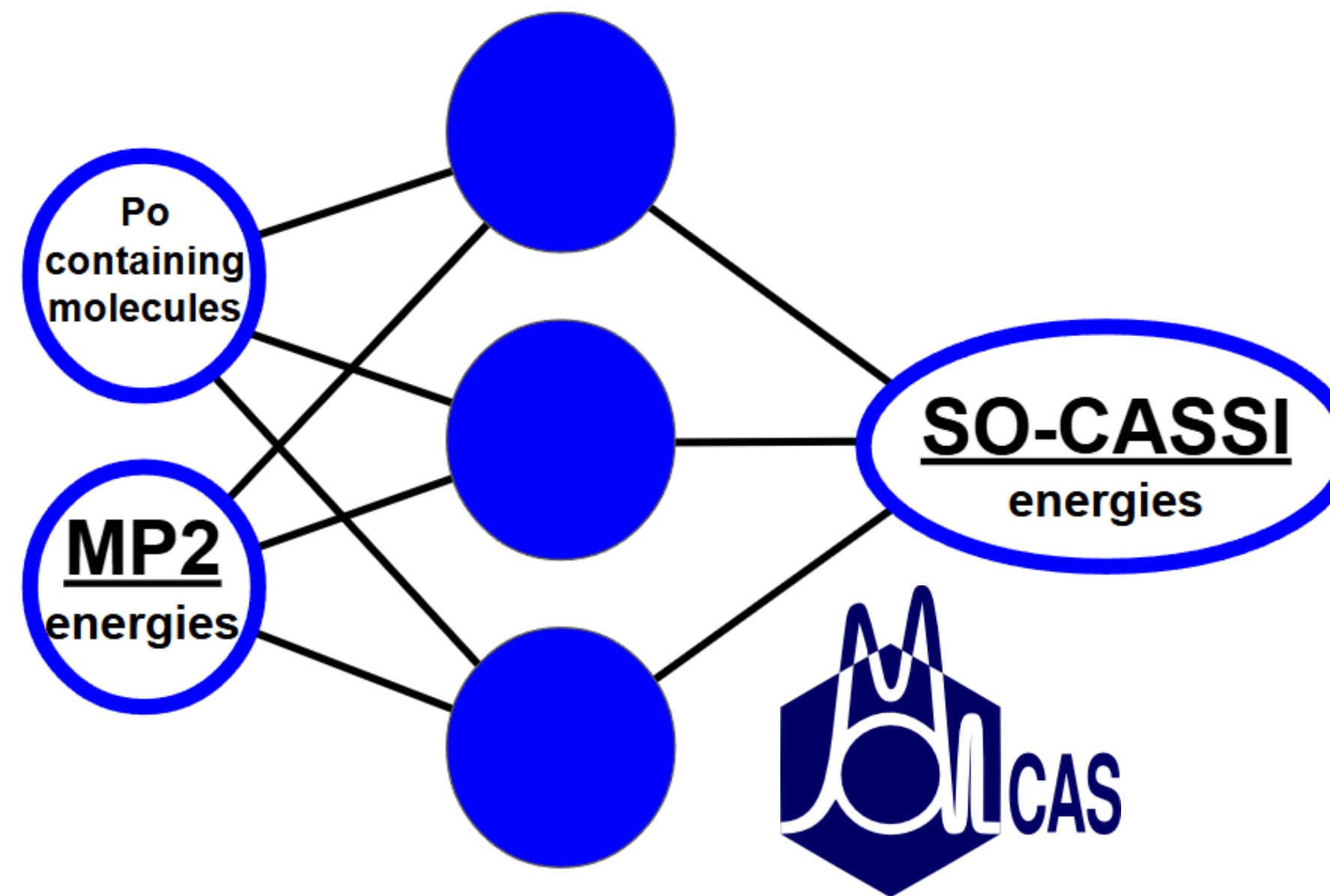
Formation of Volatile Polonium Compounds in MYRRHA



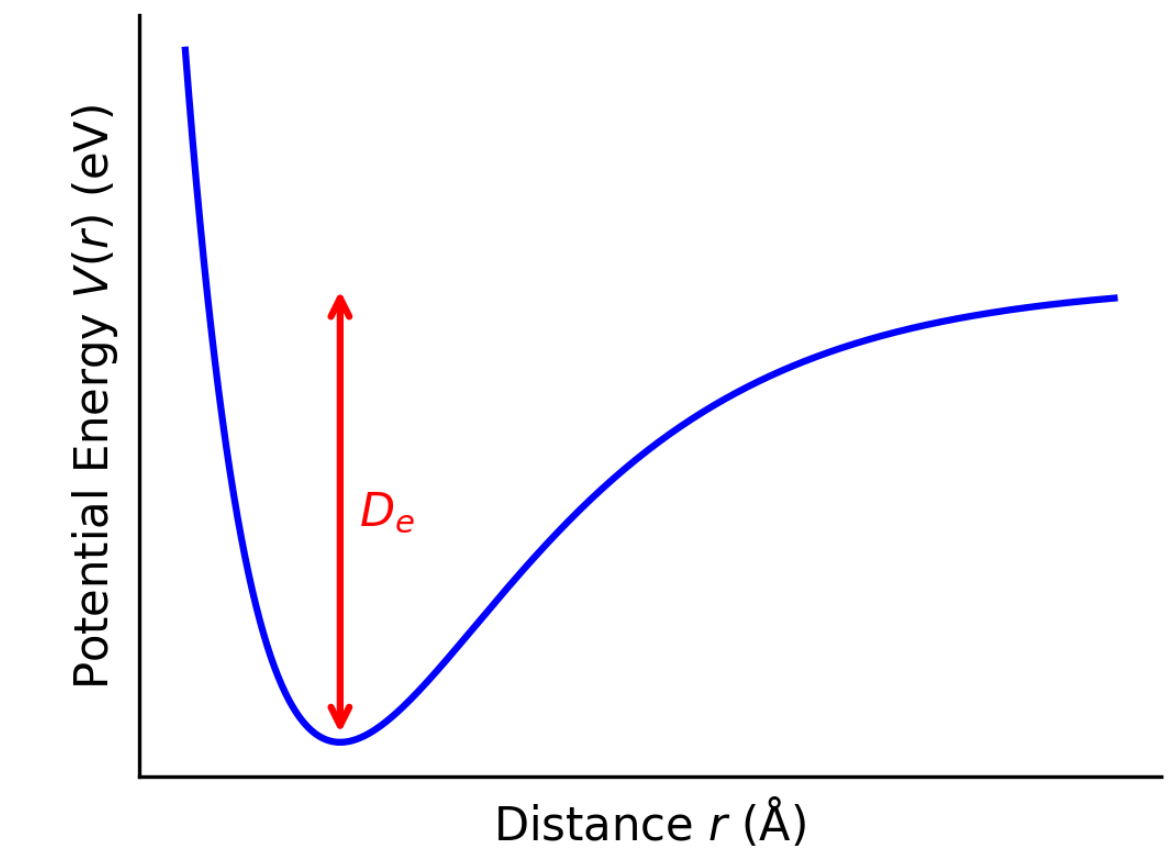
methods



CASSCF & MS-CASPT2 are used for every symmetry irrep and spin state. A state interaction (SI) calculation is performed with spin-orbit (SO) interactions included.



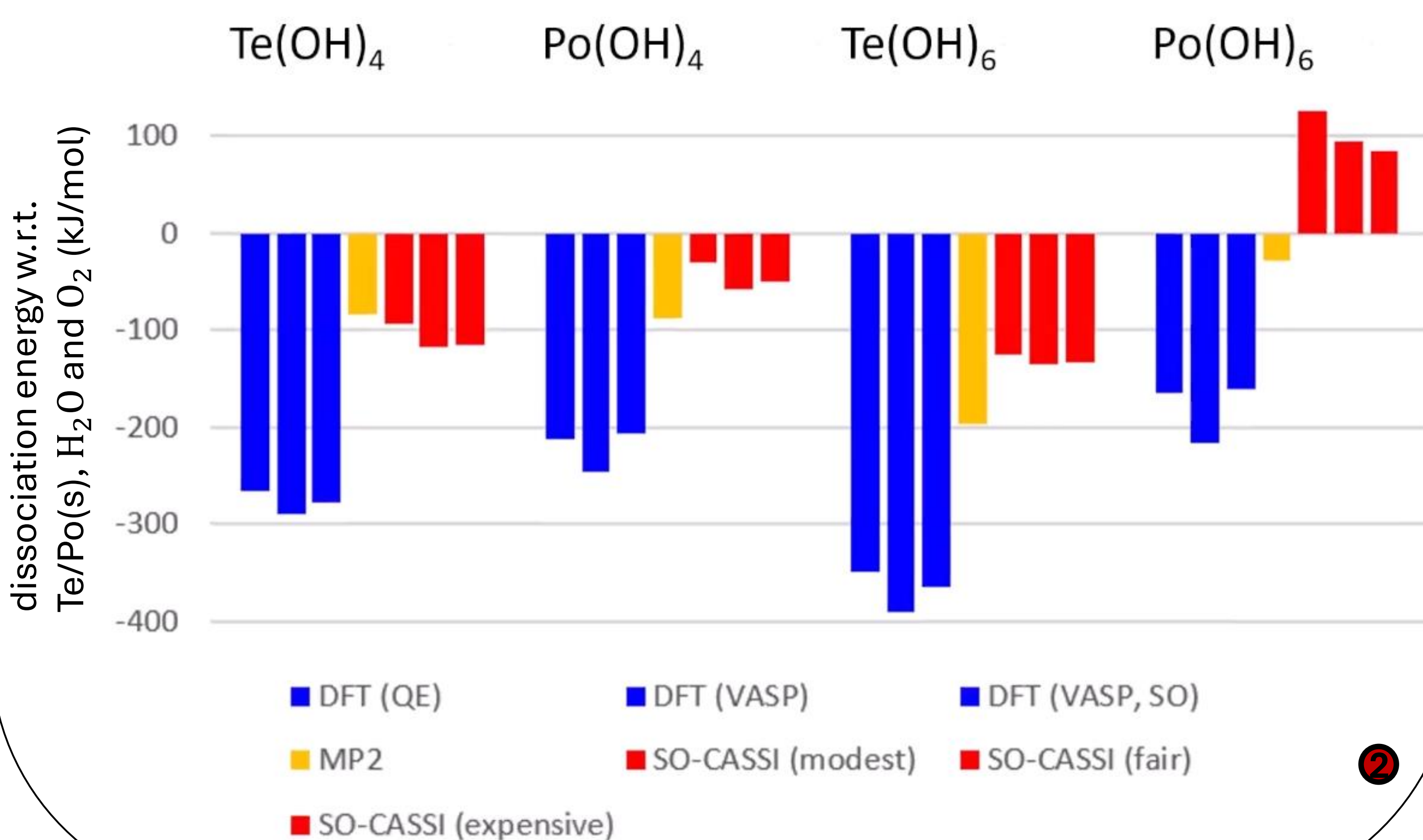
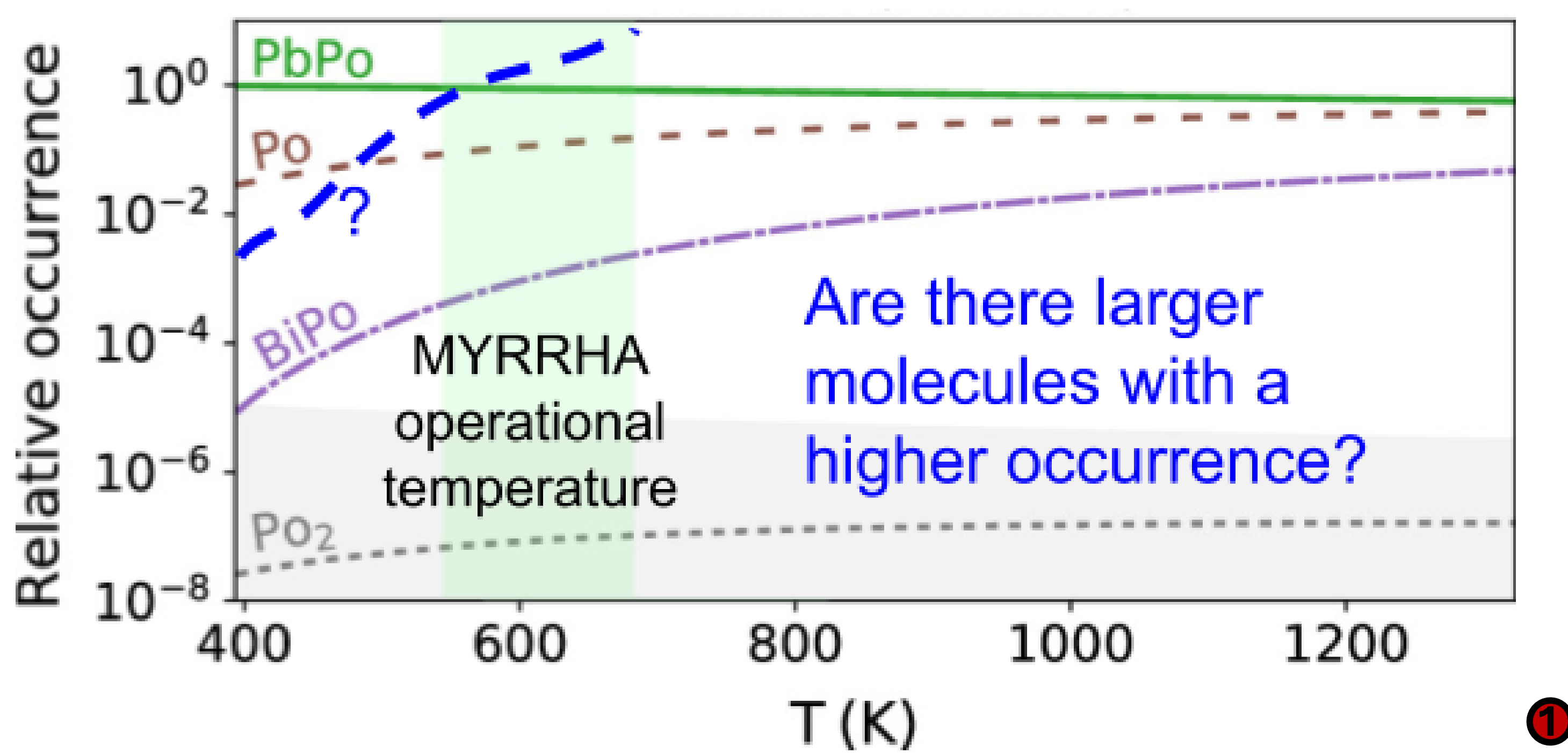
For every Po containing molecule of interest, a neural network is trained on rattled configurations, MP2 and SO-CASSI energies.



$$G_{tot} = n \sum_{mol} x_i \Delta G(T, D_e, \omega_e, x_e, \alpha_e) + nRT \sum_{mol,at} x_i \ln(x_i)$$

SO-CASSI energies over increasing distance between the molecules' constituents result in a dissociation profile from which the relative occurrence of the molecule, x_i , can be derived using: $\min(G_{tot}) \rightarrow x_i$

previous work



our planned approach

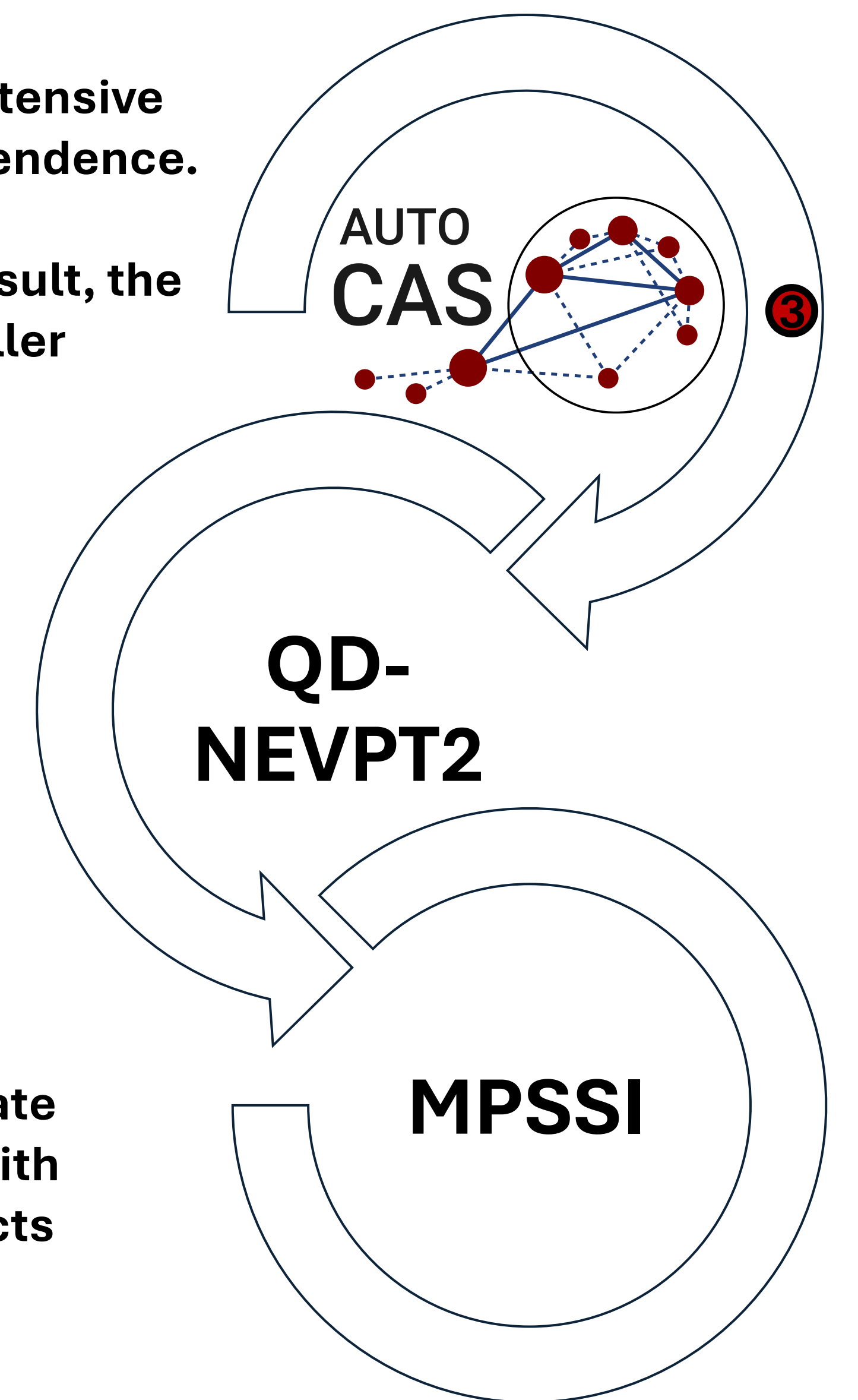
	Geometry	Energy
DFT	qualitative	not reliable
MP2	qualitative	fair approximation
SO-CASSI	too expensive	meets experiment

- CAS selection is time extensive due to its geometry dependence.
- Based on the $\text{Po}(\text{OH})_6$ result, the focus should be on smaller molecules.

Multistate version of N-electron valence state perturbation theory (NEVPT) doesn't suffer from intruder states unlike CASPT2.

Matrix Product State State Interaction combined with spin orbit coupling effects will be used.

DMRG-based workflow \rightarrow competitive from CAS(14,14) ④
 TeO_2 benchmark \rightarrow large active space test case



① Mertens, M. A., Aerts, A., Infante, I., Neuhausen, J., & Cottenier, S. (2019). Po-containing molecules in fusion and fission reactors. *The journal of physical chemistry letters*, 10(11), 2879-2884.
 ② Okan Köksal et al, submitted
 ③ Li Manni, G., Fdez. Galván, I., Alavi, A., Aleotti, F., Aquilante, F., Autschbach, J., ... & Lindh, R. (2023). The OpenMolcas Web: A community-driven approach to advancing computational chemistry. *Journal of chemical theory and computation*, 19(20), 6933-6991.
 ④ Freitag, L., Keller, S., Knecht, S., Lindh, R., Ma, Y., Stein, C. J., & Reiher, M. (2018, November). A quick user guide to the SCINE-QCMAquis software suite for OpenMolcas