

Figure S1. iTLC of [⁸⁹Zr]Zr-DFO-KN035 radiolabeling.

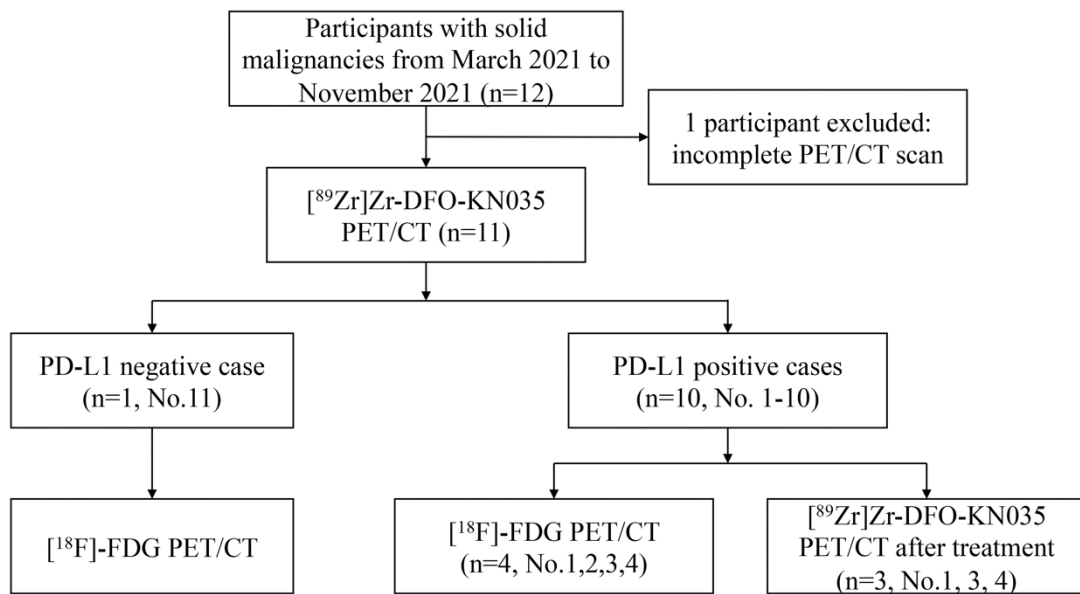


Figure S2. Study Graphset tables.

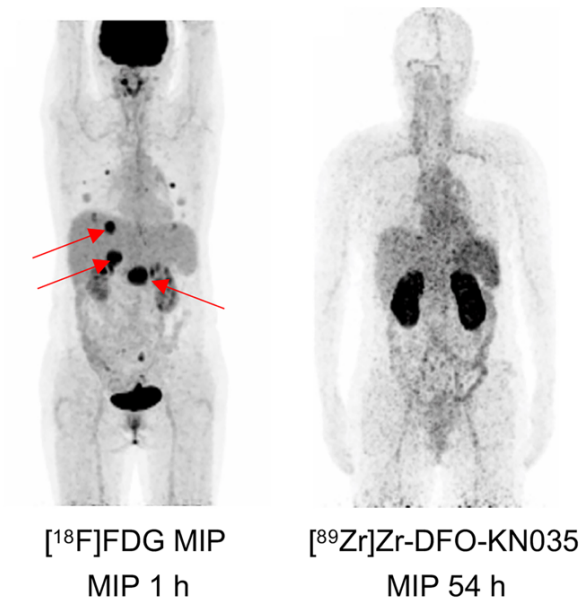


Figure S3. Comparison of the [¹⁸F]FDG and [⁸⁹Zr]Zr-DFO-KN035 PET imaging. MIP images of a PD-L1-negative patient (colorectal cancer and liver metastasis) obtained with the PET tracers [¹⁸F]FDG (scanned at 1 h after injection) and [⁸⁹Zr]Zr-DFO-KN035 (54 h).

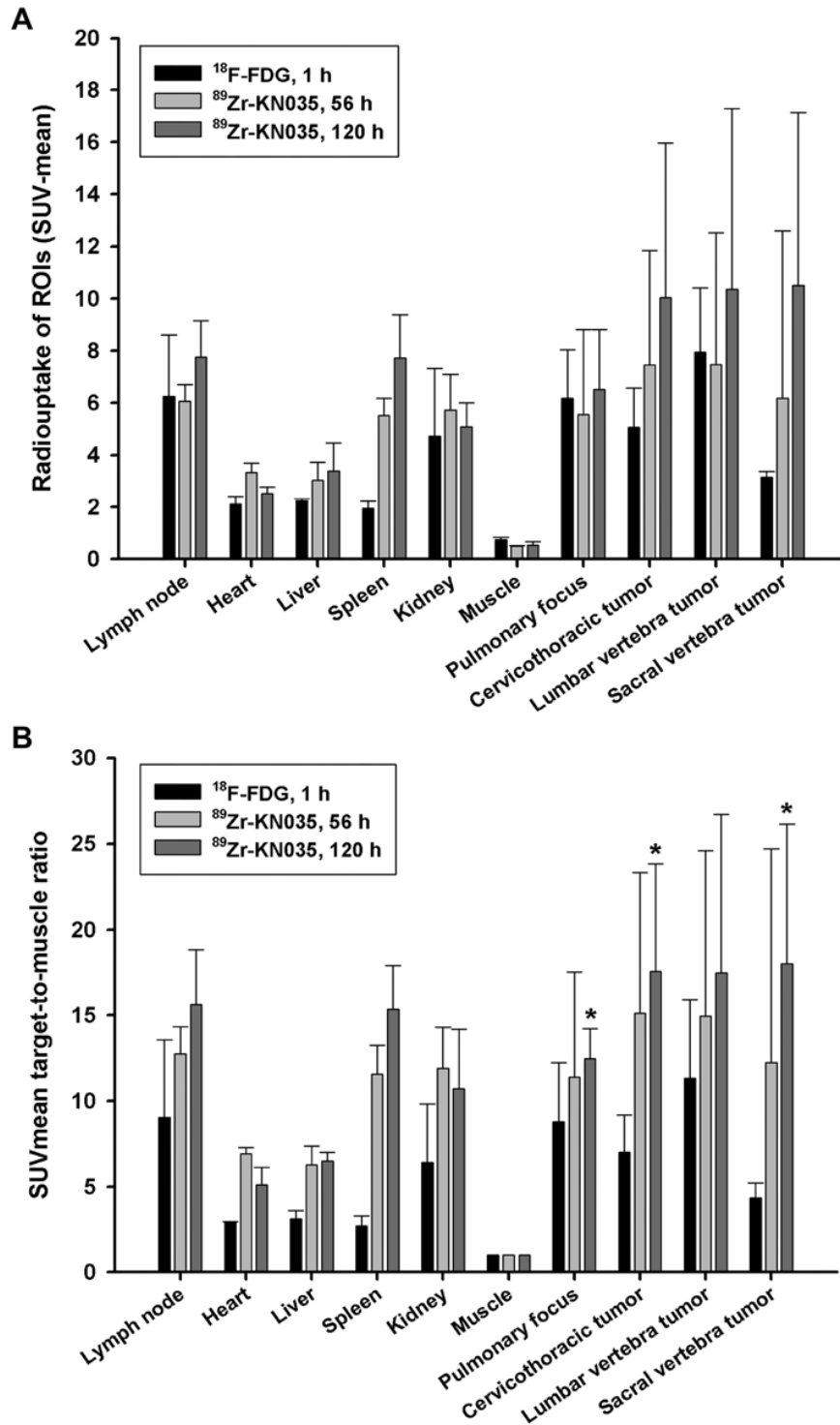


Figure S4. Comparative analysis of two PET/CT methods for the imaging diagnosis of primary lung cancer and multiple metastases in PD-L1-positive patients. (A) Statistical chart of radioactive uptake values (SUVmean) at different regions of interest (ROIs) in the ^{18}F FDG (1 h) and ^{89}Zr Zr-DFO-KN035 (56 and 120 h) PET imaging of three PD-L1 positive patients with

lung primary cancer and multiple metastases. Data are expressed as the mean \pm SD, and $n = 3$ for each site. **(B)** Statistical chart of the target-to-background ratios of SUV_{mean} ($[^{18}\text{F}]\text{FDG}$ 1 h and $[^{89}\text{Zr}]\text{Zr-DFO-KN035}$ 56 and 120 h) of different ROIs of tissues (background tissue and muscle). Data are expressed as the mean \pm SD, and $n = 3$ for each site. One-way ANOVA was used and, * $p < 0.05$ vs. $[^{18}\text{F}]\text{FDG}$ (1 h) PET group.

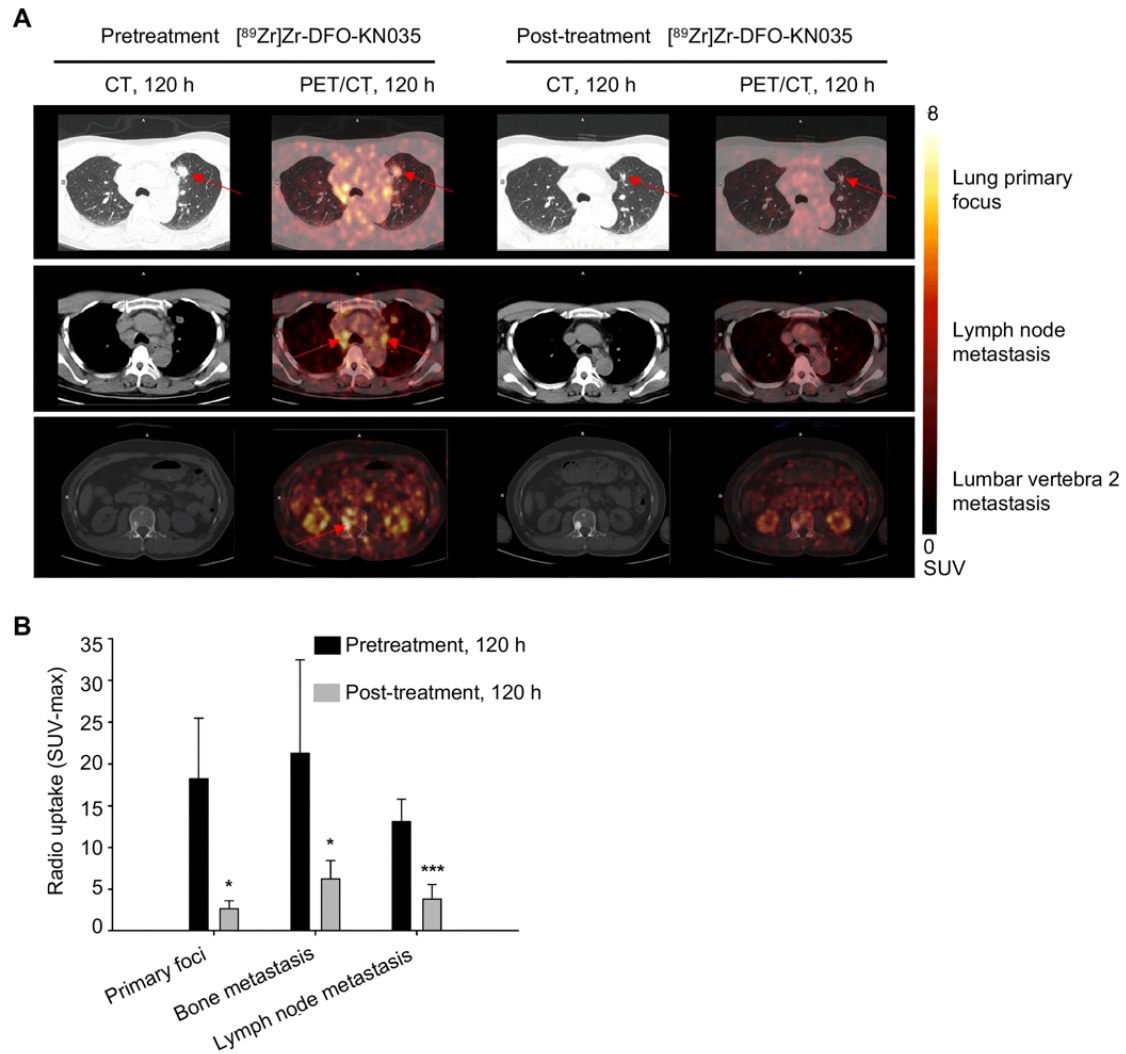


Figure S5. Comparison of primary tumors and metastases in PD-L1-positive patients using ^{89}Zr Zr-DFO-KN035 immunoPET before and after treatment. (A) Representative (Case No.1) CT and PET/CT images of the layers of primary focus (first row), lymph node metastasis (second row), and bone metastasis (third row, lumbar vertebra 2) sites in the same patient with PD-L1-positive cancer (lung primary cancer with bone, lymph node, and brain metastasis) obtained by using ^{89}Zr Zr-DFO-KN035 before (120 h) and after (120 h) treatment. Red arrows show the locations of primary tumor and metastasis focus sites. (B) Statistical chart and comparison of the radioactive uptakes (SUVmax) of the primary and metastatic tumors in ^{89}Zr Zr-DFO-KN035 immunoPET imaging before (120 h) and after (120 h) treatment. Data are expressed as the mean \pm

SD, and $n = 3-7$ for each site. Paired samples t-test was conducted. * $p < 0.05$, ** $p < 0.01$, and *** $p < 0.001$ vs. pretreatment group.

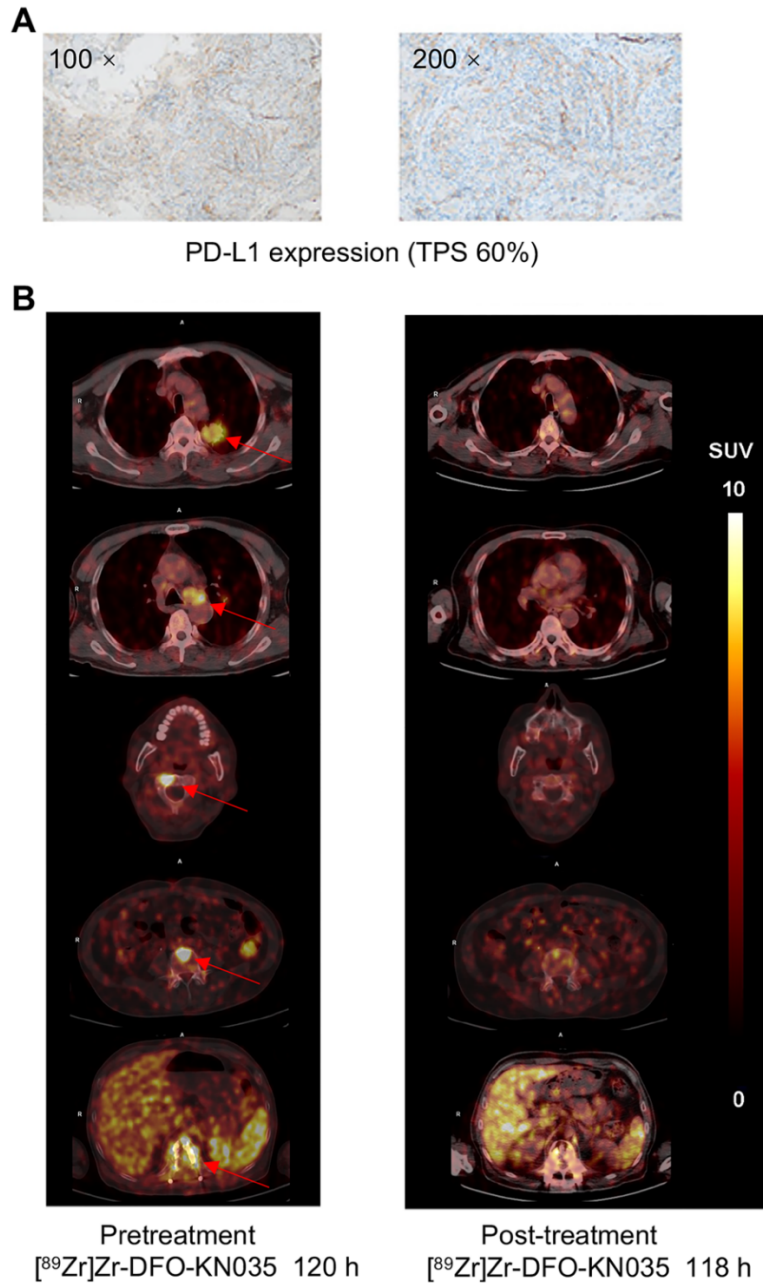


Figure S6. PET imaging using [⁸⁹Zr]Zr-DFO-KN035 before and after anti-PD-1 therapy of a patient (Case No.3) with PD-L1-positive lung cancer. (A) Immunohistochemical result of a PD-L1 positive patient (TPS = 60%) with lung cancer and multiple bone metastases. (B) Representative PET/CT images of the layers of lung primary cancer (first lane) and some bone metastatic sites (lower lanes) in the same patient taken by using [⁸⁹Zr]Zr-DFO-KN035 at pre- (120 h) and post-treatment (118 h).

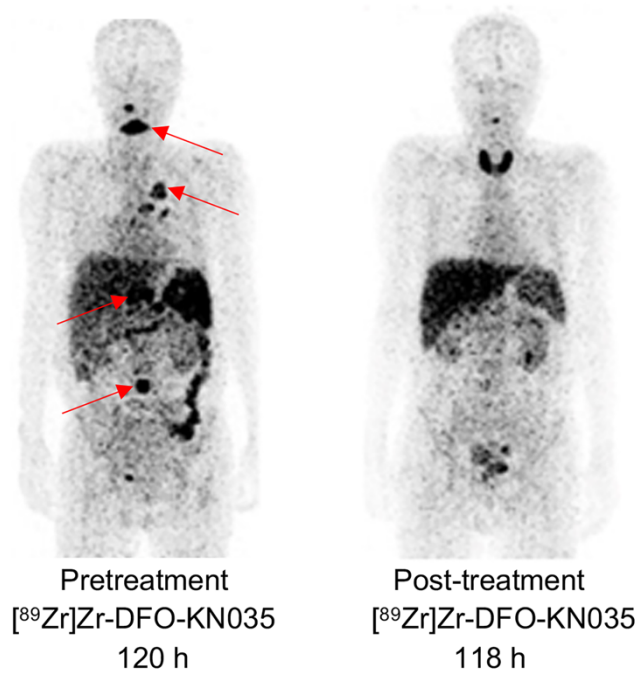


Figure S7. PET imaging using [⁸⁹Zr]Zr-DFO-KN035 before and after anti-PD-1 therapy of a patient (Case No.3) with PD-L1-positive lung cancer. (A) MIP image of a PD-L1 positive patient (TPS = 60%) with lung cancer and multiple bone metastases. (B) MIP images of the patient obtained by using the molecular-imaging tracer [⁸⁹Zr]Zr-DFO-KN035 before (120 h post-injection) and after (118 h post-injection) anti-PD-1 treatment.