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Article

Radon measurements in water for human use in the Sessa Aurunca, Cellole-Caserta district: a cross sectional study

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Abstract. *Background*. 222Radon and fluorine in ground water can be dangerous for human health. Radon is carcinogen of class one, excess of fluorine cause alterations in the process of bone calcification and calcium metabolism. Levels of radon and fluorine in the water should be monitored.
Objective: The aim of the study was to determine the concentration of radon and fluorine in the waters sources of the Communes of Sessa Aurunca and Cellole (Caserta).
Methods: A cross-sectional study in a sample of wells in Caserta area was conducted. Data about depth, nature of aquifer, centration of fluorine and radon were collected through sampling and measurement with Alpha guard. Descriptive, univariate analysis using Mann-Whitney test for non-normal distributions were applied. The correlation between depth of well and concentration of radon and fluorine was conducted with Pearson correlation.
Results: The sites submitted to analysis were 21 wells. Concentration of radon had a mean of 86.7 Bq/l and standard deviation 92.3; fluorine had a mean 1.4 mg/l, standard deviation 0.6. The univariate analysis showed a significant difference between vulcanite and sand clays rocks in radon concentration (p=0.08). The correlation analysis showed non significant association between concentration of radon, fluorine and depth of well, respectively (r=0.27; P=0.23) and (r=0.21; P=0.35).
Conclusions: The study showed a close relationship between radon and lithological characteristics, and no correlation with depth of wells. It suggests to actively monitor radon concentration both water for human consumption and living environment of the territory.

Keywords: ground water, radon, fluorine, measurement.

Introduction

Radon (Rn) is a natural radioactive gas formed from uranium decay. Two isotopes of the Radon are of interest for their concentration in the environments: Radon222 and Radon220. Radon 222 decays within a few days, forms decay products including polonium-218 and polonium-214; all these isotopes emits alpha radiation(http://www.epicentro.iss.it/problemi/radon/radon.asp)

Radon is in the earth's crust, the amount depends on uranium and radio content of rocks that varies according to the type of rock or soil. The gas propagates into the air and can dissolve in water; considering it is odorless, colorless and tasteless strict controls must be applied to water for human consumption. Irving Kaplan "Nuclear Physics" second edition.(Department of Nuclear Engineering Massachusetts Institute of Technology). Radon concentrations in surface waters such as rivers and lakes are generally very low, usually below 1 Bq/l. Concentrations in ground water vary from 1 to 50 Bq/l for sedimentary rocks, from 10 to 300 Bq /l in the case of wells and from 100 Bq /l to 50.000 Bq /l for waters from crystalline rock.

The World Health Organization (WHO), through the International Agency for Research on Cancer (IARC), has classified the radon belonging to group one of carcinogenic substances for human beings.

The relationship between exposure to ionizing radiation and health effects is well known. UNESCAR 1982, Ionising radiation source and biological effects, United Nation Scientific Committee on the Effects of Atomic radiation 1988 Report to the General Assembly, 1988. Radon is dangerous because the alpha particles can damage the DNA of the cells causing lung cancer due to inhalation and stomach cancer for ingestion. http://www.epicentro.iss.it/problemi/radon/radon.asp

Chronic exposure to high contributions of Fluorine in water can be dangerous too; it can cause alterations in the process of bone calcification and calcium metabolism. Levels of fluoride in the water should be monitored too. http://old.iss.it/binary/publ/cont/Pag1_174Rapporto97_9.pdf

The Environmental Protection Agency (EPA) estimated that Radon in drinking water, causes 168 death for cancer every year, 89% lung cancers and 11% are stomach cancers. EPA (Enviromental Protection Agency) acqua October 1999 Bruno RC (1983). Source indoor Radon in houses: A Review, J Air Pollu.Contr. Assoc. 133,105. Fonti EPA Marcinowski F.(1992). Nationwide survey of residential Radon levels in the USA. Fonti Epa University of Minnesota Enviromental and occupational Healt.

Many countries have issued regulations or recommendations to ensure that radon concentration levels do not exceed reference values called "action levels".

Regarding ground water, the guidelines provided by WHO and the European Commission, recommend controls if radon concentration exceeds 100 Bq/l. Raccomandazione della Commissione 2001/928/Euratom sulla tutela della popolazione contro l'esposizione al radon nell'acqua potabile; Gazzetta ufficiale delle Comunità Europee n. L344, 28 dicembre 2001. The national legislation on the potable water establishes that the value of the "Fluoride" parameter must be included within the limit of 1,5 mg/l. http://www.camera.it/parlam/leggi/deleghe/01031dl.htm

Regional and Provincial Environmental Protection Agencies (ARPA and APPA) realized a national survey about radon exposure in houses. Data shows great variability of average regional concentrations, from a few dozens of Bq/m3 to over 100 Bq/m3. http://www.isprambiente.gov.it/it/temi/radioattivita-ambientale/Radon/la-situazione-in-italia Differences are due to geological characteristics and vary locally within the context of individual regions. Campania is the third Italian region for concentration of Rn-222 in the houses. http://www.isprambiente.gov.it/files/temi/tabella-radon.pdf A national monitoring of radon in aquifer is missing, but there are local measurement for example the determination in Veneto Bertolo A, Bigliotto C. Radon concentration in waters of geothermal Euganean basin--Veneto, Italy. Radiat Prot Dosimetry. 2004;111(4):355-8. and in Etna area D'Alessandro W, Vita F. Groundwater radon measurements in the Mt. Etna area. J Environ Radioact. 2003;65(2):187-201.

This study focuses on the municipality of Sessa Aurunca and Cellole (Caserta) that record high level of radon both in the well and the source waters also the fluorine measurement was performed.
The aim is to determine the concentration of radon and fluorine in the waters sources of the Communes of Sessa Aurunca and Cellole (Caserta). Department of Prevention - Area 6 - Health District no. 43 of the ASL EC/2, the section intended for human consumption, directed sampling and monitoring.

Methods

Study design and study setting:

Data for this cross-sectional study were collected in 2005, resampled and double-checked in 2016 through analytical measure of radon and fluorine in water for human consumption from well. The study performed in Italy and in accordance with the STROBE statement. Vandenbroucke JP, von Elm E, Altman DG, et al. Strengthening the Reporting of Observational Studies in Epidemiology (STROBE): explanation and elaboration. Epidemiol Camb Mass 2007; 18:805–35. doi:10.1097/EDE.0b013e3181577511

The choice of site:

The sites of study were the wells in the municipality of Sessa Aurunca and Cellole (Caserta).
Elements kept in consideration in the choice of the sites have been:

1. Uniformity of distribution of wells in the territory;
2. Geologic and hydrogeological characteristics of the area;
3. Availability of other analytical determinations on the waters;
4. Practical characteristics such accessibility, reachability and possibility to do good sampling.

Withdrawal methods:

For the determination of the Radon in water, an instant sampling was carried out for each well. Two different methods were implemented, as it is known from literature that the sampling methods are of fundamental importance and Radon is not very soluble in water (solubility coefficient: 0.250 to 0 ° C).
The first method involves the collection of water through a 100 ± 1 ml syringe and transfer in a 150 ± 5 ml Pyrex test tube. The second method is with a small diameter rubber tube of 0.4-0.5 mm: one end is inserted into the delivery pipe and the other inside the same tube used with the previous

method. The sampling with this system allows greater accuracy and avoid the formation of air bubbles and bubbling at the time of transfer into the tube. The diameter of the tube allows the formation of a constant flow in laminar regime, especially in the passage in the test tube.

All picking and storage operations were carried out with attention to avoid losses and preserve the representativeness of the sample.

Once in the laboratory, the tubes were brought to a volume of 100 ml and the samples were analyzed using an ionization chamber active system: AlphaGuard.

Instrument measure:

The experimental measures were done using the AlphaGUARD, an instrument for the determination of Radon. AlphaGUARD is universally used for the determination of Radon and its decay products in soils, water, indoor and outdoor environments and building materials. The instrument provides the Radon concentration in Bq/m3 which is converted by a conversion factor in Bq/L, on the basis of environmental parameters such as: temperature (°C), pressure (mBar) and relative humidity (% rH).

Measure of sampling error:

Different sources of uncertainty were considered in the experimental measures:

1. Systematic error: instrumental error for the calibration of the instrument. It is esteemed around 5% - 7%;
2. Measure error: it is referred to variation of the environmental parameters. It is esteemed around 15% - 18%.

Verification of collecting and maintenance:

The uncertainty in the phase of collecting of the champion has not been quantified for the impossibility to bring the tool in situ. It is included therefore in the measure error. The uncertainty in the phase of maintenance has been reduced by the imminent analyze of champions coming in the laboratory. The error of analysis was of 20%.

Statistical analysis:

The statistical analyses included descriptive and correlation analyses. Descriptive analysis include characteristic as municipality, well, depth and concentration of radon and fluorine for each well. Univariate analysis was carried out for which the Normality test Kolmogorv - Smirnov was preliminarily applied. The test then used to evaluate the mean difference between acquifer groups and radon concentration was: Mann-Whitney test for non- normal distributions. The correlation between depth of well and concentration of radon and fluorine was applied with Pearson correlation. The correlation between fluorine and radon was still studied.

Results

The sites submitted to 222Radon analysis were 21 wells in the municipality of Sessa Aurunca and Cellole. Depth varied between 60 to 300 meters. Sessa Aurunca involved 17 (77%) of sampled well, 5 (23%) were in Cellole. Concentration of radon had a mean of 86.7 Bq/l and standard deviation 92.3; fluorine had a mean 1.4 mg/l, standard deviation 0.6. Geographical description and the depth of each well is reported in table 1. Synthesis measure are reported in **table 2.**

Table 1. Sample characteristics

Municipality	Well	Label of well	Depth	Aquifer	Radon Bq/l	Fluorine mg/l
Sessa A.	Sorgente 1	1	200	Vulcanite	61.62	0.7
Sessa A.	Sorgente 2	2	40	Vulcanite	90.13	0.2
Sessa A.	Valogno	3	200	Vulcanite	167.1	1.5
Sessa A.	Raccomandata1	4	120	Vulcanite	167.6	0.8
Sessa A.	S. Agata 1	5	140	Vulcanite	154	1.2
Cellole	Cellole Freiole	6	60	Vulcanite	31	1.2
Cellole	Cellole Campo	7	90	Sands clays	34.4	0.7
Sessa A.	Piedimonte	8	140	Vulcanite	79.1	0.5
Sessa A.	Cupa	9	140	Vulcanite	22.1	2.4
Sessa A.	Rongolisi	10	180	Vulcanite	168.7	1.3
Sessa A.	Lauro	11	120	Vulcanite	61.7	1.6
Cellole	Baia Felice	12	130	Sands clays	22.2	1.4
Sessa A.	S. Carlo	13	300	Vulcanite	66	2
Sessa A.	Terme	14	140	Vulcanite	420	2.9
Sessa A.	Caseificio	15	80	Sands clays	11.2	1.5
Sessa A.	Az. Agricola	16	80	Vulcanite	65.3	1.7
Sessa A.	Mulino	17	60	Vulcanite	65.7	1.6
Cellole	Sgorbi	18	60	Sands clays	14.9	1
Sessa A.	La Foce	19	100	Sands clays	62.3	1.2
Sessa A.	S. Castrese	20	60	Vulcanite	27.9	2
Sessa A.	Baia Domizia	21	100	Sands clays	29.6	1.8

Table 2. Concentration summary measures

Radon	Mean (SD)	86.7 (92.3) Bq/l
	Median (range)	62.3 (11.20-420)
Fluorine	Mean (SD)	1.4 (0.6) mg/l
	Median (range)	1.4 (0.20-2.90)

Bar chart of radon and fluorine concentration in well waters are reported in a bar chart in table 2 and table 3. The red line in the graph represents the concentration value above which the current legislation provides for the implementation of mitigation measures. Five wells (24%) are above the threshold of radon centration, 8 wells (38%) instead are above the threshold concentration of fluorine.

Table 3. Concentration of Radon in well

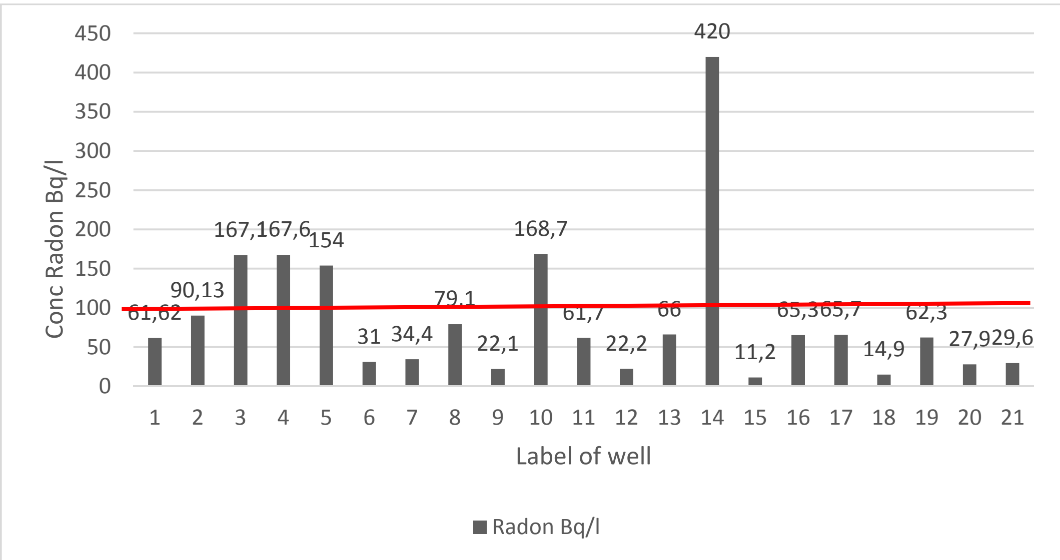
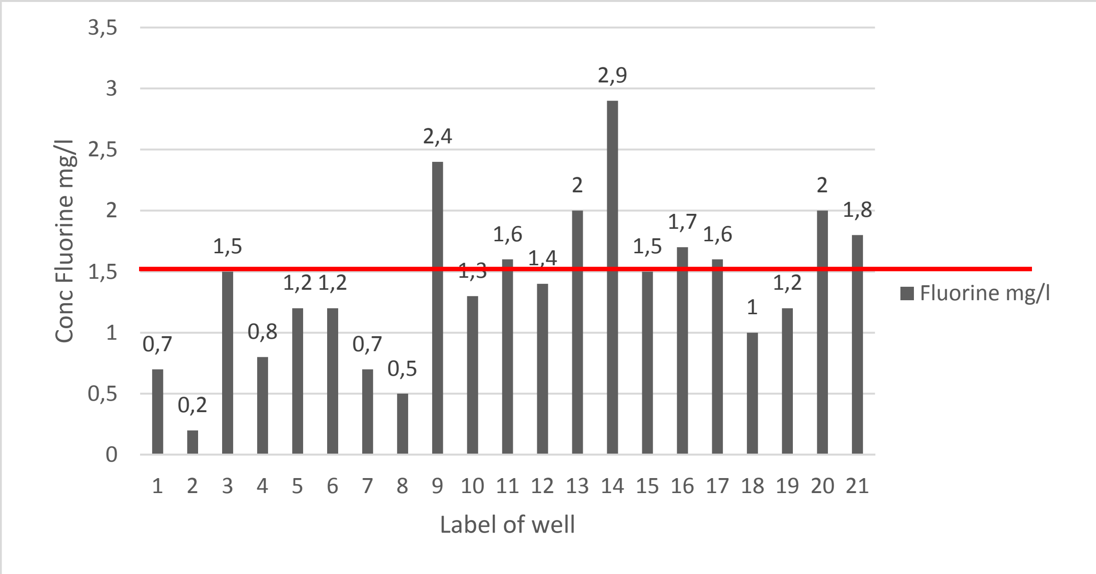


Table 3. Concentration of Fluorine in well



The univariate analysis shows a significant difference between vulcanite and sand clays rocks in radon concentration (p=0.08).

Table 4. Univariate analysis of radon concentration

		N° wells	Mean Rank	p*
	Sand clays	6	5.3	
Radon	Vulcanite	15	13.3	**0.08**

*Mann Whitney test

The correlation analysis showed non significant association between concentration of radon and depth of well (r=0.27; P=0.23) and between concentration of fluorine and depth of well (r=0.21; P=0.35). Results are reported respectively in table 5 and 6.

Table 5. Correlation between depth of well and concentration of Radon

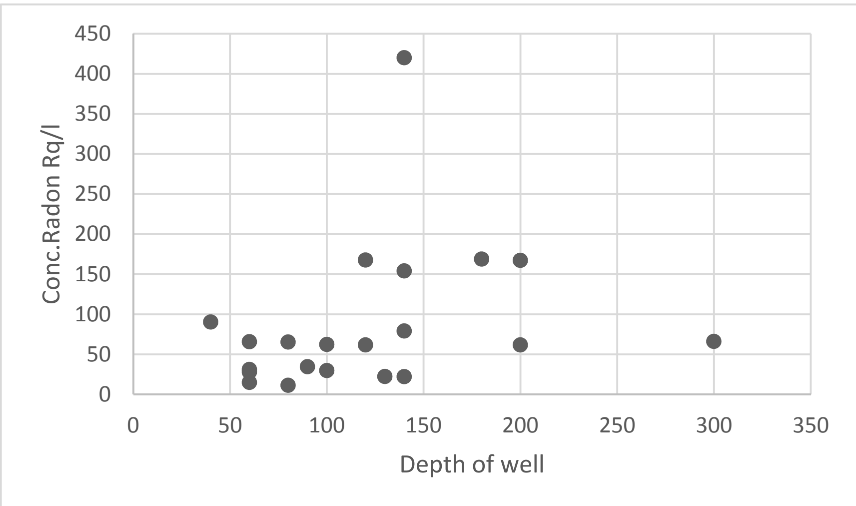
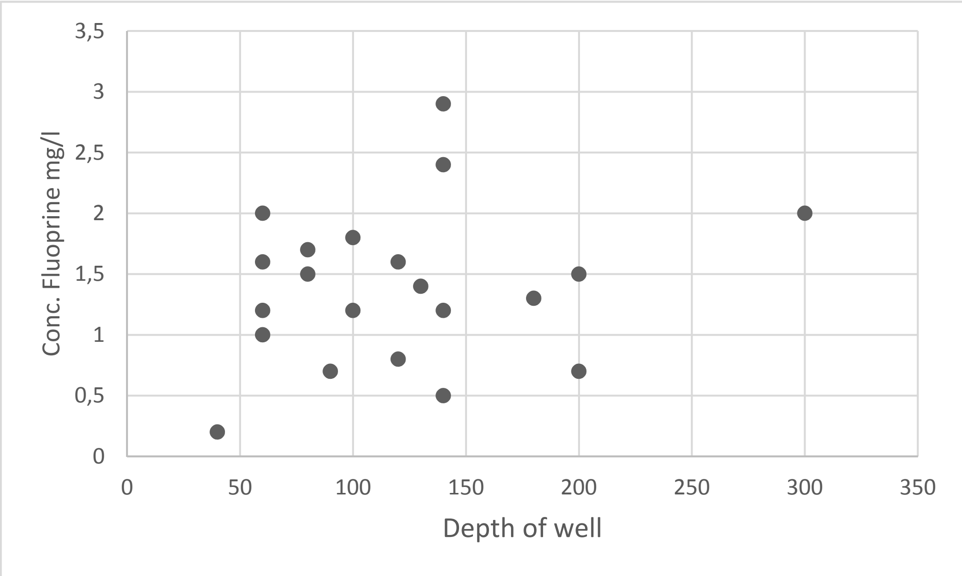
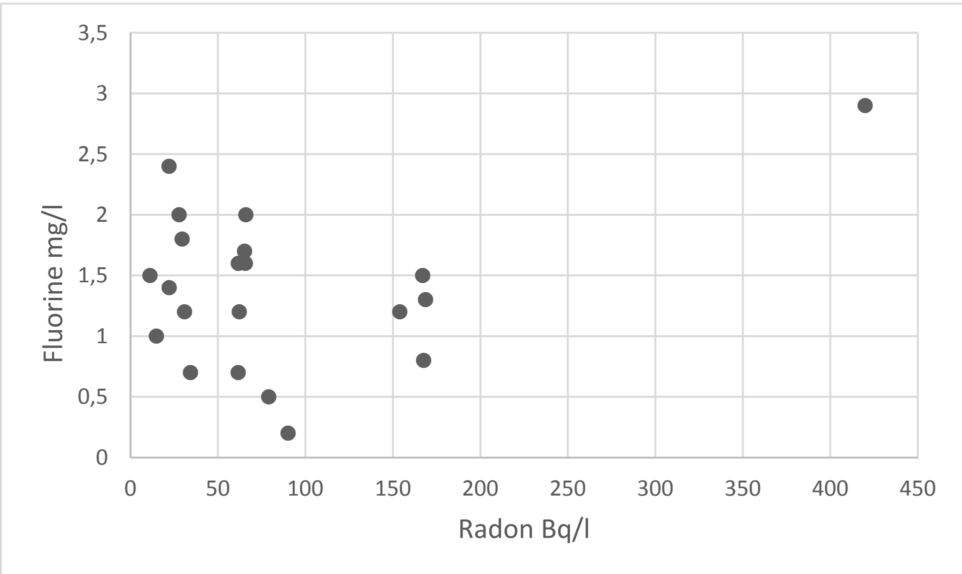


Table 6. Correlation between depth of well and concentration of fluorine



The correlation analysis between concentration of radon and fluorine showed non significant association too (r=0.32; P=0.15) Results are reported respectively in table 6.

Table 6. Correlation between fluorine and radon



Discussion

The data considered showed that the analyzed territory introduces waters destined to the human consumption coming from wells of depth between the 50 and 300 uniformly distributed in the territory.

The values of Radon activities vary widely from around 10 Bq /l to 170 Bq/l with a peak of 420 Bq/l. However, it suggests that there is interaction between several strata due to the poor or non-existent isolation of different aquifer determined by natural causes or related to the construction of wells. The maximum value of 420 Bq/l was found in a well made for mineral water research of recent construction. It was build to capture of a single aquifer located at the depth of 140 meters from the ground with water at a temperature above 30°C. This probably confirms what was previously mentioned that the value in all the wells is influenced by the presence of several overlapping aquifers having different values of radon concentration.

The aquifers have average concentrations of fluorine in many cases high and higher than those found in surface aquifers.

The study shows a closer relationship between radon and lithological characteristics consistent with the data available in the literature (http://www.epicentro.iss.it/problemi/radon/radon.asp). In fact the greatest values are found in aquifers of volcanic rocks and after a certain contact time. The decrease in the values in plain area is related to the type of reservoir rock (sands and clays) too.

In addition there is a poor correlation between gas concentrations and depth, so values do not increase proportionally with the increase in the depth of the aquifer.

For fluorine, however, there are data, albeit unpublished but certain, that directly correlate the value of the fluorine with the depth of the aquifer: in some perforation the strata intercepted during the drilling and survey of a well show increasing values with increasing groundwater depth.

The reference level of 100 Bq/l, defined by the European Commission Recommendation 2001/928/Euratom (Raccomandazione della Commissione 2001/928/Euratom sulla tutela della popolazione contro l'esposizione al radon nell'acqua potabile; Gazzetta ufficiale delle Comunità Europee n. L344, 28 dicembre 2001), is exceeded by a quarter of the wells. Also many wells exceed the fluorine threshold of C.M.A. set by Decree 31/01. Decreto legislativo 2 febbraio 2001, n.31 "Attuazione della direttiva 98/83/CE relativa alle acque destinate al consumo umano"; Supplemento ordinario alla G.U. n. 52, 3 marzo 2001.

The available data and the statistical elaborations allowed to circumscribe an area in which the waters present alert values of the quantity of Radon present. This area is recognized in the circumvulcanic area of the Roccamonfina.

Strength and limitations

The major strength of the study is that, although certainly not exhaustive, is the first that define radon concentration in Sessa Aurunca and Cellole in the Area 6 Health District no. 43 of the CE / 2 ASL. It focused attention in a territory where 222Radon concentration exceed the threshold values. Nevertheless, the present work is affected by some limitations that should be acknowledged. Firstly, measurement experience were not carried out on site, secondly, the investigations did not involve the determination of the chemical-physical characteristics of the aquifer and other radioactive elements.

Conclusion

Despite of these limitations this study makes important contributions to current literature monitoring radon concentration in an area where reference level of radon are often exceed. The study shows a close relationship between radon and lithological characteristics, and no correlation with depth of wells.

It suggest to actively monitor radon concentration both water for human consumption and living environment of the territory. It would be important to proceed with radon measurements during the construction of new perforations.

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