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Application of the Pulsed Fast/Thermal Neutron-Gamma Method for Soil Elemental Analysis

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Neutron technology for elemental analysis

Area of application

- Detection of threat material (explosives, drugs and dangerous chemicals)
- Archaeological site surveys and provenance studies
- Determination of elemental composition of human and animal tissues
- **Soil science**
- Real-time elemental analysis of bulk coal moving on conveyor belts
- Well logging during oil exploration
- Planetary science

Background of evolution

- Availability of commercial products: portable pulse neutron generators, high efficiency gamma detectors, reliable electronics, and measurement processing software
- Detailed knowledge of neutron interactions with nuclei (processes, neutron database of cross-sections)

Neutron-Gamma Methods

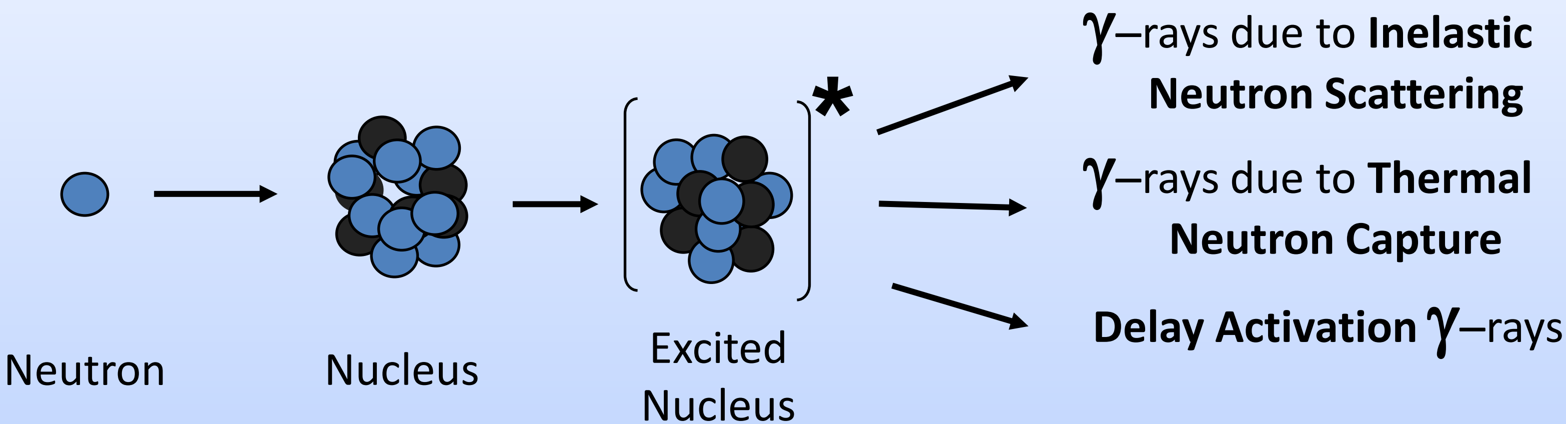
- PGNAA - Prompt Gamma Neutron Activation Analysis
- PFNA - Pulsed Fast Neutron Analysis
- **PFTNA - Pulsed Fast/Thermal Neutron Analysis** (Womble et al., 1995)
- PFNTS - Pulsed Fast Neutron Transmission Spectroscopy
- API - Associated Particle Imaging

Advantages

- Non-Destructive
- *In situ*; No sample preparation
- Multi-elemental analysis of large volumes
- Impact of local sharp changes in element content are negligible
- Scanning capability

General Principal of the N-G method

Neutron interaction with an atomic nucleus



Each kind of nucleus and process produces gamma-rays of a particular energy

Soil Element	Applied for analysis			
	Kind of Neutrons	Process	Cross-Section, b	Characteristic gamma line, MeV
Si	Fast	INS	0.52	1.78
O	Fast	INS	0.31	6.13
H	Thermal	TNC	0.33	2.22
C	Fast	INS	0.42	4.43
N	Thermal	TNC	0.08	10.8
Cl	Thermal	TNC	43	1.64

Scope of the Soil Element and Knowledge Importance

1. **Carbon**

- An universal indicator of soil **quality** that can impact many environmental processes, such as soil carbon sequestration, fertility, erosion, and greenhouse gas fluxes

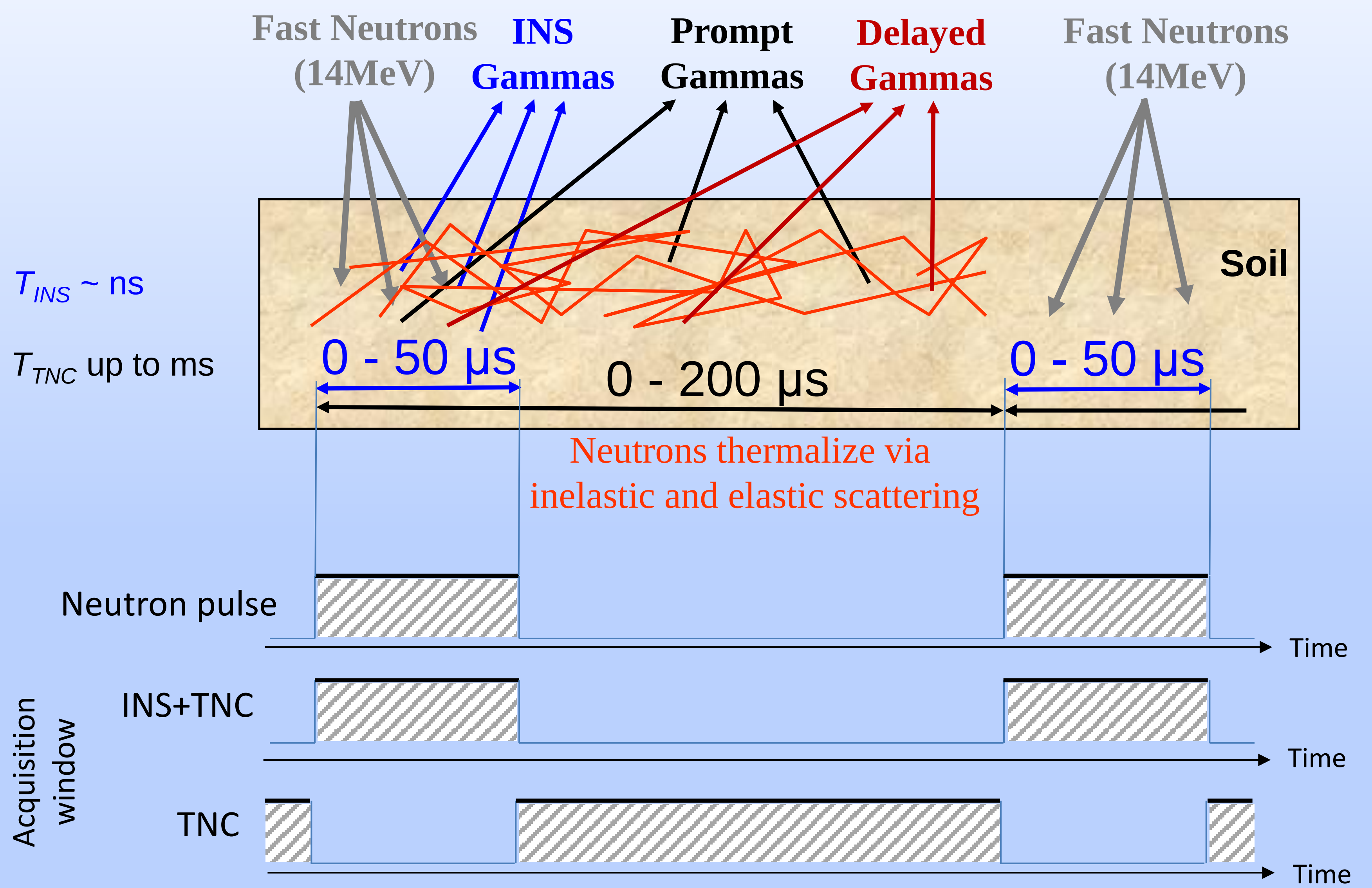
2. **Nitrogen**

- Detection of dangerous hidden objects in soil (explosives) by presence of nitrogen and C/N ratio

3. **Chlorine**

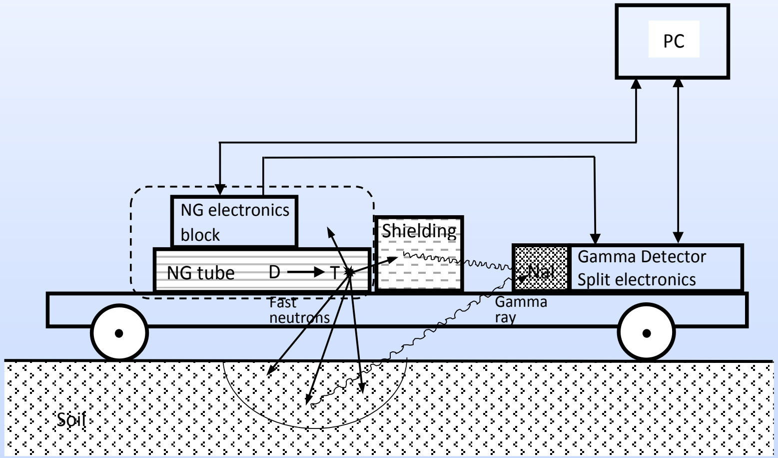
- Detection of soil chlorine (contamination and distribution) and underground objects that contain chlorine

Time Separation of INS+TNC and TNC Gamma Spectra for the Pulsed Fast/Thermal Neutron-Gamma Method

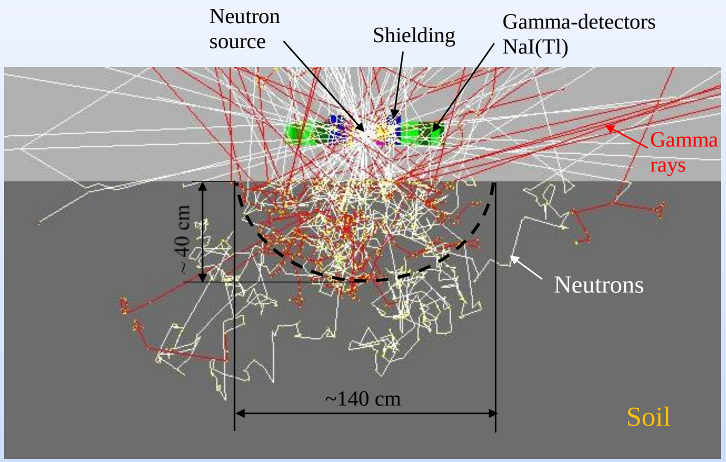


Soil Carbon Measurements

Field System Geometry and Design



MC simulation screen shot



Dimensions: 75 x 23 x 95 cm^3

Weight: ~ 300 kg

System Components:

Neutron Generator MP320

3 Gamma Detectors - NaI(Tl)

Neutron Detector

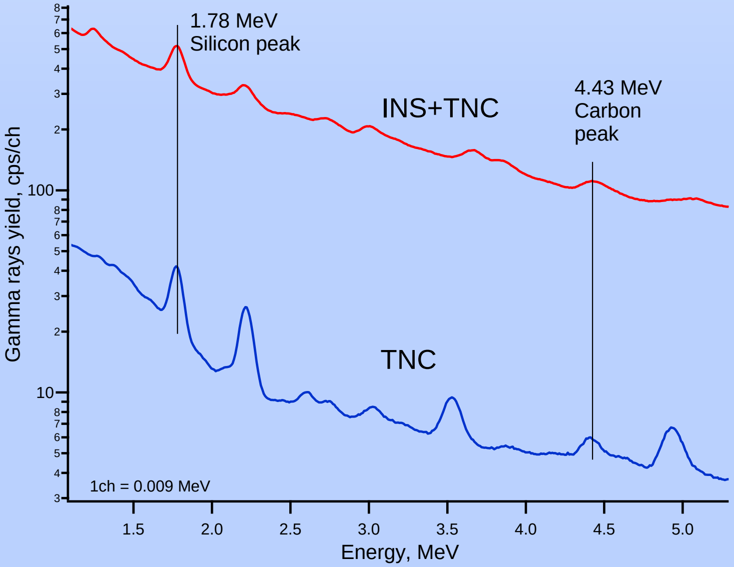
4 Batteries

Inverter

Laptop

Shielding

Experimental Soil Gamma Spectra

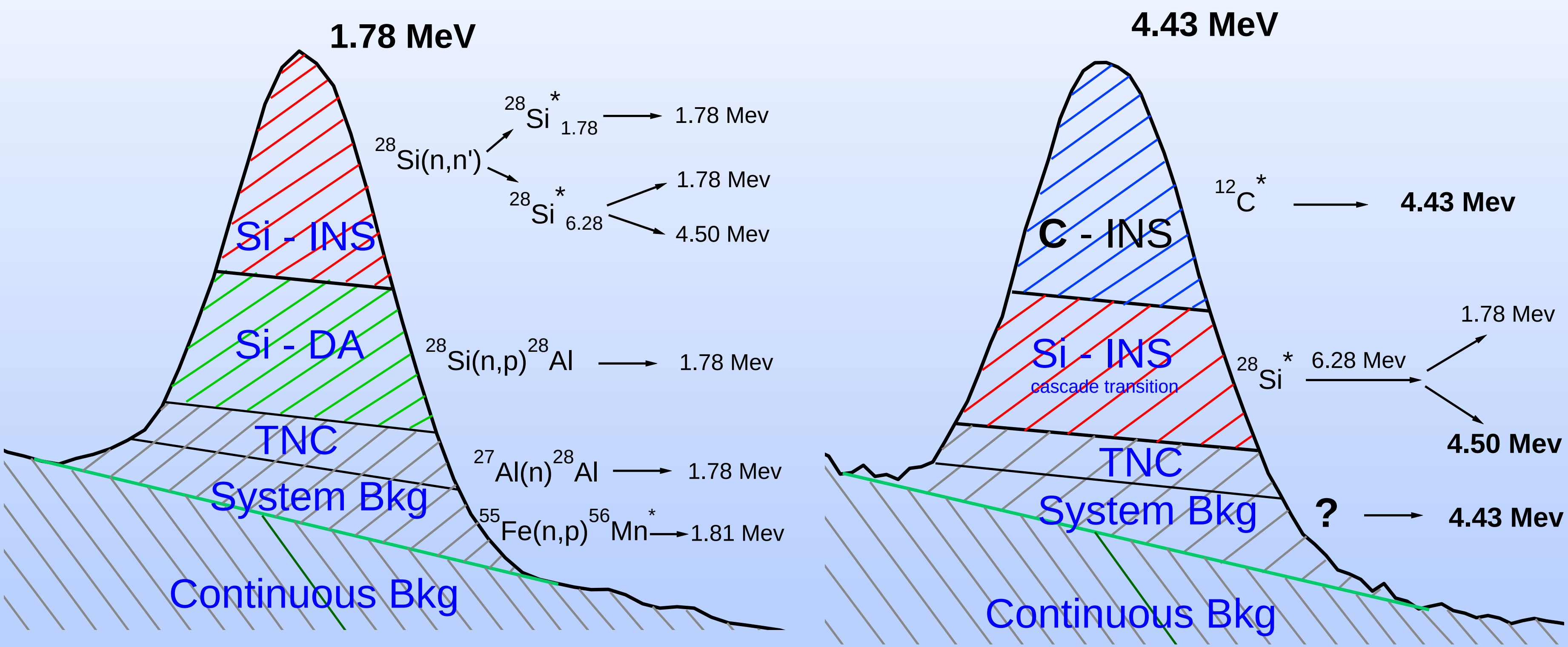


Problems

- Presence of other soil elements
  - **Separating** the carbon peak area at 4.43 MeV from other gamma rays

The **separation** carbon peak area at 4.43 MeV

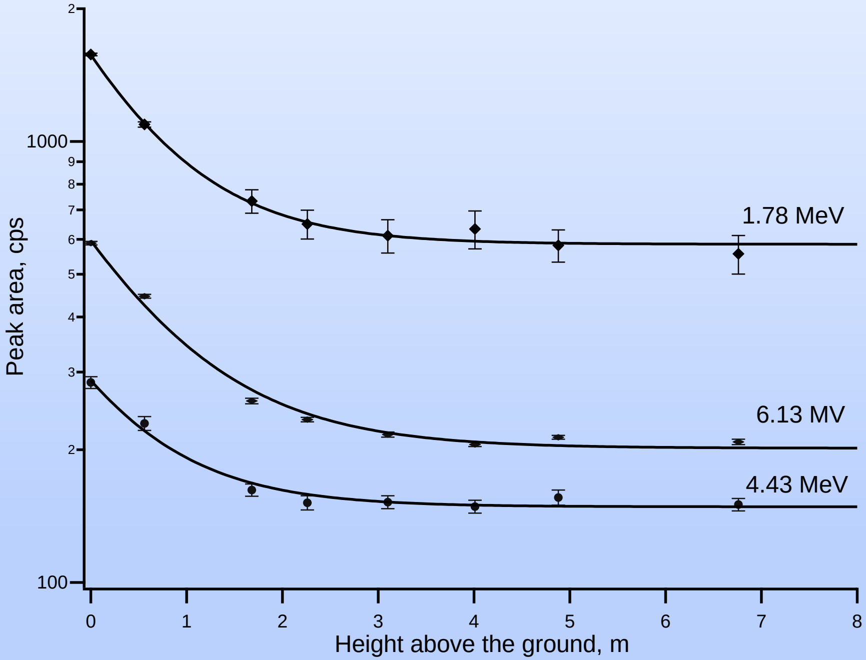
Peak Compositions in raw INS+TNC spectrum



- ✓ The main peak of interest at 4.43 MeV contains **interference from the silicon gamma line**
- ✓ This interference can be taken into account through correlation with the 1.78 MeV peak
- ✓ Gamma rays from **TNC** processes are present in both peaks
- ✓ **System background** is present in both peaks

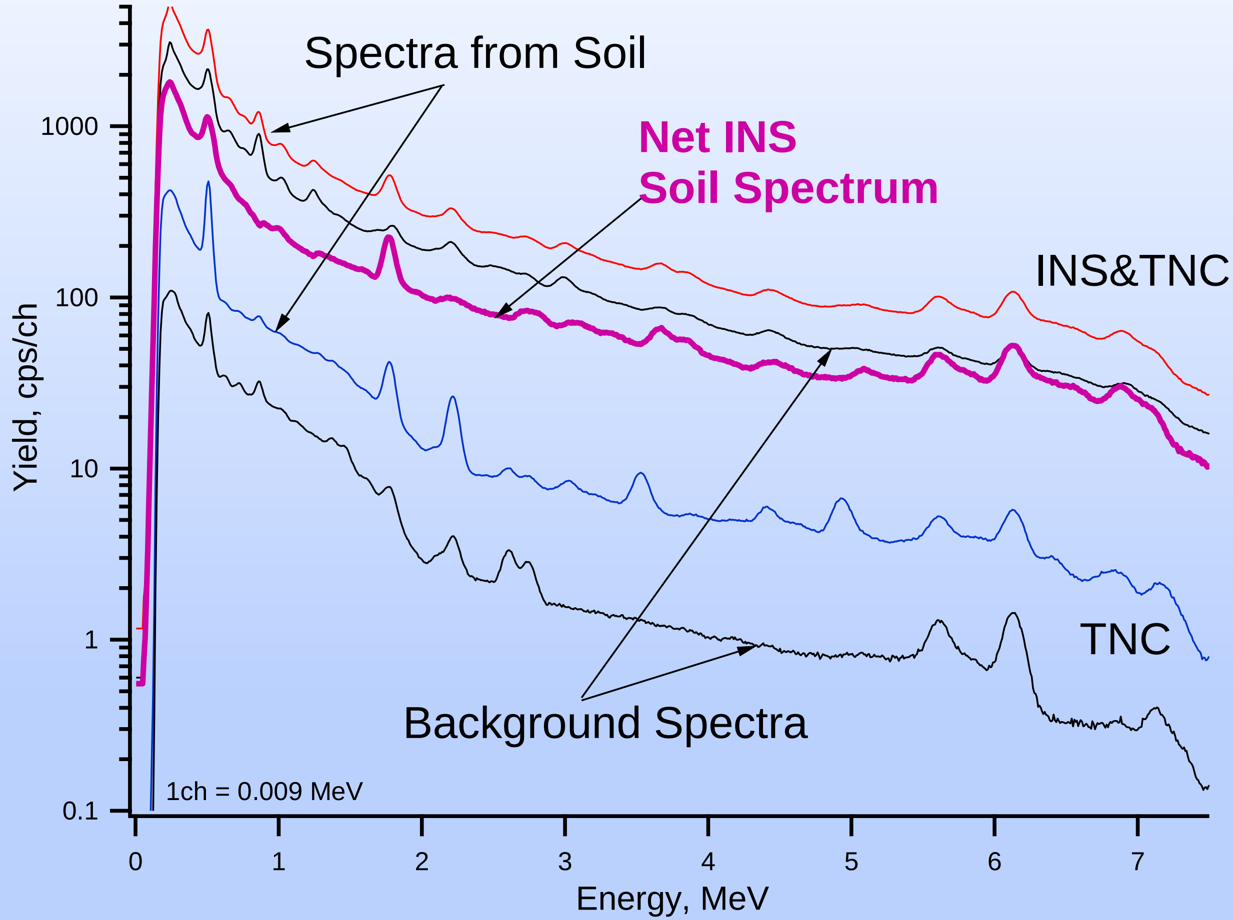
Background Measurement

Peak Areas vs Height in Net INS spectra



Effects of neutron interactions with soil are **negligible at heights > 4 m**

Net INS spectrum



Net INS Spectrum =

$$\text{Net-INS}_{Soil} - \text{Net-INS}_{Bkg} = (\text{INS\&TNC} - \text{TNC})_{Soil} - (\text{INS\&TNC} - \text{TNC})_{Bkg}$$

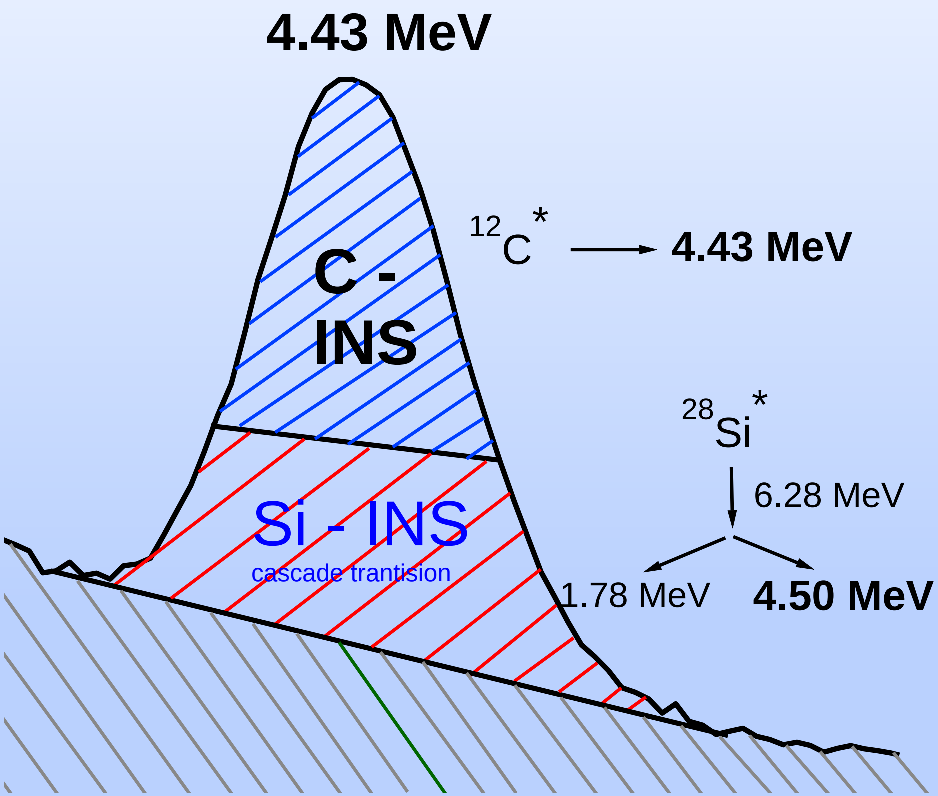
The **separation** carbon peak area at 4.43 MeV

Peak Compositions

The cascade transition of excited Si at 6.23 MeV to ground state goes through the 4.50 MeV and 1.78 MeV levels

$^{16}O(n,n`\alpha)^{12}C^{*}$ - ???

$^{14}N(n,t)^{12}C^{*}$ - ???

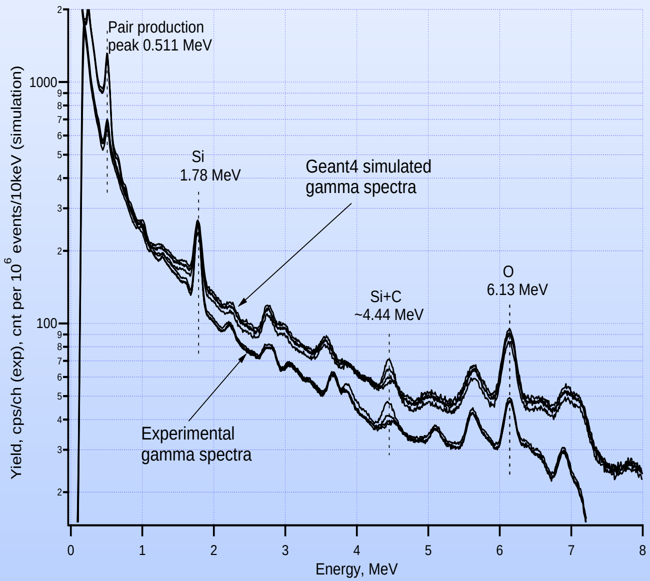


- ✓ The main peak of interest at 4.43 MeV contains **interference from the silicon gamma line**
- ✓ This interference can be taken into account through correlation with the 1.78 MeV peak

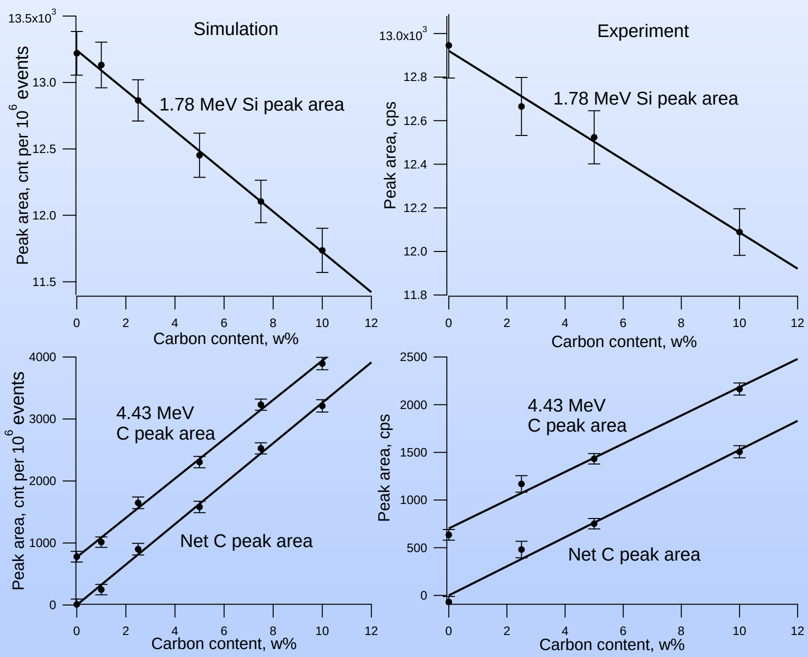
The **separation** carbon peak area at 4.43 MeV

Extraction of the carbon signal from peak with centroid at 4.43 MeV

Net INS spectra



Silicon and Carbon Peak areas vs Carbon content



Reference pits: 150 cm x 150 cm x 60 cm^3
sand + carbon homogeneous mixture;
Carbon content 0-10w%

f_1 = 0.058 **f_2 = 0.054**

Net C peak $area_i$ = 4.43 MeV C peak $area_i$ - $f_i \cdot$ 1.78 MeV Si peak $area_i$

i=1 (simulation), 2 (experiment)

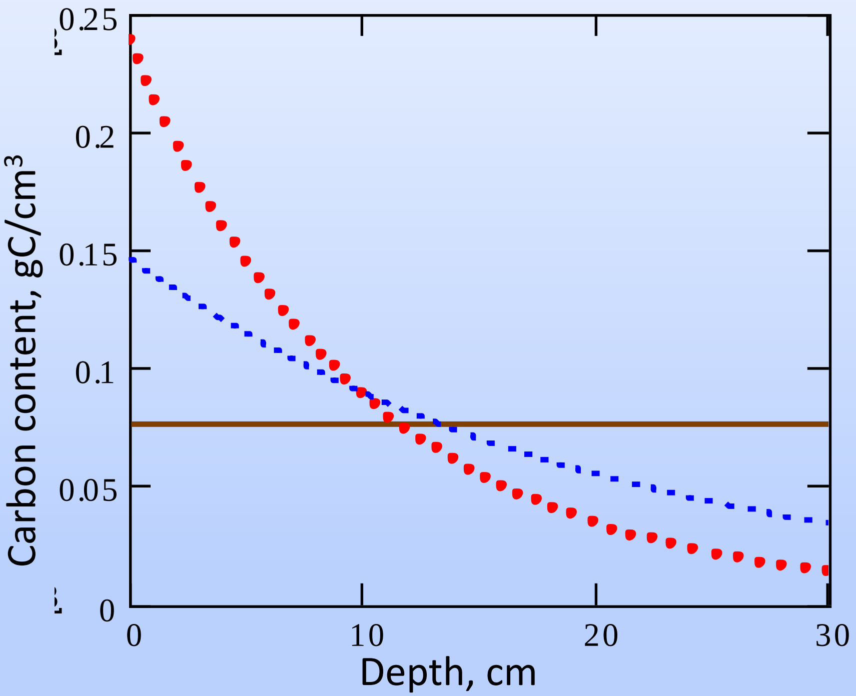
Si cascade transition coefficient f = 0.0547 (Herman et. al., 2007)

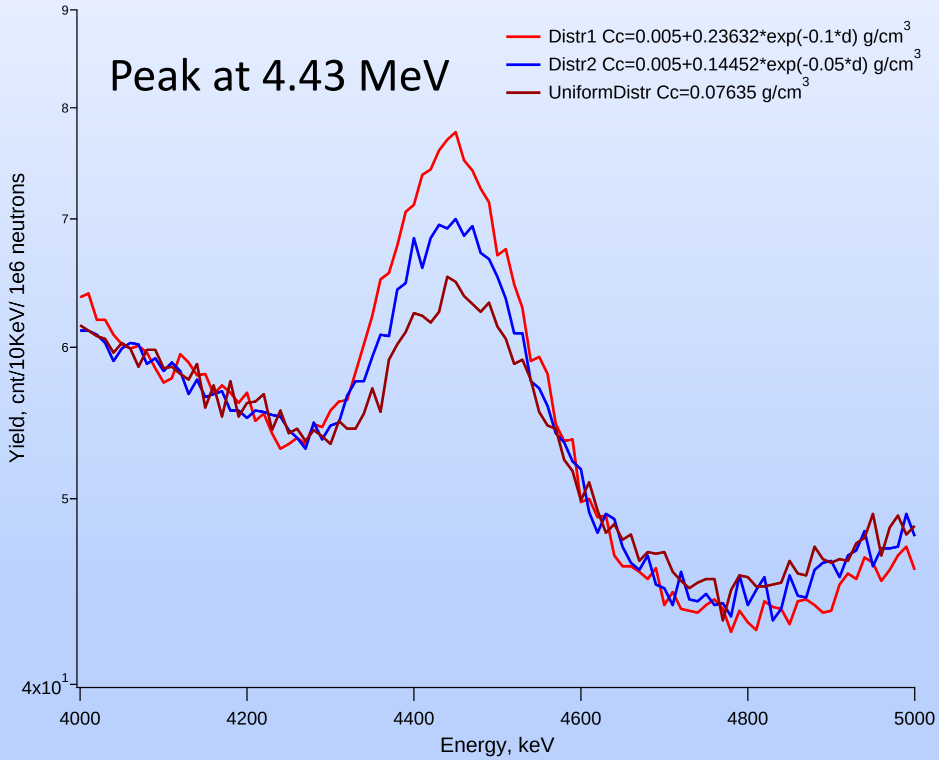
Problems

- Effects of soil carbon distribution
  - Attributing the INS signal to **certain** soil carbon **characteristics**

Monte-Carlo Simulation
for different carbon distributions

Carbon surface density at 30 cm = const





Net C peak area = 4.43 MeV C peak area - f · 1.78 MeV Si peak area **≠ const**

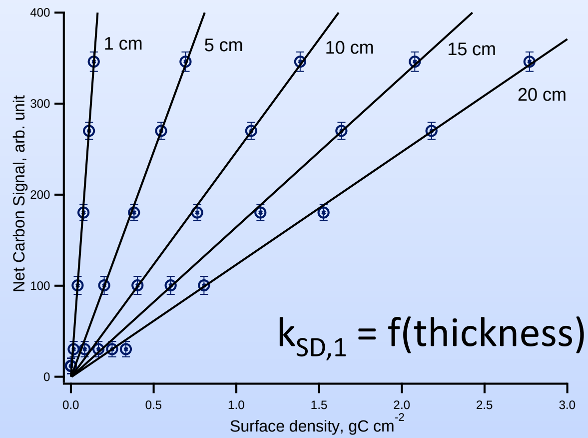
Carbon INS peak area can't characterize the soil carbon content expressed in carbon surface density at 30 cm

Calibration of INS signal vs. different soil characteristics for homogeneous carbon distribution

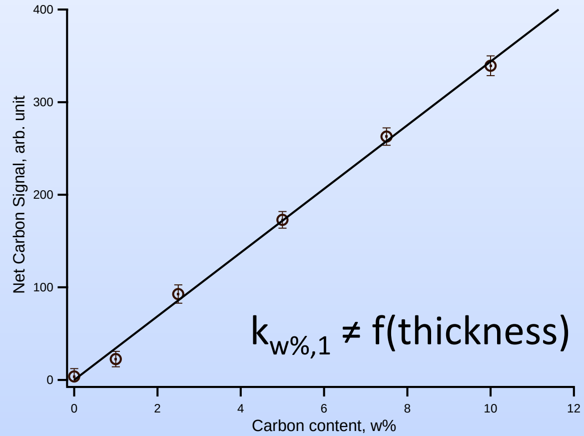
Net Carbon Peak Area = k · Carbon content

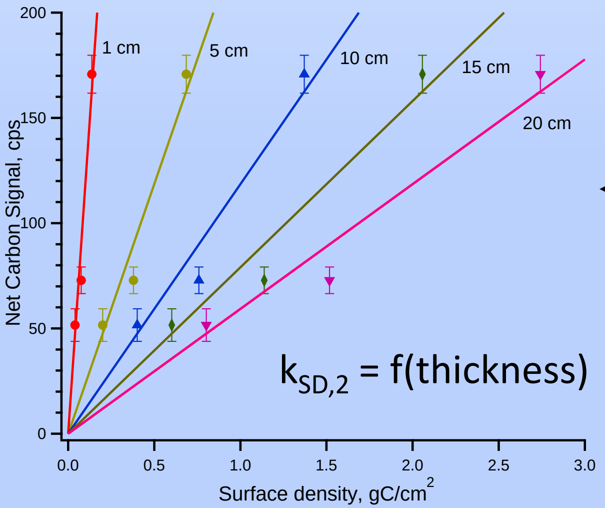
vs. Surface Density in the 1-20 cm soil layer, **gC/cm^2**

vs. average Carbon Weight % in soil layer, **Cw%**

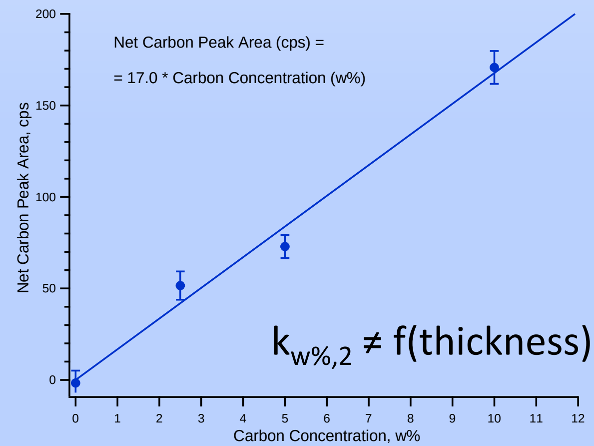


← simulation →





← experiment →

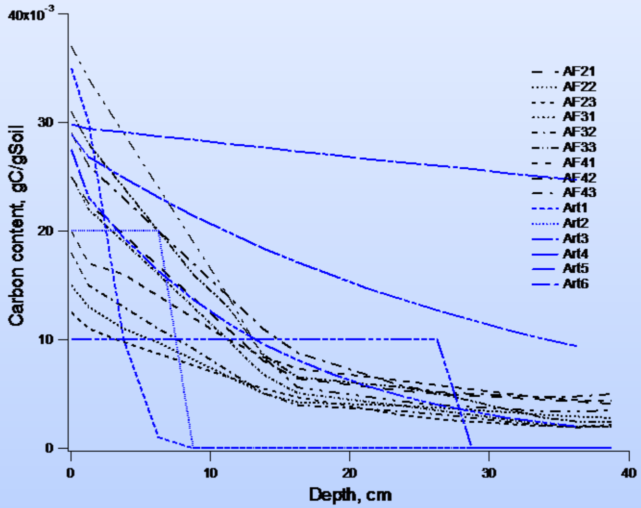


Which soil characterization parameter reflects the INS signal from soil with an unknown carbon depth distribution shape ?

MC Simulation and Field INS measurements
of soil with non-uniform carbon distribution

Carbon Distribution Field Data

from dry combustion analysis and artificial



$AvgCw\%(th)_{DC}$ = f(Thickness)

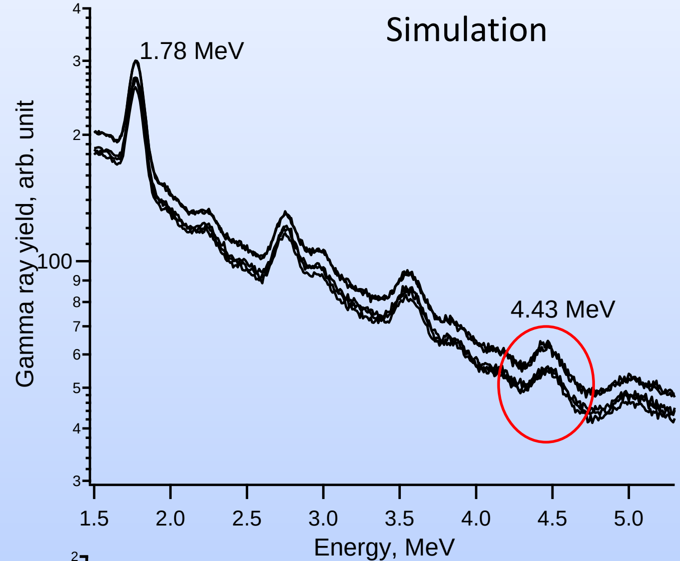
$SD(th)_{DC}$ = f(Thickness)

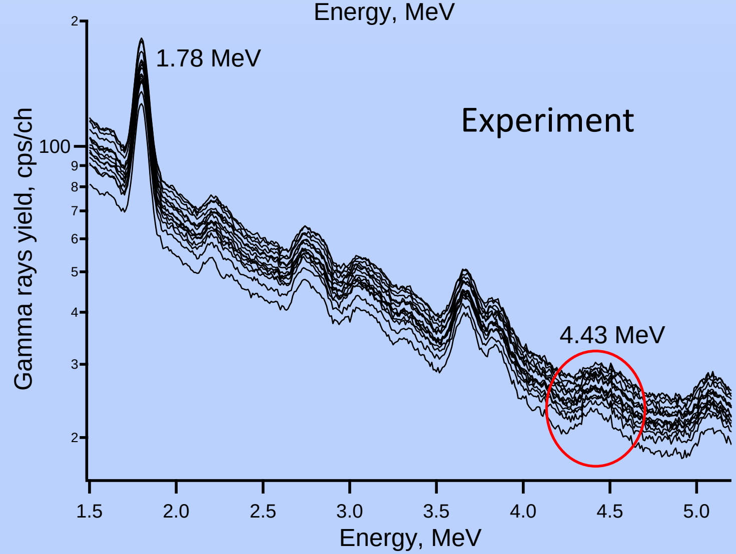
$Cw\%_{INS,i}$ = Net C Peak $Area_i$/ $k_{w\%,i}$

$SD(th)_{INS,i}$ = Net C Peak $Area_i$/ $k_{SD,i}(th)$

i=1 (MC simulation), 2(experiment)

Net INS spectra

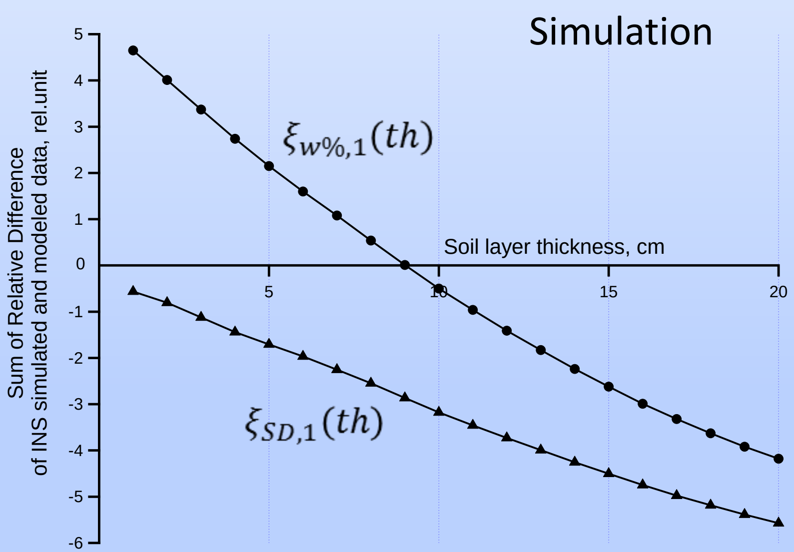


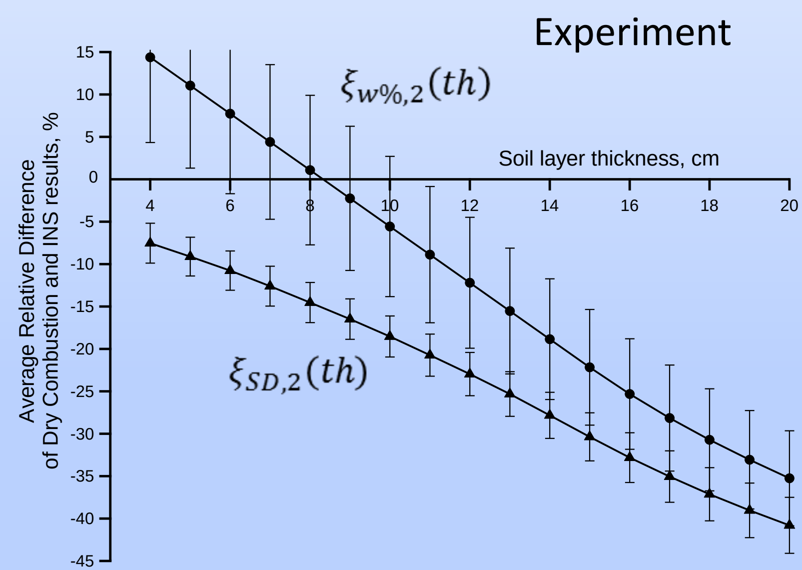


Comparison of INS and carbon depth profile distribution data

$$\xi_{w\%,i}(th) = \frac{1}{N}\sum_{n}\frac{AvgCw\%(th)_{DC} - Cw\%_{INS,i}}{Cw\%_{INS,i}} \cdot 100\% \;\; ; \qquad \xi_{SD,i}(th) = \frac{1}{N}\sum_{n}\frac{\left[SD(th)_{DC} - SD(th)_{INS,i}\right]}{SD(th)_{INS,i}}$$

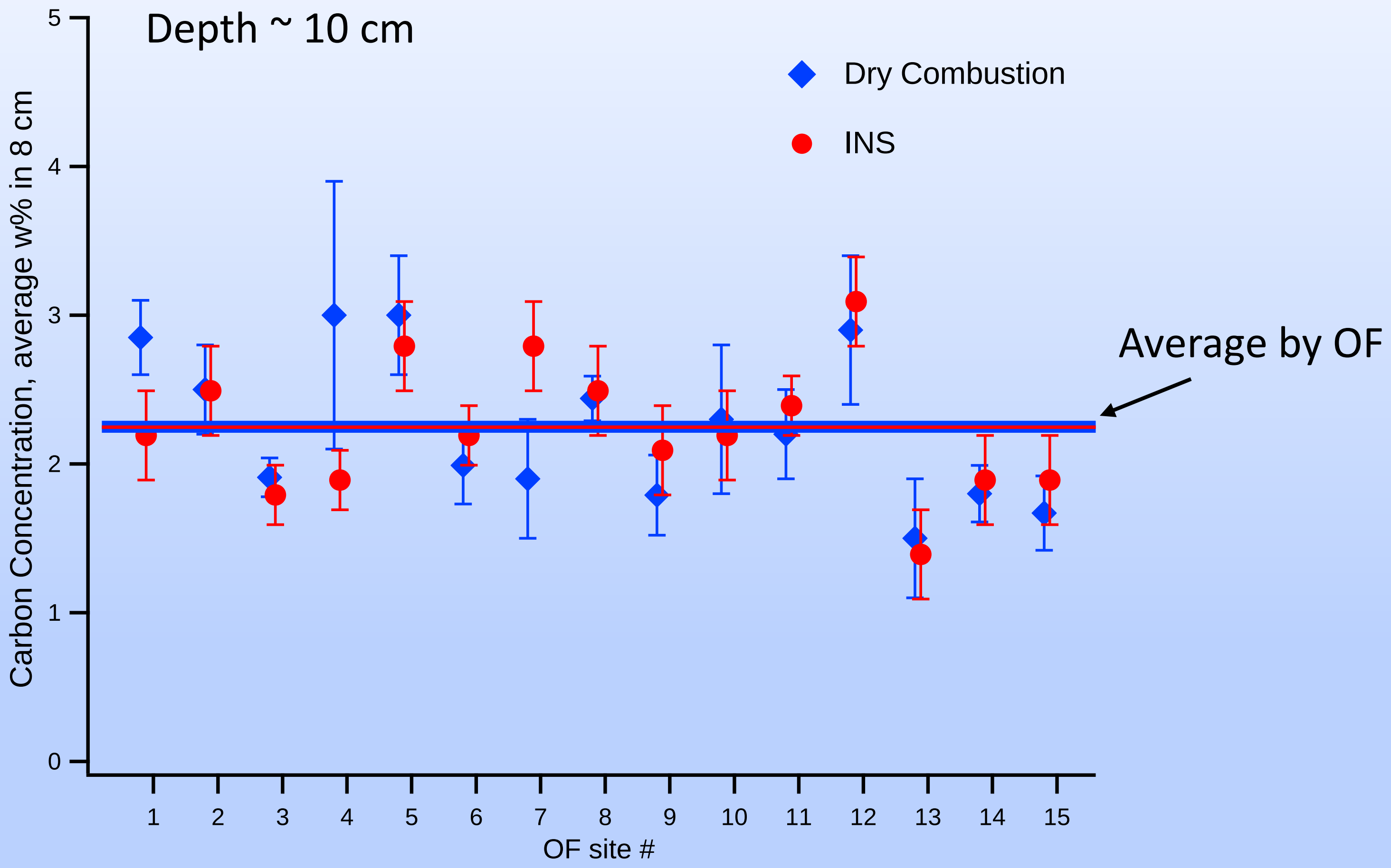
i=1 (MC simulation), 2(experiment)





The value of carbon weight percent coincides with the average weight percent **at thickness ~10 cm** for **any** carbon distribution

INS Field Measurements Compared to Dry Combustion



Experimental results demonstrate that the INS **signal** can be attributed to the **average carbon soil content in ~10 cm upper soil layer**

Comparison of INS and Dry Combustion

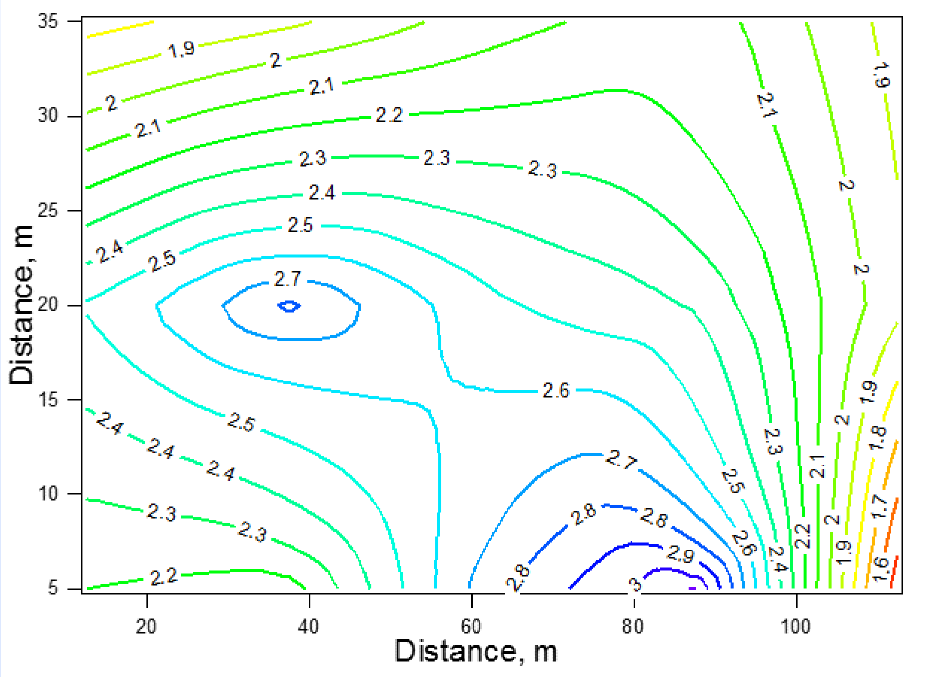
Location	Site # or Plot #	MINS measurements			Dry Combustion measurements		
		Carbon, w%	STD, w%	Plot Average ±STD, w%	Carbon, w%	STD, w%	Plot Average ±STD, w%
Camp Hill Open Field	OF1	2.20	0.29	**2.23±0.45**	2.85	0.25	**2.25±0.51**
	OF2	2.51	0.29		2.54	0.31	
	OF3	1.76	0.22		1.91	0.13	
	OF4	1.88	0.23		2.99	0.94	
	OF5	2.82	0.25		3.03	0.37	
	OF6	2.15	0.21		1.99	0.26	
	OF7	2.77	0.32		1.92	0.41	
	OF8	2.52	0.25		2.44	0.15	
	OF9	2.06	0.26		1.79	0.27	
	OF10	2.17	0.27		2.25	0.45	
	OF11	2.39	0.22		2.23	0.30	
	OF12	3.11	0.31		2.91	0.47	
	OF13	1.44	0.25		1.49	0.42	
	OF14	1.93	0.29		1.80	0.19	
	OF15	1.86	0.27		1.67	0.25	
Camp Hill Applied Field 2	AF2-1	1.22	0.38	**1.59±0.45**	2.00	0.34	**1.48±0.46**
	AF2-2	2.09	0.37		1.14	0.34	
	AF2-3	1.46	0.37		1.31	0.08	
Camp Hill Applied Field 3	AF3-1	1.44	0.43	**1.77±0.37**	1.96	0.34	**1.90±0.53**
	AF3-2	1.68	0.37		1.34	0.34	
	AF3-3	2.17	0.39		2.4	0. 8	
Camp Hill Applied Field 4	AF4-1	2.59	0.42	**2.33±0.34**	1.58	0.34	**2.12±0.46**
	AF4-2	2.47	0.37		2.35	0.34	
	AF4-3	1.94	0.45		2.42	0.14	

Experimental results demonstrate that the INS **signal** can be assign to the **average carbon soil content in ~10 cm upper soil layer**

Comparison of INS and Dry Combustion

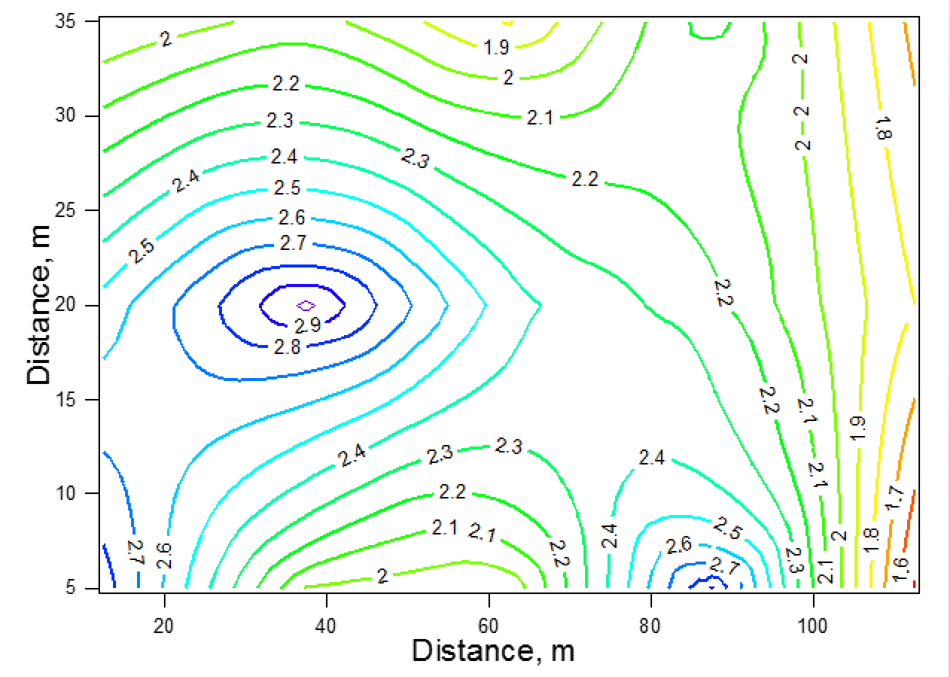
Mapping of carbon content

INS system



Working Time = 2 days

Dry combustion



Working Time = 2 months

Maps plotted using INS and Dry combustion data are similar

Conclusions for Carbon Measurements

- A reliable mobile **system based on the Pulsed Fast/Thermal Neutron-Gamma method** for soil carbon measurements was **developed** and **constructed**:

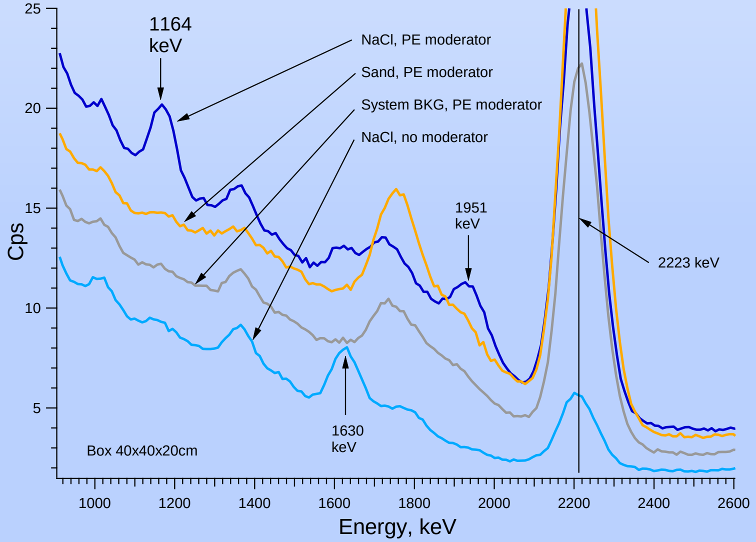
- The procedure for **net carbon signal** extraction was defined
- The **net carbon signal** is **directly proportional** to the average carbon weight percent in the **~10 cm** upper **soil layer** for **any** carbon distribution
- The **duration** of one INS measurement is **30-60 minutes**
- The **minimal** measurement level of soil carbon content was estimated to be ~ **1.0 w%** with standard deviation within **± 0.5 w%**

Chlorine measurements

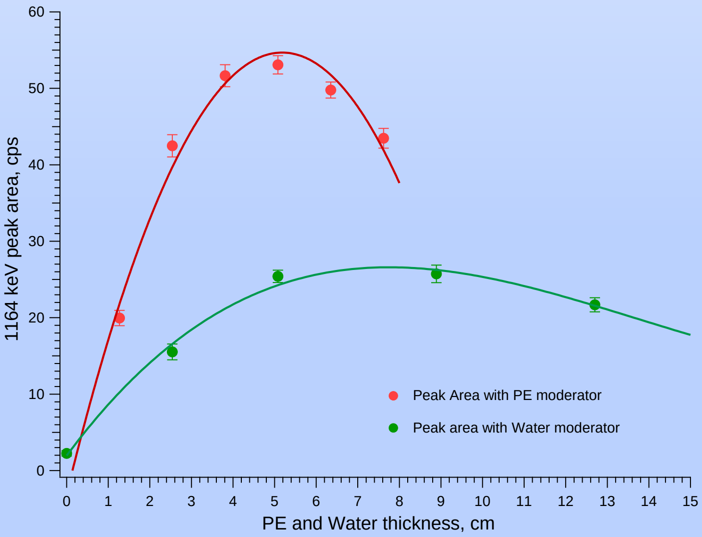
- Sources of chlorine contamination: Chlorinated hydrocarbons, components of explosive ordinance (trinitrochlorobenzene, ammonium perchlorate, and tetraaminecopper perchlorate)
- The ability to detect chlorine contamination or chlorine-containing objects buried under soil quickly and remotely is desirable.
- Cl-35 TNC -> 1.16 MeV-> 43 b
- The field system for carbon measurement with an additional moderator is suitable for chlorine measurement

Moderator Selection

TNC spectra with and w/o moderator

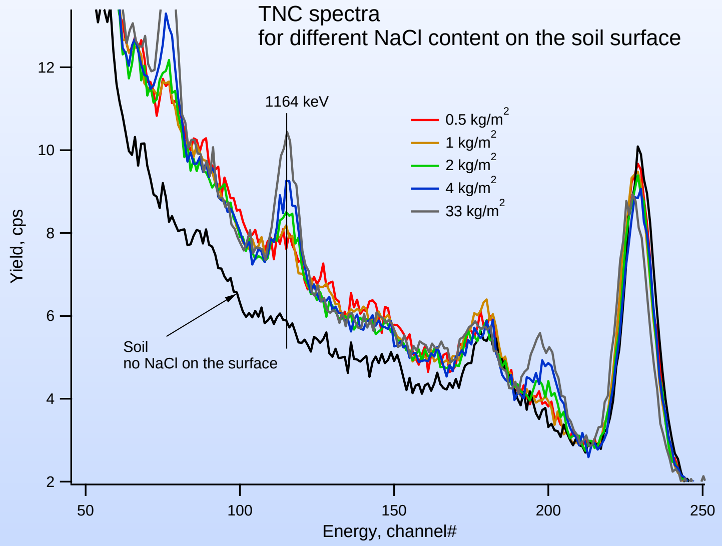


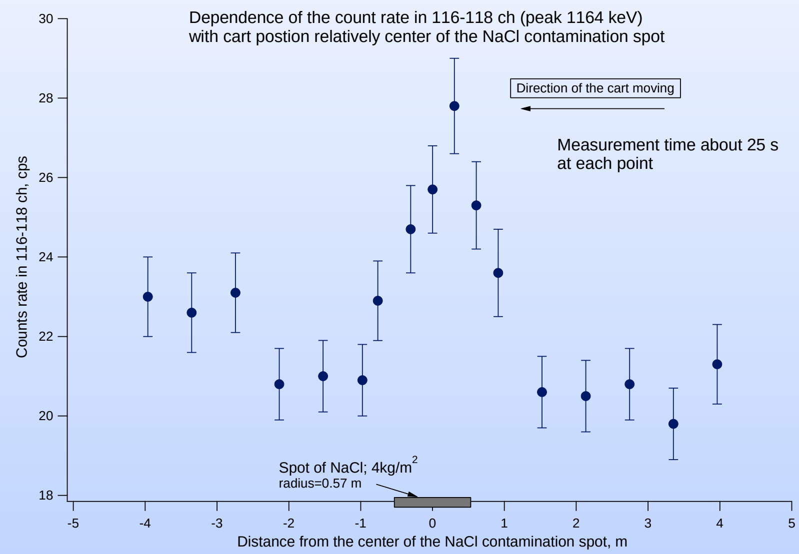
Dependences of the 1.16 MeV peak area vs moderator thickness



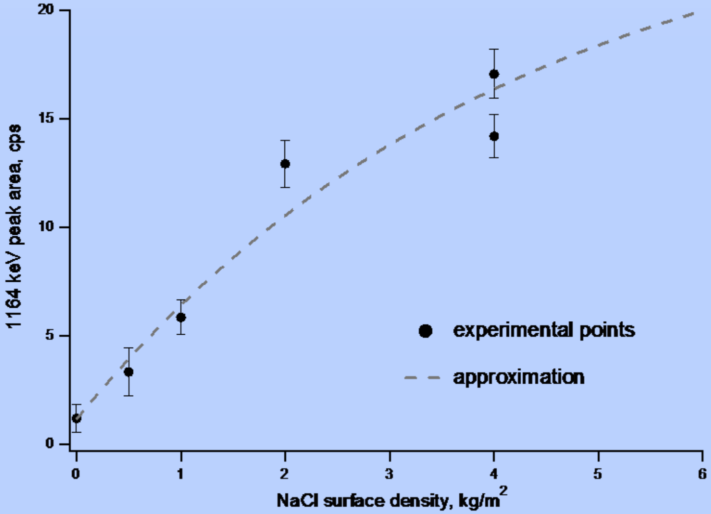
Polyethylene moderator with thickness of 5 cm was chosen for further measurements

Chloride Surface Contamination Measurements





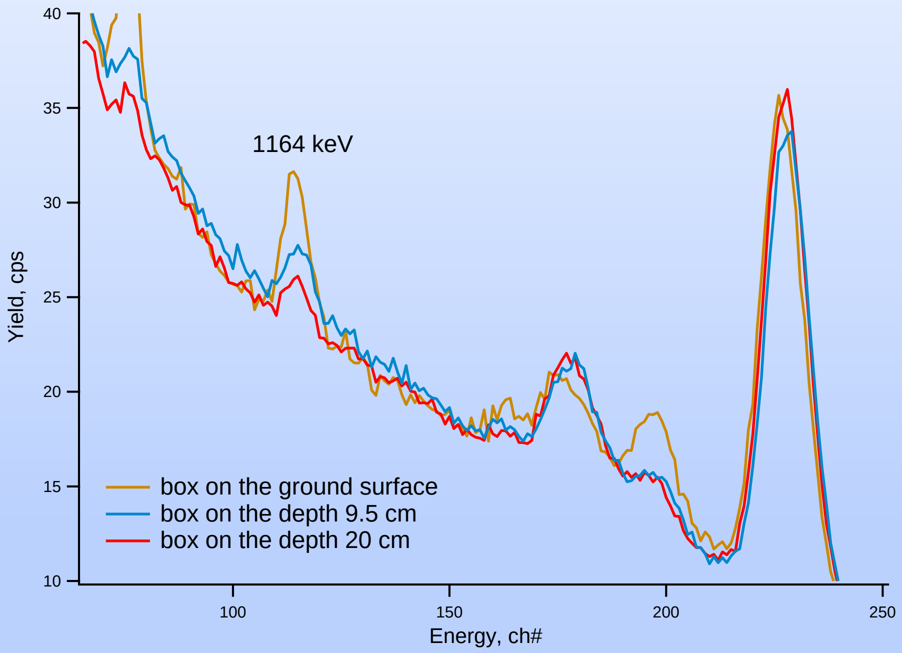
Dependences of the 1.16 MeV peak area vs NaCl surface density



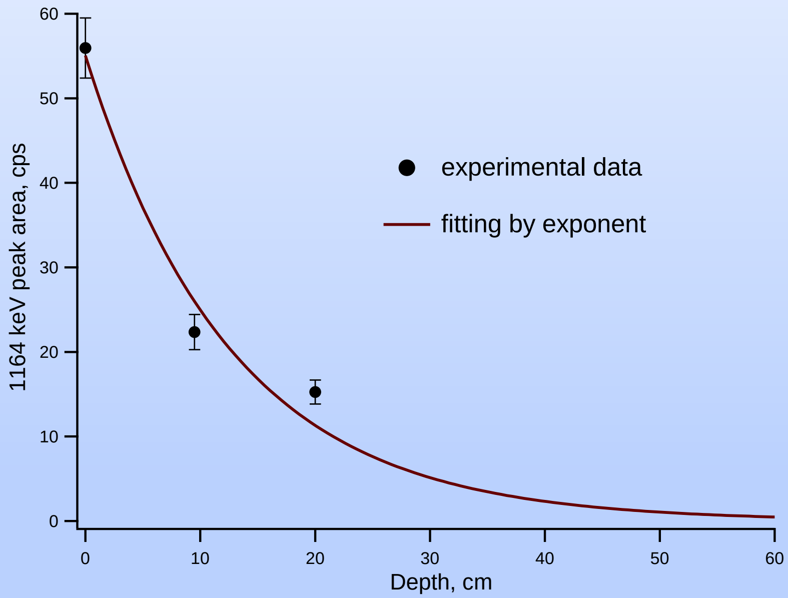
The current mobile system can determine a contamination spot at levels of several tenths of a kilogram chlorine per square meter in the “pseudo”-scanning regime

Buried Chloride Object Measurements

TNC spectra of the box with 6.5 kg NaCl
with size 34x27x7 cm^3



Dependences of the 1.16 MeV peak area
vs Depth to Buried Object

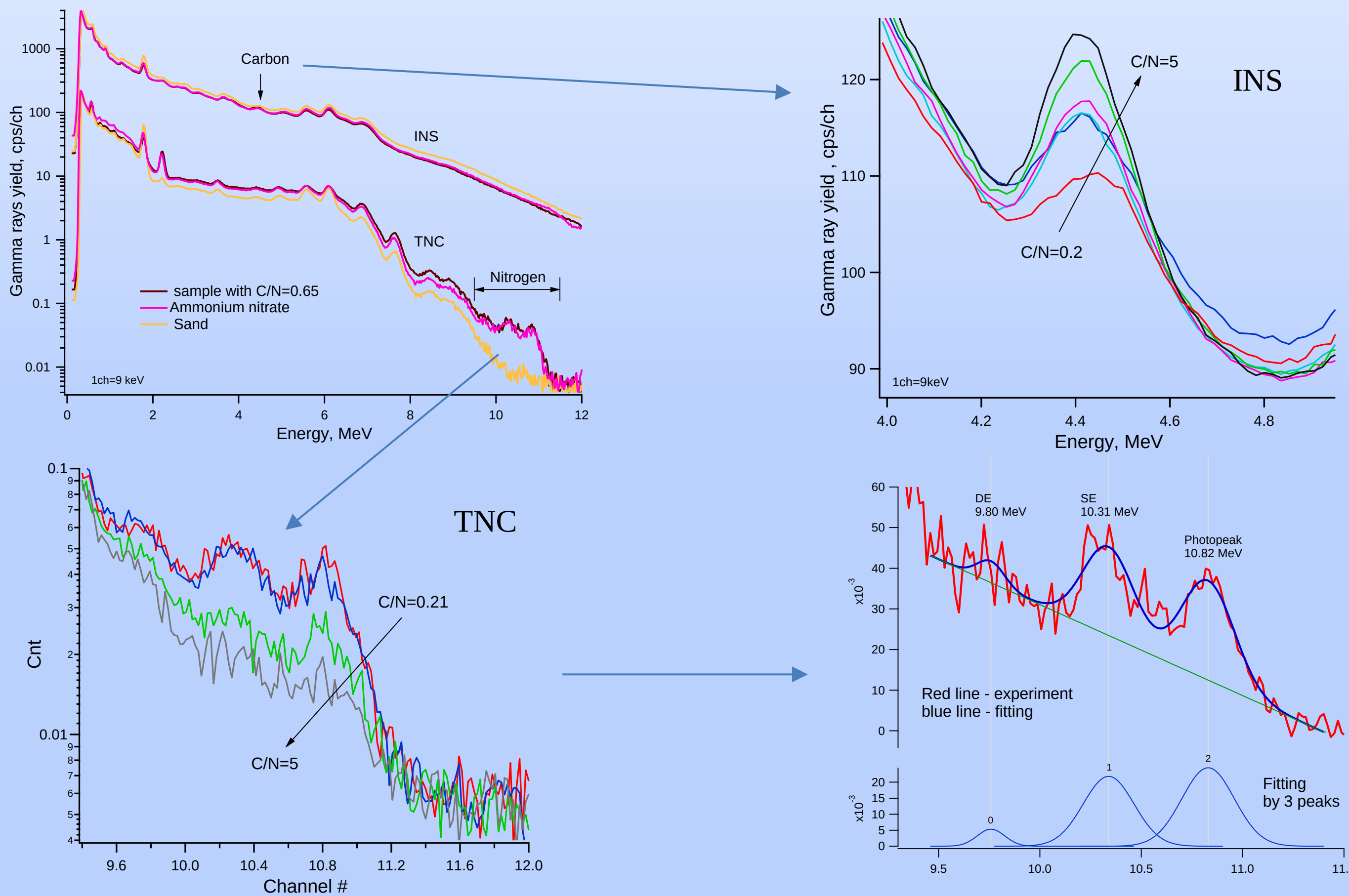


The current mobile system can detect objects with several kilograms of chloride buried at a depth of ~30-40 cm under the soil surface

Nitrogen Measurements
by Pulsed Fast/Thermal Neutron-Gamma Method

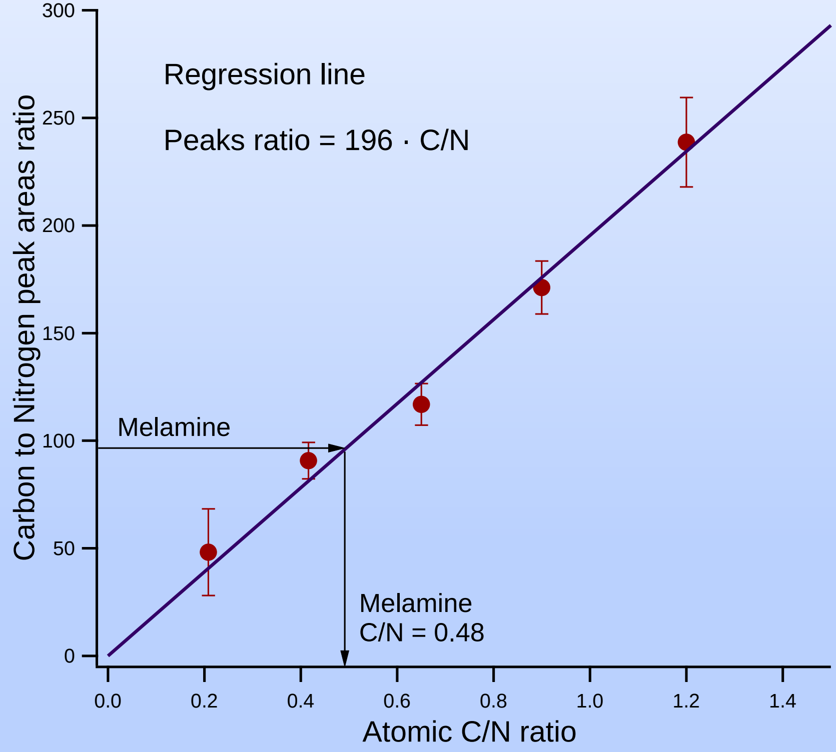
- The non-invasive determination of the C/N atomic or mass ratio is used for detecting explosive materials in studied objects
- C-12 INS -> 4.43 MeV -> 0.42 b;   N-14 TNC -> 10.82 MeV-> 0.08 b
- Field system for carbon measurement is suitable for C/N measurement
- Samples m=30 kg; Volume = 40x40x20 cm^3; Atomic C/N=0.2 – 1.2; Density = 0.96 – 0.81 g/cm^3

Gamma spectra of carbon-nitrogen mixtures with different C/N ratios



C/N measurements

The ratio of the carbon (INS) to nitrogen (TNC) peaks
vs C/N atomic ratio in samples



The accuracy of C/N ratio determination is ± 0.03

(for comparison, the accuracy of C/N ratio determination in Mitra (2012) is ± 0.25)

Conclusions

- The procedures determining soil carbon content by the Pulsed Fast/Thermal Neutron-Gamma Method were developed;
- This method can define the soil carbon content in the upper soil layer much faster than the traditional dry combustion method;
- This method can be used to detect chlorine contamination or chlorine-containing objects buried under soil quickly and remotely;
- The accuracy of the C/N ratio determination of dangerous objects is several times better than other previously described variants of neutron-gamma analysis.

Future Work

- Optimize the Pulsed Fast/Thermal Neutron System design to **improve sensitivity** and **accuracy**
  - Number of the detectors
  - Shielding
  - Geometry
- Testing the workability of the system in the **scanning regime**

List of Publications

- Poster at this Conference: Monte-Carlo Gamma Response Simulation on Fast/Thermal Neutron Interactions with Soil Elements. 2016. Kavetskiy A., G. Yakubova, H.A. Torbert, and S.A. Prior.
- Yakubova G., L. Wielopolski, A. Kavetskiy, H.A. Torbert, and S.A. Prior. 2014. Field Testing a Mobile Inelastic Neutron Scattering System to Measure Soil Carbon. Soil Sci. 179:529-535.
- Kavetskiy A., G. Yakubova, H.A. Torbert, and S.A. Prior. 2015. Continuous versus pulse neutron induced gamma spectroscopy for soil carbon analysis. Appl. Radiat. Isotopes 96:139-147.
- Yakubova, G., A. Kavetskiy, S.A. Prior, and H.A Torbert. 2015. Benchmarking the inelastic neutron scattering soil carbon method. Vadose Zone J., doi:10.2136/vzj2015.04.0056.
- Kavetskiy A., G. Yakubova, S.A. Prior, and H.A Torbert. 2016. "Hot background" of the mobile inelastic neutron scattering system for soil carbon analysis. Appl. Radiat. Isotopes 106:299-311.
- Yakubova, G., A. Kavetskiy, S.A. Prior, and H.A Torbert. 2016. Application of Geant4 simulation for analysis of soil carbon inelastic neutron scattering measurements. Appl. Radiat. Isotopes 113:33-39.

Thank you!

Please, any questions?

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