Vectorized internal radiotherapy relies on radionuclides coupled to biological vectors capable of selectively targeting tumour tissues. In this context, terbium isotopes are of particular interest because different terbium radionuclides can potentially be used both for therapeutic and diagnostic approach (Theranostic) with the same vector and chelator for complementary purposes: assessing biodistribution before therapy and delivering the therapeutic dose. This thesis focuses mainly on a potential theranostic couple: Tb-155, a radionuclide suitable for SPECT imaging, and Tb-161, a therapeutic radionuclide that can also be imaged through its low energy gamma emissions. Particular attention is given to Tb-156, which can be co-produced with Tb-155 during standard cyclotron production and whose high energy photons can degrade the quality of Tb-155 SPECT images.

This work combines experimental acquisitions, Monte Carlo simulations with GATE 10 and tomographic reconstruction. Experimental data were acquired within the PRISMAP framework using the ALBIRA S108 preclinical SPECT system at CHUV Lausanne, in particular with the NEMA NU 4-2008 image-quality phantom. These acquisitions were used to characterize the camera response and to validate the simulation workflow against measured spectra, reconstructed images, profiles and image quality metrics. Imaging experiments with Tb-161 showed that its low energy gamma emissions can provide usable SPECT images, although image quality remains limited by photon energy and system response. Experiments with a nominally pure Tb-155 source confirmed the feasibility of Tb-155 SPECT imaging, while a Tb-156-contaminated source showed the strong impact of radionuclidic impurities.

After validation, ALBIRA digital twin was used to simulate controlled levels of Tb-156 contamination. A second system, THIDOS, a mobile gamma camera with tungsten parallel hole collimation developed locally within the REV group at IJCLab, was also implemented as a medium energy comparison SPECT camera. The results show that the impact of Tb-156 strongly depends on the imaging system. With ALBIRA, only a few percent of Tb-156 generate a substantial detected contribution in the Tb-155 reconstruction window, leading to a rapid degradation of signal to noise ratio and cold cylinder contrast. For ALBIRA-like low energy systems, the Tb-156 activity fraction should remain close to 1\% and should not exceed about 2\% for reliable quantitative or contrast sensitive Tb-155 imaging. With THIDOS, tungsten collimation more effectively rejects high energy photons and provides better tolerance to contamination. For THIDOS-like medium energy systems, the acceptable fraction is less restrictive: it should remain below about 5\% for strict quantitative imaging, while 10\% remains more usable than in ALBIRA but is not considered optimal.

This thesis also includes dosimetric studies of Tb-161, Tb-155 and Tb-156 using two chelators, LiHOPO and TPACY. Tb-156 led to a substantial increase in absorbed dose to organs, while Tb-161 and Tb-155 produced dose values comparable to those reported in the literature. The comparison between chelators showed that LiHOPO provided better tumour retention, corresponding to a higher tumour-to-healthy-tissue uptake ratio.

Overall, the results show that Tb-155 is relevant for SPECT imaging, but that radionuclidic purity and camera design are critical for limiting the impact of Tb-156. They also support the interest of Tb-161 as a therapeutic isotope with imaging capability. This work highlights the need for an integrated approach combining isotope production, radiochemistry, instrumentation, Monte Carlo simulation and image reconstruction to define realistic conditions for the use of terbium isotopes in vectorized internal radiotherapy.