

**Supplementary Figure 1**. The distribution of m/z distances (mass accuracies) between theoretical and observed peaks in appropriately sized error tolerance windows. For low mass accuracy data,  $\pm 0.25$  Da window covers the mixture of a unimodal distribution (signal peaks) and the flat distribution (noise peaks). For high mass accuracy data,  $\pm 0.025$  Da window covers the range. LuciPHOr attempts to learn these distributions from each dataset, with a user-defined mass tolerance window. Green and red lines indicate the fitted densities for matched and random peaks respectively.