Development of a new portable instrument for in situ activity measurement of Radionuclides in Nuclear Medicine

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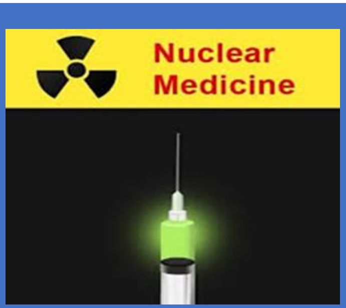
Aim of the Project



This project concerned with the development of portable detector for in situ activity measurement of radiopharmaceuticals, of interest in nuclear medicine . This project is aimed at providing sufficient metrological support for activity and dose measurement in Molecular Radiotherapy (MRT), a technique widely used in medicine for fighting against cancer.

Short half-life β-γ radionuclides have many applications in nuclear medicine and require the development of fast measuring methods in order to standardize their activity. For that reason we combined pure beta channel with gamma channel NaI(Tl) to implement $4\pi\beta-\gamma$ coincidence technique (TDCR) for activity measurement of those radionuclides with low uncertainty. In this project we calibrated TDCR and studied some pure beta emitter radionuclides (H3, Ni63, C14, Tc99, Sr90, and Y90). They have many applications in environmental, industry and medicine science

Radionuclides	Emission	Half-Life	Application
H3	$\beta-$	12,312 years	Radiotracer in radioluminescence
Ni63	$\beta-$	98,7 years	Detection of explosives traces, Electron capture detector in gas chromatography
C14	$\beta-$	5700 years	Radioactive tracer for medical test and radiocarbon dating in archology
Tc99	$\beta-$	$211{,}5 \times 10^3$ years	Diagnostic imaging procedure in nuclear medicine use this isotope as a radioactive tracer in SPECT
Sr90	$\beta-$	28,8 years	Radioactive tracer in medical studies
F18	$\beta+$	1,8289 hours	Nuclear medicine -Diagnosis (PET)
C11	$\beta+$	20,61 min	Nuclear medicine- Diagnosis and therapeutics (PET)
Y90	$\beta-$	2,684 days	Nuclear medicine and radiation oncology communities for radiation therapy

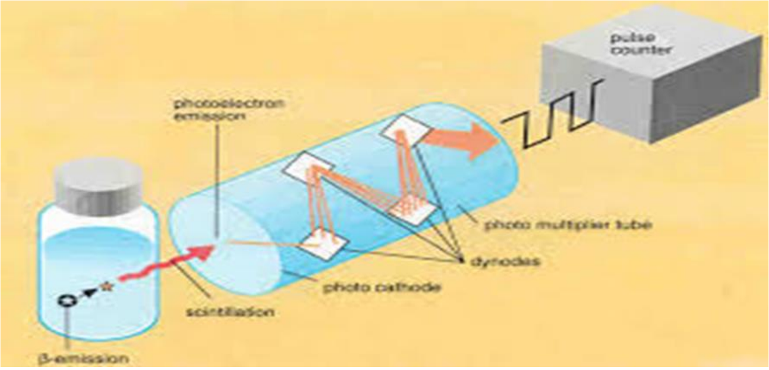
Beta emitter radionuclides and their applications

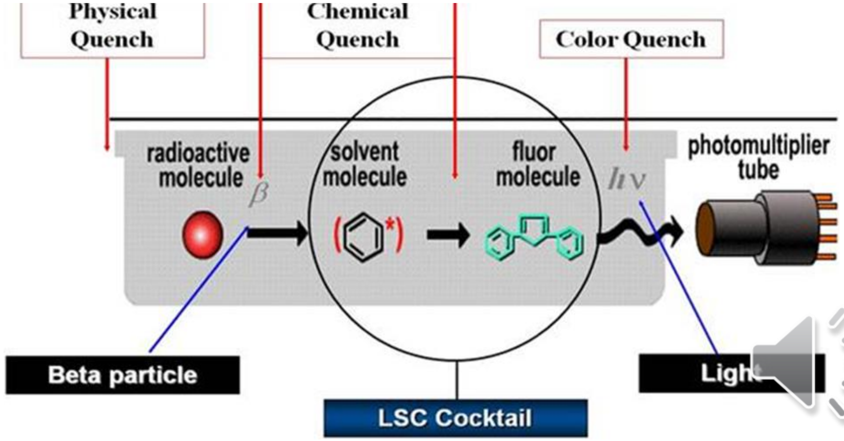
Liquid Scintillation Counting (LSC)

Liquid scintillation counting (LSC) has been a very popular technique for the detection and quantitative measurement of radionuclides. These include principally alpha and beta emitters, but may also include weak gamma, X ray and Auger electron emitters.

1. The liquid scintillation technique involves placing the **sample containing the radioactivity** into a container (plastic or glass), called **scintillation vial**, and adding a special scintillation cocktail.
2. Basically, the liquid scintillation process is the conversion of the **energy of a radioactive decay event** into **photons**. The number of photons released by the scintillator is directly proportional to the energy of the beta particles
3. The detection of the scintillator light is done by using a photomultiplier (PMT), which converts a small amount of light into electrons (via the photoelectric effect), and amplifies the number of electrons many times (via a cascade), delivering a "strong" electrical pulse at its output

Liquid Scintillation Process

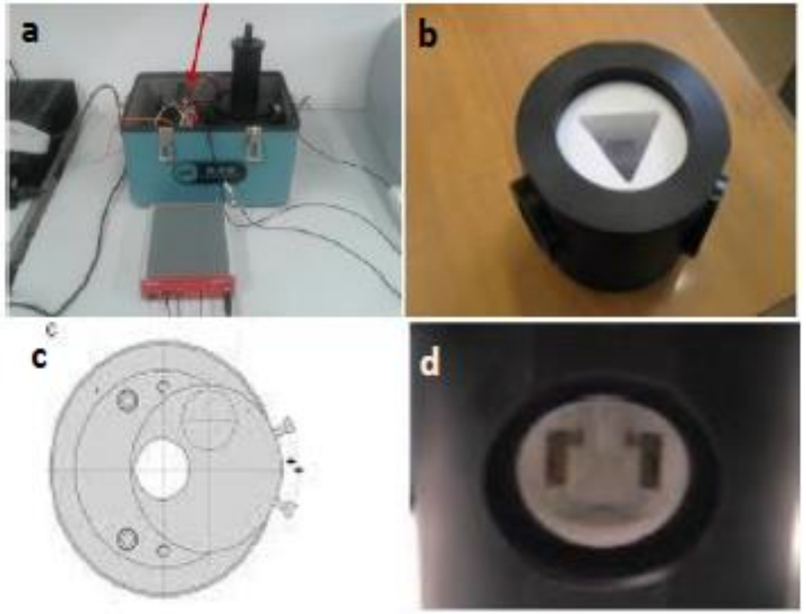




TDCR Detection System

The Triple to Double Coincidence Ratio (TDCR) is the most sensitive method which is used for determining absolute activity in the Liquid Scintillation Counting (LSC). It is especially developed for pure beta and pure electron capture (EC) radionuclide in the LS cocktail without using any reference source.

- The detection efficiency can be calculated by using a statistical and physical model of the photon distribution which is emitted by the scintillating source in Liquid scintillator counter.
- Due to the low penetrative power of beta radiation, the detection efficiency of beta emitters, especially the low energy ones, is quite low; the quantification of beta emitting isotopes is quite difficult without LSC.

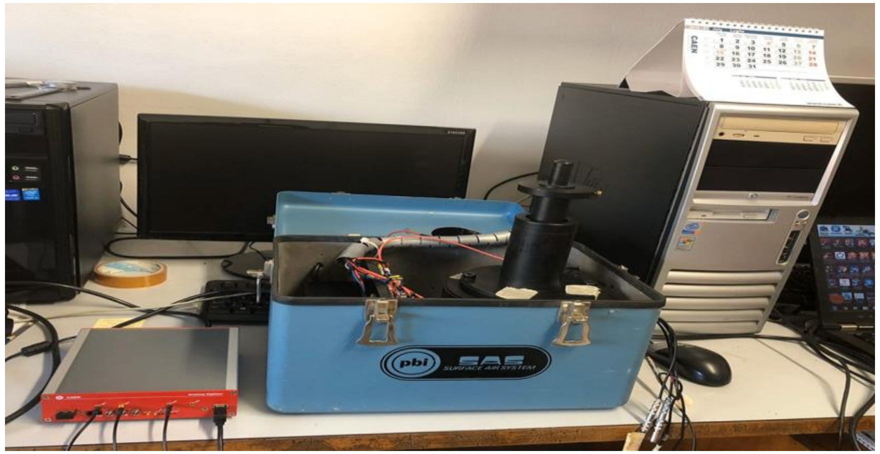


TDCR system and Optical chamber

1- Optical Chamber

- The new ENEA -TDCR counter optical chamber has an inner prismatic shape with equilateral base (L=60 mm and H=73 mm). The optical chamber is made from white PTFE (Teflon) and it is surrounded by a black PTFE cylindrical box of ($\emptyset = 150\ mm\ and\ H = 150\ mm$), outer cylindrical shape to fit precisely with this box.
- It is supplied with a lift and a shutter that can be used to remove or insert the vials inside the optical chamber without turning off the PMTs.
- The total weight of the portable TDCR detector is less than 6 kg.

The TDCR detection system



2- Photomultipliers

- For the portable TDCR detector at ENEA three Hamamatsu Photonics type R7600U-200 square package type of photomultiplier tubes (PMTs) are selected.
- Photomultiplier tubes with small dimensions ($30\ mm \times 30\ mm$) are used, with **10 dynodes**. They are arranged around the optical chamber in a 120-degree planar geometry.
- The selected PMTs have a high quantum efficiency, high gain, short time of response, relative wide range wavelength about (300 – 600 nm), and relatively low supply voltage (approximately 900 V) with the cathode grounded to operate in the photon counting mode. The PMTs are Powered by three compacts (dimensions: $46\times24\times12\ mm$^3, weight: 31 g), on-board type HV power supply units (C4900 series, provided By Hamamatsu Photonics).

Hamamatsu R7600U-200 PMT

3-Digitizer

In different National Metrology Institutes (NMIs) recently carried out a new improvement in radionuclide metrology with FPGA-based nuclear. DT5720 CAEN Digitizers version is platform independent instrument housing high speed (250 MS/s) multichannel ADCs with local memory and FPGAs for real-time data processing.

The electronics which are used for the TDCR applications, must have high power to record very fast signals. In different National Metrology Institutes (NMIs) recently carried out a new improvement in radionuclide metrology with FPGA-based nuclear.

TDCR detector is equipped with DT5720 Desktop CAEN digitizer, and is portable.

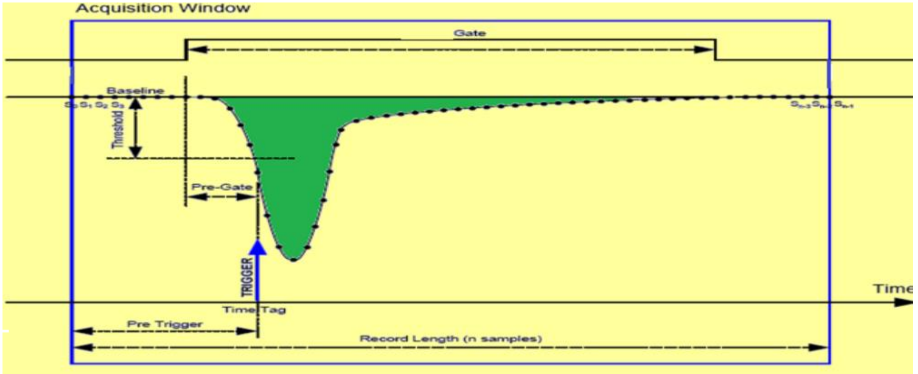
The computer remotely controls the digitizer via CAEN Control Software, which runs on Linux or Windows operating systems; the software gives the possibility to independently set the acquisition parameters (gate length, DC offset, pulse polarity, threshold level, etc.) for each channel..

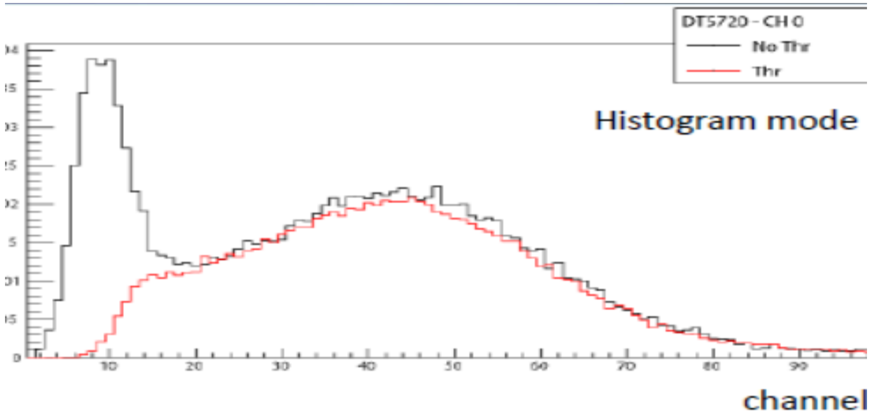
(DT5720) Digitizer

Data Analysis

- The portable TDCR detector was directly linked to the desktop-type CAEN digitizers DT5720.
- This version of digitizer allows the setting of the acquisition gate width in time for each triggered pulse, in order to cover the whole input signal.
- The physical information is carried out by the amplitude as a function of time and by the total charge Q within the gate. Q is proportional to the energy of the particles and can be calculated by performing the gate integration over the input pulse.
- The DT5720 module elaborates and records the data over the threshold and saves into a file the charge Q and a stream of Trigger Time Tag (TTT).
- In order to equalize the response of each single PMT a pulse shape spectrum for each channel is recorded without any vial inside the optical chamber .
- We selected the high voltage of the 3PMTs about 900 V to have the same maximum peak of Single Electron Peak SEP and the threshold about 45 and 8 respectively for each 3PMTs channel.

acquisition window





Single Electron Peak (SEP) of Ch0

Experimental Setup

- The sources contained **10 mL of Ultima Gold (UG)** as a liquid scintillator in 20 ml glass vials and approximately **10 mg of radioactive solution** (with different aliquots of CCl_4 as a quenching agent).
- **Radionuclides**
- Two low-energy beta nuclides (^{3}H and ^{14}C).
- Medium-energy beta nuclide (^{99}Tc).
- High-energy beta emitter nuclide (^{90}Sr, ^{18}F and ^{90}Y).
- To perform the TDCR analysis the charge integration value Q is recorded with a **Gate= 24 ns**.
- The radiochemistry laboratory of the ENEA- INMRI prepared the sources. The master solutions are checked by high energy-resolution HPGe spectrometry to perform a preliminary gamma-impurity check

Radionuclides	Mean Energy of Beta Spectrum (keV)	Experimental TDCR
H-3	5.68	0.3106
C-14	49.16	0.8903
Tc-99	94.6	0.9492
Sr-90	196	0.9757
F-18	249.5	0.9917
Y-90	926.7	0.991

- A blank source is prepared for background measurements, containing only 10 ml of UG.
- We set the coincidence resolving time $t_c = 140$ ns and dead time $t_{dead} = 50$ µs.
- The signal information from each PMT (3 channels) are recorded by the digitizer and the CAEN analysis software.

$$\frac{T}{D} = \frac{\varepsilon_T}{\varepsilon_D} = TDCR$$

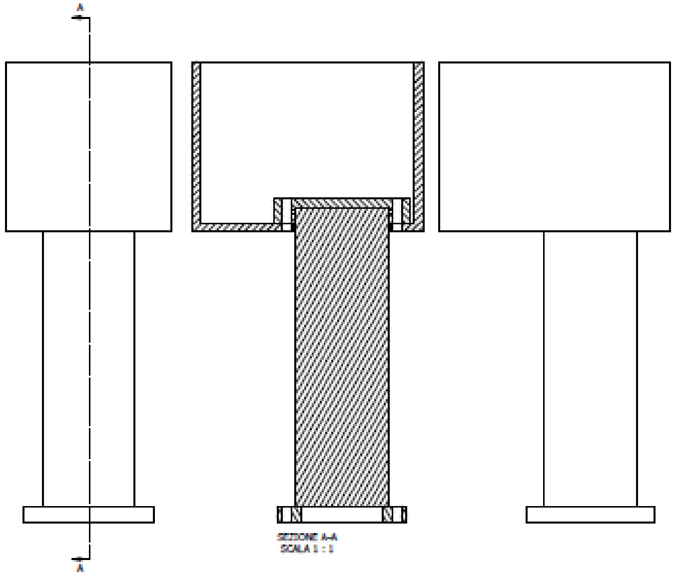
- Next, we extract single counts of each PMT (A, B, and C), their sum (S), the real time (t_{real}), the live time (t_{live}) and the coincidence counts AB, BC, AC.
- TDCR parameter can be calculated by taking into account **the ratio of the count rate of triple coincidence T to the logical sum of double coincidence D, which is equal to the ratio of the efficiency of triple coincidences (ε_T) to the efficiencies for the logical sum of the double coincidences (ε_D) for high energy beta emitters with uncertainty estimated of about 1%.**

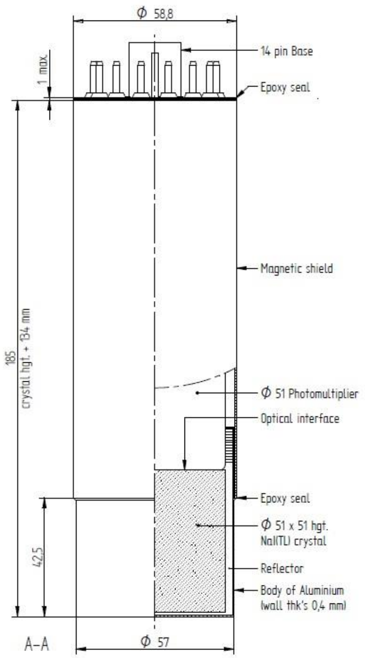
Monte Carlo simulation technique

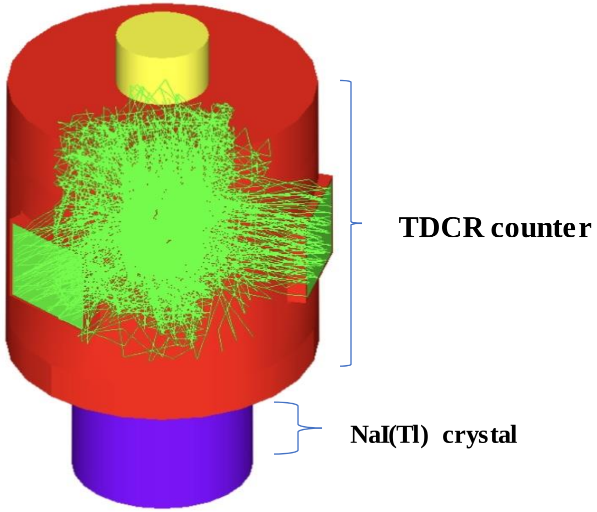
- In this project Geant4 codes are developed for modeling the TDCR counter and simulating the different physical processes taking place in the optical chamber.
- GEANT4 is a “Toolkit for the simulation of the passage of particles through matter". It is widely used, freely available, continuously updated and provides extensive physics libraries and geometry design classes. It provides simulations based on theory, data, or parametrization.
- Geant4 can be used to create such virtual reality we need to model the particle-matter interactions, geometry and materials in order to propagate elementary particles into the detector and define the primary particles. Transports a particle step-by-step by taking into account the interactions with materials and external electromagnetic fields until the particle loses its kinetic energy to zero.
- In liquid scintillation counting, Monte Carlo is used to calculate the probability of the interaction of beta particles in a given scintillator and the absorbed energy distribution. The absorbed energy distribution of photons is most often calculated in discrete energy bins.

Radionuclides	Mean Energy of Beta Spectrum (keV)	Monte Carlo Simulation TDCR Parameter	Uncertainty (Δ%)
H-3	5.68	0.3634	± 0.1122
Ni-63	17.434	0.7317	± 0.1309
C-14	49.16	0.91	± 0.1372
Sr-90	196	0.9732	± 1.4223
Y-90	926.7	0.9961	± 1.4143

4π(LS)β–γ detection system





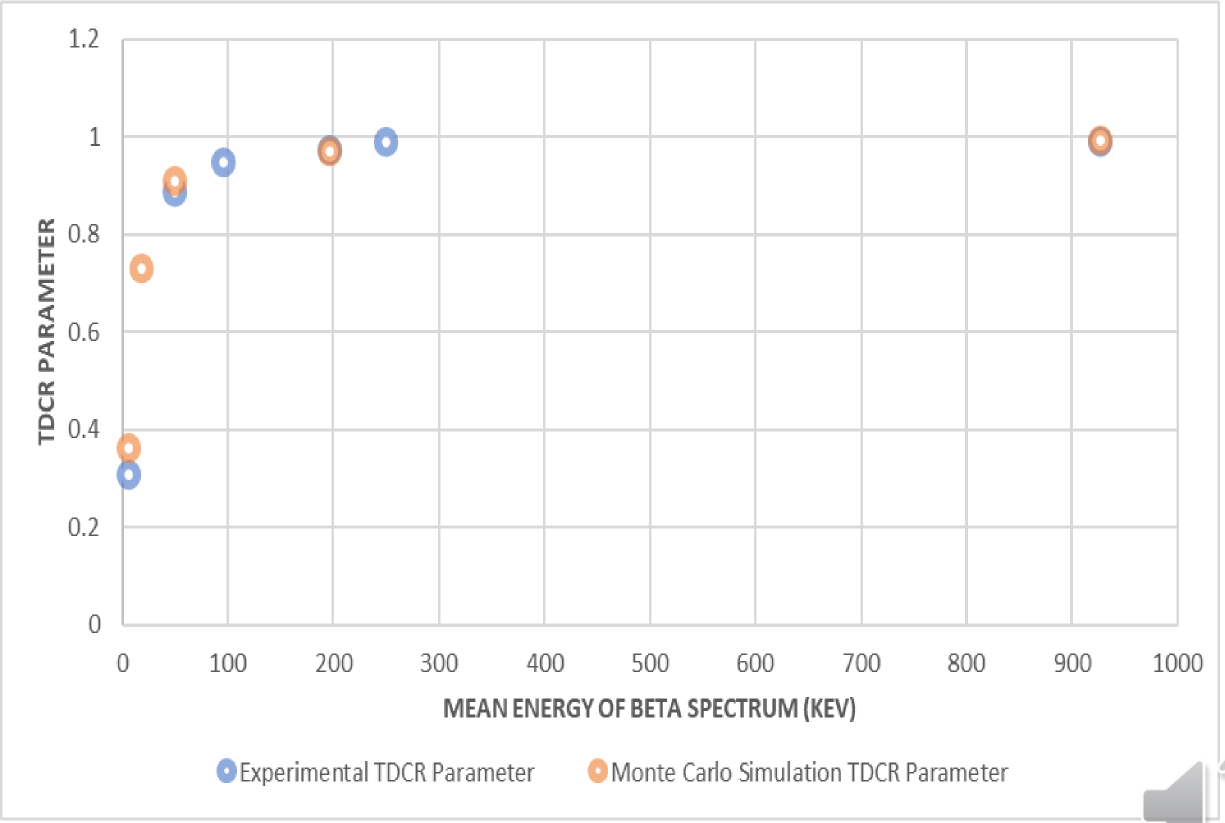


Simulated 4π(LS)β–γ detector system (GEANT4 Code)

- The LS β-channel is equipped with a complete TDCR setup as described before. The 4π(LS)β–γ detection system is based on the use of a standard TDCR system for liquid scintillation measurements in the beta channel and an (3”×3”) NaI(Tl) scintillator detector in (0.4 mm) thick of Aluminum housing as the gamma channel which is implemented at the bottom of the TDCR counter.
- Basically, a detection setup for coincidence counting can be described as two detection β- and γ-channels which are connected through specialized electronics allowing coincidence counting.
- The TDCR model is extended to 4π(LS)β–γ coincidence counting to enable the simulation of the efficiency–extrapolation technique by the addition of a γ-channel. This simulation tool aims at the prediction of systematic biases in activity determination. Especially developed for the standardization of beta and pure electron capture emitters, the TDCR method can be extended to simple beta-gamma and EC-gamma decay-scheme nuclides.
- The electronics and coincidence analysis Software will be developed in collaboration with CAEN and that this is my work for the 2nd PhD year.

Result

- TDCR parameter depends on the energy spectrum of beta emitter radionuclides, for high-energy beta emitter the TDCR parameter approaches the unity.
- This new device can be used as a traveling tool for in-situ measurements. It is results opened new interesting forwarding-looking for activity measurements of beta and alpha emitters. This device has a particular benefit in many applications in the field of nuclear medicine and in the nuclear energy industry.



Thanks for your attention