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Sub soil measurements in Sweden of radioxenon and radioargon

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POSTER

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ABSTRACT

The most important indicators for an underground nuclear explosion during a CTBT on-site inspection are the radioactive xenon isotopes ^{131m}Xe, ^{133}Xe and ^{133m}Xe and the radioactive argon isotope ^{37}Ar. Knowledge of how these isotopes vary and potentially correlate in different types of soil is essential to be able to discriminate between the natural background and a signal from a nuclear explosion.

A series of measurements has been performed at different depths within a limited area in the region of Kvarntorp (Sweden), a location with known elevated uranium content in the ground. To investigate variations of the naturally occurring noble gas concentration in soil gas over time and at different depths soil gas sample collection was carried out over a period of two weeks. The analytical results from the collected samples will be discussed in relation to radon levels, soil uranium content and environmental parameters such as the meteorological conditions.

Balloons being filled with soil gas extracted using FOI samplers (seen in the middle of the photo). The sampling rate is approximately 6 L/min at a sampling depth of approximately 3 m.

Objectives:

- Build capacity, test and develop methods and equipment
- Study the natural background of radioxenon and radioargon
- Collect data for modeling and better understanding
- Multiple samples from the same location
- Gas collection at different depths

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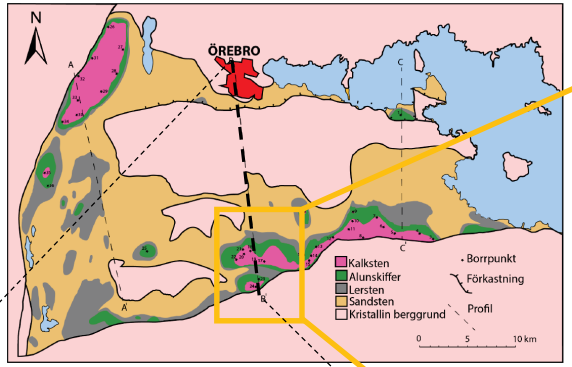
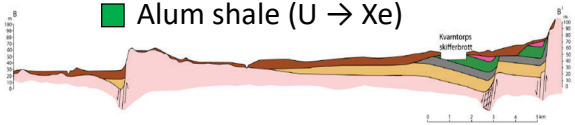
INTRODUCTION

Sampling sites 2016-2019

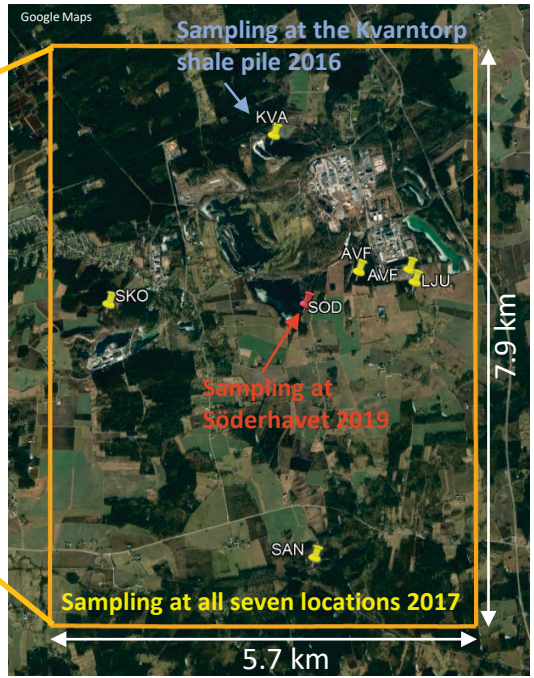


Limestone (Ca → Ar)

Alum shale (U → Xe)



Andersson et al., 1985: The Scandinavium Alum Shales. *Sveriges Geologiska Undersökning (SGU) Ca 56*, 1-49



Soil gas sampling in the region of Kvarntorp (Sweden), an area with known elevated uranium content in the ground. This region was selected to increase the possibility to detect radioxenon in the ground. Three sampling campaigns have been carried out during 2016-2019. The pictures illustrates the alum shale and limestone profiles in the area and the sampling locations.

Sampling campaign 2016:
https://doi.org/10.1016/j.jenvrad.2018.12.006
Sampling campaign 2017:
https://doi.org/10.1016/j.jenvrad.2020.106458

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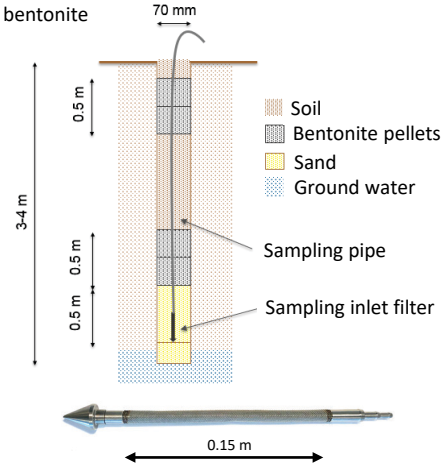
METHODS

Sampling sites and hole preparation

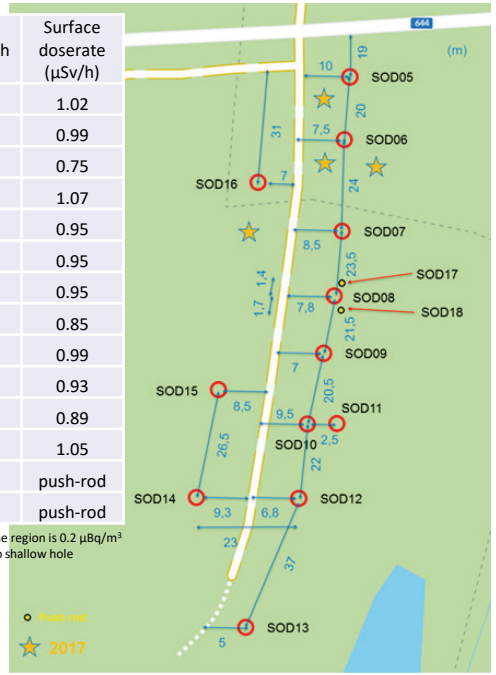
In total 12 holes were augered within 5 hours to a depth of approximately 3-4 m, limited by the ground water. At a later stage two additional sampling sites were setup using push rods to a depth of approximately 0.9 m, see the map to the right. Soil samples were extracted during augering at each meter.

The holes were first backfilled with sand above the water table. Then a sampling inlet connected to a 6 mm tube was inserted. A 0.5 m thick layer of sand was poured around the sampling inlet and thereafter the hole was sealed using the excavated soil and two plugs of bentonite granulates. The bentonite pellets were first poured in the hole and thereafter water was added to make the bentonite expand and seal the hole.



Sampling site	Depth (m)	Surface doserate (µSv/h)
SOD05	3	1.02
SOD06	3	0.99
SOD07	3	0.75
SOD08	3	1.07
SOD09	3	0.95
SOD10	3	0.95
SOD11*	1.5	0.95
SOD12	3	0.85
SOD13	2.5	0.99
SOD14	2.5	0.93
SOD15	3	0.89
SOD16	3	1.05
SOD17	0.9	push-rod
SOD18	0.9	push-rod

Normal background in the region is 0.2 µBq/m³
* Not sampled due to too shallow hole



The site spacing was selected to minimize effects from sampling simultaneously from nearby sites. The exception are the two push rods (SOD17 and SOD18) which where positions < 2 from site SOD08 to investigate possible disturbances.

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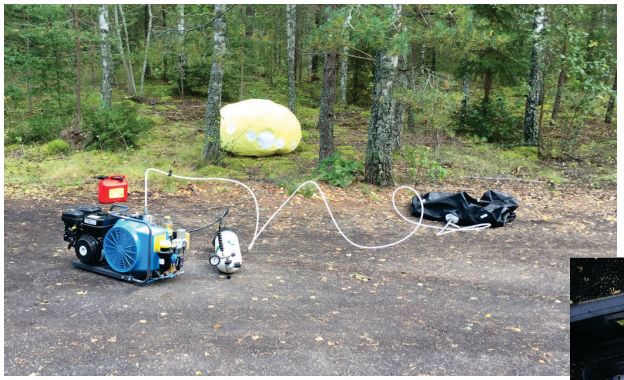
METHODS

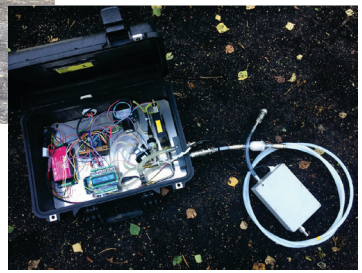
Sample collection



The soil gas was sampled into balloons (typically at 6 l/min). The sample sizes ranged from 1 to 5 m^3, but most samples were 2-3 m^3. Sampling was carried out during day time only. No extra traps for removal of CO_2 nor water was used. After the collection was completed the gas was compressed into scuba bottles and transported to the laboratory in Stockholm for analysis.

An in house developed sampler was used to log the sampling flow rate, concentration of CO_2 and O_2 continuously during the sample collection. Radon was measured at discrete points in time using a MARKUS unit (temporarily) interconnected in the gas flow. A first prototype of a radon measurement directly in the gas flow was tested and evaluated.

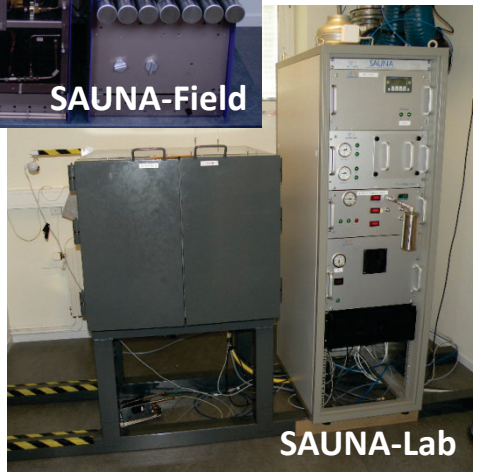
The sample collection was carried out during two weeks in September 2019. Below the FOI sampler is shown.



Lab processing and sample analysis

The soil gas from the scuba bottles was processed in SAUNA-Field and measured either directly or transferred to the SAUNA-Lab system via archive bottles.

A PSA pre-filter was added to SAUNA-Field in order to remove CO_2, hence no extra filters were required (as compared to the sample collection and processing in 2016 and 2017). A few samples were combined for increased sensitivity.



The average delay between sampling and measurement of xenon in the laboratory was 4.5 days, ranging from two up to nine days

Argon samples were “collected” into bags both directly on site and from the drain of SAUNA-Field system. The samples were then shipped to the University of Bern for analysis.

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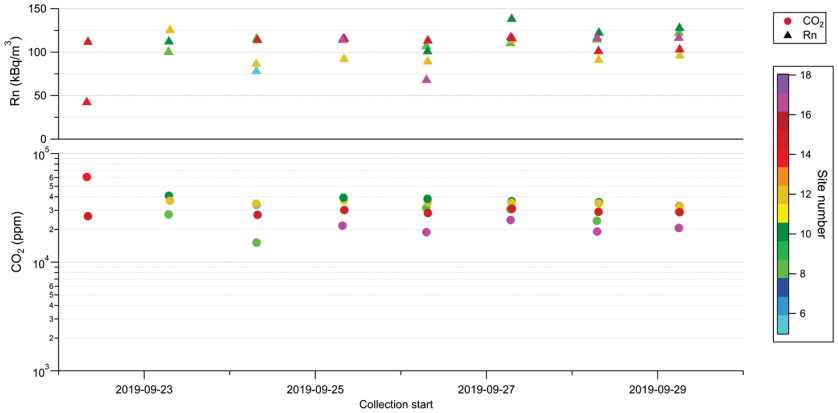
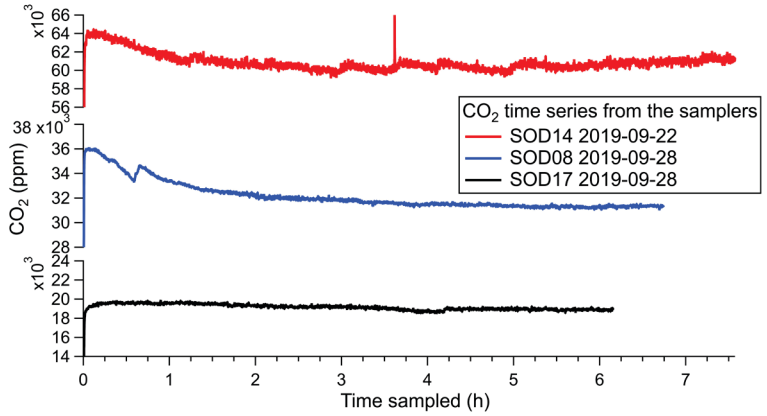
RESULTS

Radon and CO_2 concentrations during sampling



Average CO_2 and radon concentrations during sampling at the different sites and collection dates. Both concentrations were fairly stable for all sites during the sampling period of two weeks. Some sites were sampled for approximately 8 hours almost every day, but allowed to stabilize during the night. Atmospheric CO_2 concentration was approximately 400 ppm, i.e. 25-50 times lower than soil gas concentrations. The atmospheric radon concentration was less than 1 kBq/m^3 (the detection limit of the instrument).



The figure is an illustration of CO_2 concentrations logged during sampling at some different sampling sites. Site SOD17 is installed using a push rod and is located < 2 m from site SOD08 at a depth of 0.9 m. The sampling depth at SOD08 is 3 m. The CO_2 concentrations are from when sampling simultaneously from the push rods as well as the central hole. Stable CO_2 and radon concentrations have been used to indicate that the hole seals are functional.

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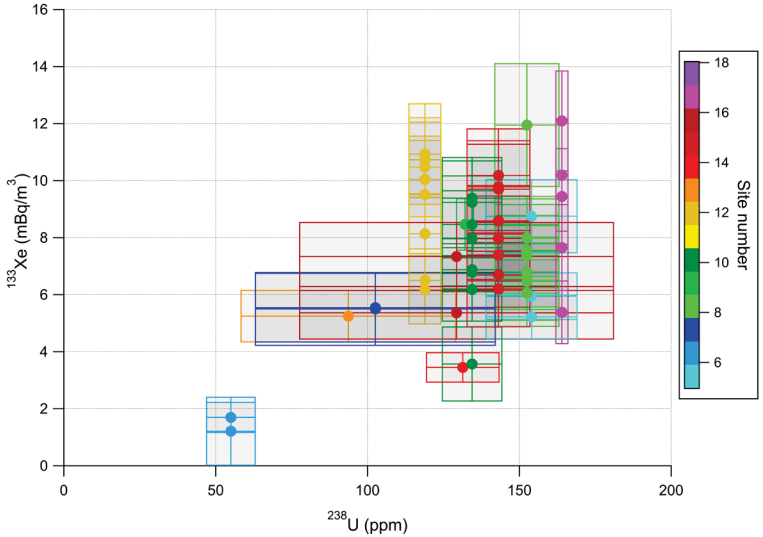
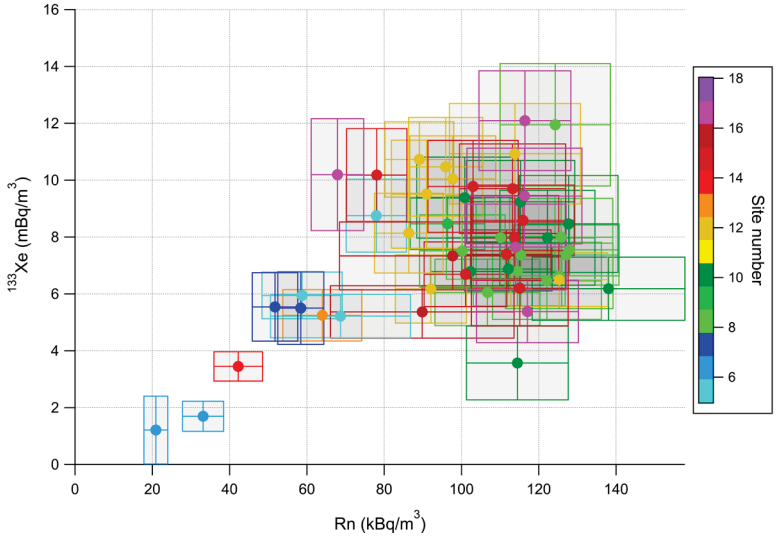
RESULTS

^{133}Xe concentration in relation to uranium content and radon concentration



The average uranium concentration was estimated from soil samples extracted at every meter during hole preparation. The variation at different depths are roughly within 30%, similar to the variation between sites. Only site SOD6 is significantly different which is also seen in measured radon and xenon concentrations.



The radon concentration was measured at discrete time intervals during sampling using an external instrument temporarily interconnected in the gas flow. The results shows no clear trend between radon and xenon concentrations (with the exception of SOD06 which seems to have a different soil composition).

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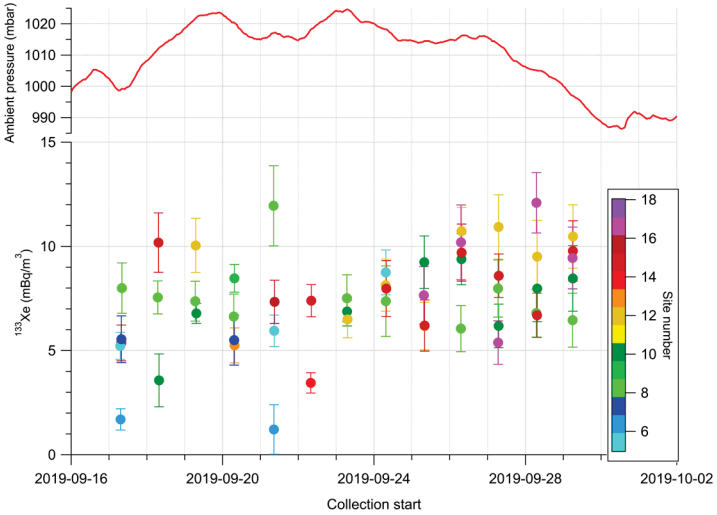
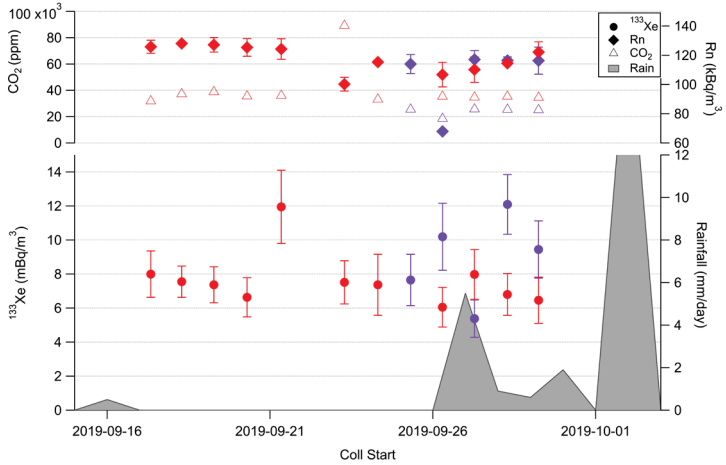
RESULTS

Atmospheric pressure, rainfall and multiple samples at different depths



Measured ^{133}Xe concentrations for the entire sampling period. The atmospheric pressure was measured at a nearby weather station and agrees with on site measurements. No obvious correlation with the xenon concentration and the pressure can be found. The absence of a trend can be due to the soil type and relatively moist ground.



Results for sites SOD08 (red) and SOD17/SOD18 (purple) which were located less than 2 meters apart at sampling depths of 3 and 0.9 m respectively. The sampling in SOD17/18 started a week after the start of sampling at site SOD08. The measured concentrations are fairly stable during the whole sampling period, and the concentrations at the deeper sampling point (SOD08) is fairly unaffected by the withdrawal of equal sized samples 2 m above the sampling point. The rainfall in the end of the sampling period seem to have no effect on the concentrations.

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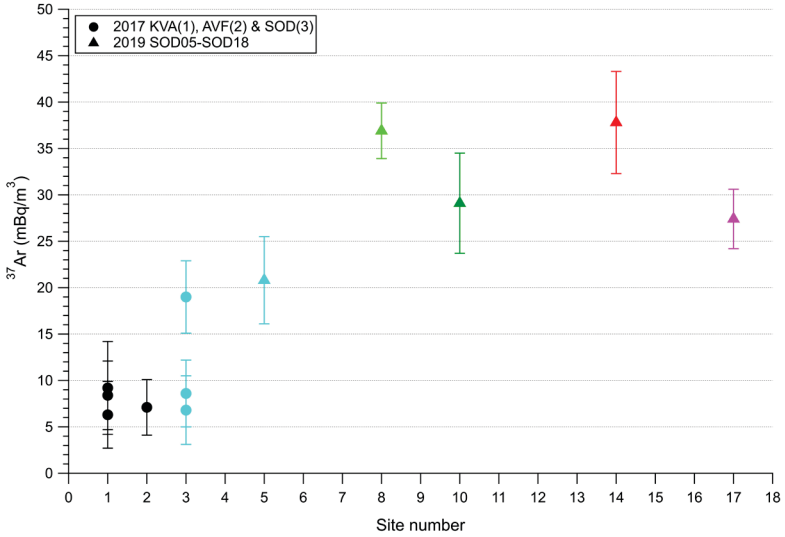
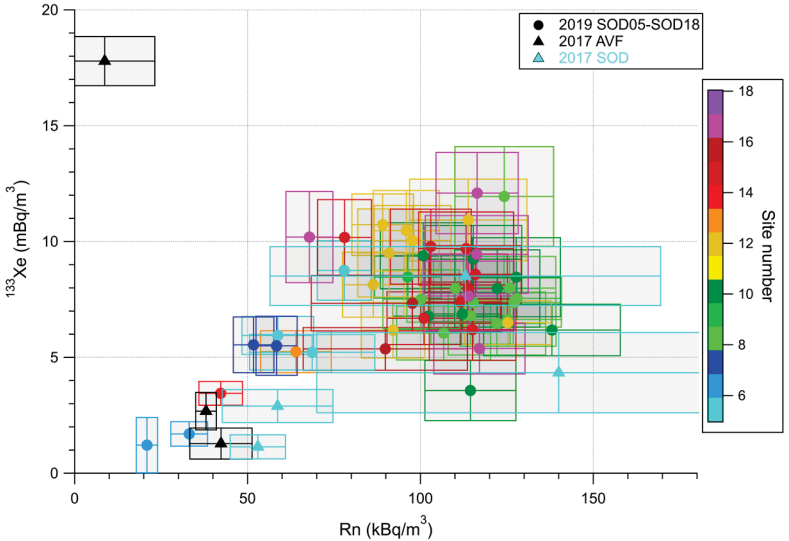
Radioargon detections 2017 and 2019



Results for radioargon analysis 2019 compared to the result from 2017. Note that the samples from 2017 are not only collected at the current site. The sampling point in 2017 (SOD03) were located close to sampling point SOD05. All samples 2017 were extracted at a depth of approximately 0.9 m using push rods.

^{133}Xe comparison 2017 and 2019



Results for radioxenon and radon analysis 2019 compared to the result from 2017. Note that the samples from 2017 are not only collected at the current site. The radioxenon detection at the current site in 2017 were located close to the sampling location SOD05. All samples 2017 were extracted at a depth of approximately 0.9 m using push rods.

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CONCLUSIONS

More than 60 soil gas samples were collected from 13 different sites during a two week period. Radioxenon (^{133}Xe) was detected at all sites and the concentration ranged from 1 to 12 mBq/m^3. Five samples were analyzed for radioargon yielding concentrations between 21 and 38 mBq/m^3 of ^{37}Ar.

Bentonite pellets were used to seal the augured sampling holes. No site showed a degradation of the seal during the sampling period. Gas was collected using an in-house developed sampler.

Only the ^{133}Xe isotope was detected, which is consistent with the difference in production yield and half-life for this isotope, as compared to the xenon isotopes ^{131m}Xe, ^{133m}Xe and ^{135}Xe. The last two isotopes could potentially have been detected if the time between sampling and measurement were reduced to a minimum, i.e. the samples measured directly in the field. To detect ^{131m}Xe, much larger sample volumes would be required due to the lower production yield and lower specific activity.

The uranium concentration in the soil was estimated to be between 40 – 175 ppm. Except for one site there is no clear relationship between estimated uranium concentration and the radioxenon concentration. The radioxenon and radioargon concentrations are fairly stable during the sampling period and equal for all sites and is in agreement with the results from 2017.

The detected concentrations of radioxenon can be seen as an upper limit of what can be expected due to natural occurring uranium in the ground. In 2016 up to 100 mBq/m^3 was detected on a site with very special transport properties, but the concentrations values from 2017 and 2019 are more representative for concentrations for a ground with elevated uranium concentrations.

A photo showing the road along which the samples were collected during a two week period. Two sample balloons are visible between the trees.