To study the Ni-embedding behavior on the ZnO monolayer, a Zn atom is replaced by a Ni atom and then the geometric optimization is conducted. Also, the formation energy (E_f) is defined to reflect the difficulty to embed a Ni atom within the ZnO surface, calculated as:

$$E_f = E_{Ni\text{-}ZnO} - E_{ZnO} - E_{Ni} + E_{Zn} \quad (1)$$

in which $E_{Ni\text{-}ZnO}$ and E_{ZnO} represent the total energy of Ni-ZnO and pristine ZnO system, while E_{Ni} and E_{Zn} represent the chemical potential of Ni and Zn atom in their bulk structure. After Ni-embedding, one can see in **Figure S1** that the Ni-O bond is measured as 1.88 Å and the E_f is obtained as -1.05 eV. We can find that the E_f here is not as negative as the E_b for Ni-decorating on the T_O site (-1.75 eV), which reveals the better chemical stability for Ni-decoration on the ZnO monolayer in comparison with the embedding manner. Therefore, we determine the Ni-decorating on the T_O site of the ZnO monolayer as the preferred Ni-ZnO morphology.

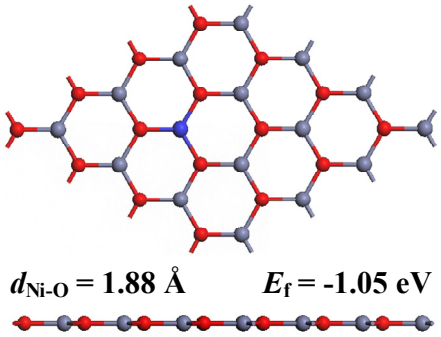

Figure S1 Morphology of Ni-embedded ZnO monolayer.

The adsorption energy (E_{ad2}) is defined to evaluate the adsorption performance of pristine ZnO monolayer upon CO and HCHO molecule, calculated as:

$$E_{ad2} = E_{ZnO/gas} - E_{ZnO} - E_{gas} \quad (1)$$

in which $E_{ZnO/gas}$ and E_{ZnO} represent the total energies of ZnO/gas system and ZnO system, respectively. The optimized CO and HCHO adsorption system as well as their band structure are plotted in **Figure S2**. One can see from **Figure S2** that the E_{ad} and Q_T for CO and HCHO systems are obtained as -0.18 eV and 0.061 e, and -0.28 eV and -0.035 e, respectively, which are much smaller than that in the Ni-ZnO systems indicating the weaker adsorption performance of the intrinsic ZnO monolayer compared with the Ni-decorated counterpart. Besides, the bandgap in the CO and HCHO systems are calculated as 1.896 and 1.887 eV, respectively. We can find that the

bandgap of the ZnO monolayer is increased by 0.027 and 0.018 eV, respectively after adsorption of CO and HCHO molecule. In comparison with the Ni-ZnO/gas systems, one can infer that the sensitivity in the pristine ZnO/gas systems are much smaller than that in the Ni-ZnO/gas systems. Therefore, the Ni-decorating is quite significant and essential to improve the adsorption and sensing properties of the ZnO monolayer upon gas species.

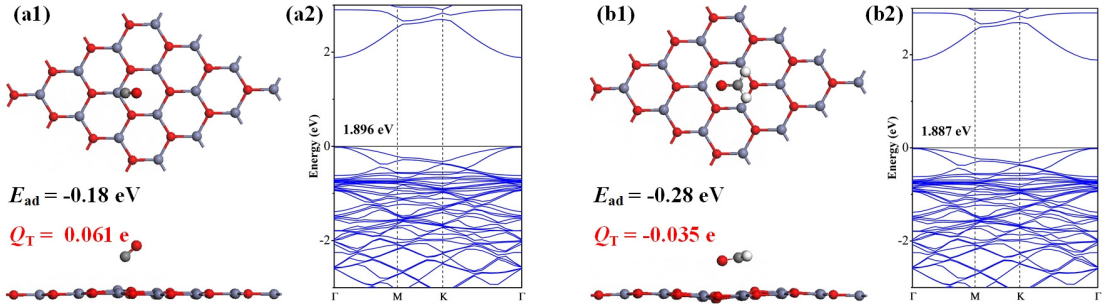

Figure S2 Adsorption configurations and band structure of ZnO/CO (a1)-(a2) and ZnO/HCHO (b1)-(b2) systems. In Band structure, the black value is the bandgap.