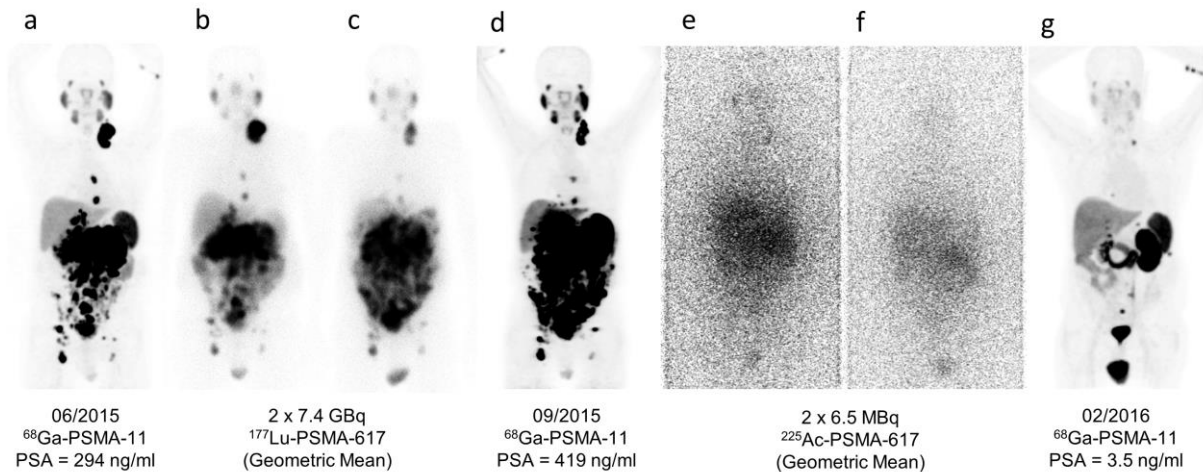


**Supplemental Figure 1:** Posttherapeutic planar scans based on the coemission of photons during alpha-decay acquired 24 h after cycles 1-3 demonstrate sufficient tumor targeting and stepwise response over the conducted treatment fractions.



**Supplemental Figure 2:** Posttherapeutic planar scans after 2 cycles of 7.4 GBq of  $^{177}\text{Lu}$ -PSMA-617 (geometric mean) demonstrate that nonresponse was not caused by insufficient tumor targeting. Emission scans during  $^{225}\text{Ac}$ -PSMA-617 treatment already imply a stepwise tumor response.