SURVEY OF RADON IN AUSTRALIAN RESIDENCES



S.A. Baggs

Hon. Research Fellow, Graduate School of the Built Environment University of New South Wales Kensington 2033

C.F. Wong

Lecturer in Physics, Queensland Institute of Technology Brisbane 4001



Present trands indicate that: (i) a problem of high levels exists in some earth-wall and dugout dwellings which, if uranium mine data are extrapolated, could indicate some increased risk of lung cancer; (ii) when ventilation rate is other than minimal, it does not appear to affect radon concentration in the expected direct inverse way.

Introduction

Some 400 years ago, in the city of Joachimsthal, Czechoslovakia, Georgius Agricola was city physician over the period 1527-1533 [1]. He recorded that an occupational chest disease existed amongst miners; it was known as 'mountain sickness' and was prevalent in the region of that city and of Schneeberg. This disease claimed the lives of some 75 percent of all miners; 100 years later it was diagnosed as lung cancer. In 1924, it was suggested that high concentrations of radon gas in the mines were responsible for these lung cancers [2].

Architects in Australia are entering a new phase in which energy-conservation

strategies are being applied to the production of buildings that rely, in part, for their thermal energy efficiency upon a reduction in the number of interior airexchange cycles. However, questions must be asked concerning the potential effect of such strategies upon the health of occupants. The incidence of radon-222, and the potentially carcinogenous radon daughters in the subject of the first stage of a monitoring programme currently being implemented in Australian buildings.

The problem of radon effects upon health has its source in the decay chain of uranium-238 (Figure 1). In this process, radium-226 occurs with a half-life of 1602 years followed by inert radon-222 (half-



Figure 1: The Uranium-238 decay chain (unimportant branches omitted).

life, 3.8 days). Its four radioactive daughters are not inert, and by attaching to airborne particulate matter (by either chemical or physical means) can be inhaled. When this happens, they lodge in the tracheobronchial and pulmonary regions of the body [3,4], subsequently decaying to release a radiation dosage. The result is a possible increased risk of disease, especially lung cancer. Radon-222 emits an alpha (5.49 Mev) and is also a risk to the lung.

People are exposed to a great range of radiation 'risks' in everyday life, and the estimated risk of contracting cancer from those sources is one in 50 000 to one in 100 000. In a lifetime there could be a maximum of one in 1000 dying from cancer caused by natural radiation; yet something like one in five cancer deaths are caused by environmental factors other than radiation [5,6].

Radon, unlike other products in the uranium series, exists in gaseous form at normal temperatures, is soluble in water and the attached fraction of daughter products (some 60 percent) can be retained on the bronchial epithelium. These filial products of polonium-218, polonium-214, lead-214 and bismuth-214, all have half-lives of less than 30 minutes (lead-210 is outside this range). Hence, the daughters present a greater cancerforming potential in the respiratory tract than radon-222. When associated with water vapour, or other small clusters of molecules, these are referred to as 'unattached'. If incorporated in aerosol particles larger than the previous by a minimum of one order of magnitude, they are referred to as 'attached' [7,8]. 'Attached' particles enter (and more efficiently deposit within the respiratory system) than 'unattached' as follows:

the absorbed [alpha particle] radiation energy may induce latent damage in cells which will eventually cause them to become malignant....this activity may penetrate the pulmony capillaries and be transported by the bloodstream to other organs before the decay of the radon daughter products [9].

As long ago as 1967, the US Federal Radiation Council advised that more needed to be known about exposure to low concentrations of radon daughters than existed then. Research to that time had been concerned with high levels of exposure, and results had been mainly derived from epidemiological studies of uranium miners in which the exposure dosage was many times greater than that occurring under normal conditions [10]. The situation has changed very little since that time.

In the 1950s, US medical surveys established a causal relationship between radon daughter intake and an increase in lung cancer amongst uranium miners of the Colorado plateau [11]. Epidemiological studies place the risk of lung cancer for uranium miners in the range of 7 to 40 x 10-5 per WLM (see Glossary [12]. To date, there has been no history of excess lung cancer incidence amongst population groups exposed to high radon daughter concentration for interior domestic environments. It is only in the past 10 years that high interior air radon concentrations have been found to exist in houses in such countries as Norway, Sweden and the United States of America (*ibid*.). Although there is a general tendency in the literature to translate uranium mining data and results into the domestic environment, this extrapolation has not yet been validated. Consequently, any positive correlation of the incidence of cancer in users of domestic environments with radon daughter dosage has not yet been found.

In above-ground buildings, radon levels are typically higher than ambient This radiation background levels. emanates from rock and soil, concrete and certain building materials, including plastics [these are discussed in detail in Reference 5]. Radon from soil and those building materials that employ rock in their manufacture, enter a building through three principal pathways: (i) floor, wall joints and cracks in concrete; (ii) floor drains with loose fitting pipes; and (iii) in tap water taken from groundwater sources.

Eventually, the objectives of a major architectural study would be:

1. to investigate the levels of radon concentration in the interior and exterior environments of various dwelling types;

- 2. to relate these results to the incidence of use of various techniques intended to increase the efficiency of thermalenergy usage in these dwellings; and
- 3. to make recommendations concerning the relationship of air exchange rates between the dwelling and its ambient atmosphere in terms of preferred levels of radon-concentration and in the context of an increasing trend towards the design (or retrofitting) of dwellings for energy efficiency.

However, this section of the research, about which the present preliminary report has been written, was undertaken as a pilot study and concerned Item 1. only.

Conventional Methods of Measuring Low Concentrations of Radon-222 in Air

Conventionally, spot sampling of radon in air is used as a means of measuring the total alpha activity in environments of low concentration. To allow the ingrowth to transient equilibrium of the radon daughters, collection for a long enough period (usually three to five hours) in a calibrated Lucas Cell has been a preferred method [13]. However, problems arise when this method is used to deal with wide-ranging concentrations of radon and when alpha radiation concentrations build up to the limit of detection. US results obtained by this method have varied widely by a factor of up to three [14]. Results in Great Britain have varied up to a factor of 10 [15]. Similar variability has been found in Canadian measurements [16].

These difficulties may be overcome, and accuracy and precision in the measurement of low level environmental radon concentrations can be improved by the use of long-term integrating radon monitors. Of the various passive type dosimeters developed, those used for this survey of 50 Australian dwellings were plastic nuclear track detectors (CR-39 manufactured by Penshore Moulding Ltd, UK, and LR-115 manufactured by Kodak-Pathe Coy). All detectors (for the 14 sites covered in this report) were left in place for at least one year, and, using the responses of occupants to a survey questionnaire circulated, the principal factors that affect air exchange rates were assessed for each sampling site.

Experimental Procedure

Detectors

For the measurement of airborne nuclides that emit alpha particles, plastic nuclear track detectors were used in this experiment [17]. The design of the sensor plate that combined two detectors of each type provided a good average and therefore, reliable results (see Appendix A).

Sampling Sites and Locations of Detectors

Fifty Australian dwellings of differing design types situated in a variety of microenvironments were chosen for the present study. These dwellings were roughly categorised as follows:

- (a) *Standard above-ground*, cavity-brickwall houses (referred to in Figure 3 as 'sa') with on-grade as well as elevated timber floors.
- (b) *Standard above-ground* houses with earth-walls (including mud-brick and rammed earth, referred to 'ew') with on-grade, as well as elevated timber floors.
- (c) *Terratecture* houses (walls and roof earth-covered, floors: concrete on-grade, referred to as 'tt').
- (d) *Earth-sheltered* houses (walls only earth-covered, floors: concrete on-grade, referred to as 'es').
- (e) Lithotecture houses (excavated into self-supporting rock ('dugouts'), referred to as 'lt').

(Note that Categories (c), (d) and (e) are together classified as 'geotecture' [18].)

Occupants of dwellings were instructed to place the plastic nuclear track detectors in four positions inside and outside their homes. These were:

- (i) the main bedroom of each dwelling (the room in which the potential radon exposure is probably an optimum on the basis of daily usage);
- (ii) the outside air under the cover of a roof overhang surrounding the dwelling to be monitored;
- (iii) the water in a toilet cistern or drinking-water supply tank; and
- (iv) the soil adjoining the monitored building at a depth of 36 cm.

Detector Processing and Calibration

Detector processing: The method of processing detectors has been described elsewhere [19] (see Appendix B).

Calibration: Since the main component of the alpha-emitting nuclides in indoor air is radon, it was decided to calibrate the nuclear track detector with radon gas [20]. (The method is also described in Appendix B.)

Survey method: A postal survey was incorporated into the programme. Dwelling occupants were required to respond to a user-survey which sought to determine the various architecturalphysical data needed to assess air infiltration and ventilation effects, as well as smoking habits and medical history (with specific reference to cancer).

Results

Experimental

Results of calibration experiments and of the exposure of the detector plates at 14 of the 50 sampling sites are presented below. Each number represents an average of four detectors on the same sensor plate.

Calibration: Experiments for calibration (previously described) were repeated for more than 10 different integrated activities of radon.

The results were used to obtain averaged calibration factors [20].

No.1

Table 1Calibration Factors

Type of Detector	Radon Activity/track density/time Bq m ⁻³ /track cm ⁻² /day ⁻¹	
CR-39	2.7	
LR-115	13.5	

The calibration factors were estimated to be accurate to about ± 5 Bq m⁻³/track cm⁻²/day⁻¹. These figures were tested with the results obtained for samples in other indoor exposures. This yielded a mean activity of 17 Bq m⁻³ measured in a timber house in Brisbane. The result compares favourably with the mean activities of 10 to 30 Bq m⁻³ measured in some private dwellings by a Melbourne group using a different detecting method [21].

Mean Activity: The track density for detectors exposed at various sites was determined in the way previously described. If the alpha activity in indoor air was entirely or almost entirely due to radon emission from building materials and ground, then the mean activity may be deduced from the track density using the calibration factor derived in the preceding csection. The results of these calculations are presented in Table 2 where the various building types are grouped together. Because of the presence of soil dust and condensation on some of the returned detectors, the data concerning water and soil have not been included.

Working level: To assess the hazards of these results, it was necessary to calculate the energy deposition and the dose equivalent delivered to the human lung. The potential alpha energy concentration (a) measured in working (WL) of any mixture (m) of radon daughters is as follows:

(a) = 3.7 f(Rn)/(mWL) (1) where:

(Rn) = radon concentration in Bq m⁻³;

f = equilibrium factor for radon daughters [22].

Table 2Mean Activity

Site No.	Indoor Bq	Outdoor m ⁻³	Building
1	81	29	lt
11	117	30	lt
12	74	28	lt
19	33	29	sa
29	38	22	sa
55	30	11	sa
26	72	30	tt
30	89	38	tt
41	60	21	tt
31	50	16	es
36	388	49	ew
38	19	16	ew
39	97	22	ew
42	90	48	ew

The equilibrium factor for radon daughters varies considerably with atmospheric conditions. The mean value obtained in the survey of private dwellings was 0.5, and the deviation from the mean for indoor air was found to be within 40 percent [11,12]. For present purposes, the mean value was adopted for the calculation of the working levels, bearing in mind that this value may lead to an underestimate of the equilibrium factor in poorly ventilated houses. The mean working level was calculated with Equation (1) and the results follow in Table 3.

It has been proved experimentally, and also from published data, that the variation of the f-factor in indoor air is less than 40%. Although it is necessary to correlate the f-factor and the air exchange figures for accurate comparison, for this report on preliminary measurements, it is sufficient to use the mean value of the ffactor published in the literature. Further measurements are needed for accurate comparison. There are two ways of approaching this problem. One is to measure the equilibrium factor for each building using, for example, an air sampling method. This would be too difficult to achieve. Another approach is to use a masked nuclear track detector in a Architectural Science Review

(2)

Site No.	Indoor	Outdoor	Building Type
1	• 11	4	lt
11	16	4	lt
12	10	4	lt
19	5	4	sa
29	5	S	sa
55	4	1	sa
26	10	4	tt
30	12	5	tt
41	8	3	. tt
31	7	2	es
36	52	7	ew
38	3	2	ew
39	13	3	ew
42	12	7	ew

Table 3

sample cup for the detection of airborne alpha-emitting nuclides. The mask is used to remove radon so that only radon daughters would be detected. The second method is feasible for future surveys but it will take time to complete.

Infiltration: From the survey data, the two major types of infiltration were estimated. The first depended on the number of air exchanges that would have occurred in the test room (where the sensor plate was located) over the monitoring period; these modified the radon build-up rate. The second depended on the total area of apertures connecting the interior air with the subfloor void, where a crawl-space existed; this facilitated radon infiltration from the soil beneath the building.

After determining whether a sub-floor void existed and whether the sub-floor surface was bare or had been sealed with concrete or some other material, the total area of crack-connections between the interior and sub-floor void were estimated from information supplied by respondents.

The air-exchange rate was determined from the number of exposed exterior windows in the test room; the purpose for which the room was used; the window type and size, as well as the length of time they were left open throughout the year



Figure 2: Increase in airflow caused by excess of area of one opening over another.

and the extent to which they were opened; the presence of outside air and sub-floor air-ventilator connections; and the number and frequency of use and extent of opening of all doors to the test room.

There are many methods used to calculate air-exchange rate [23], but a simple equation:

$$Q = EA$$

where:

 $Q = airflow (m^{3}h^{-1});$

V

- E=effectiveness of openings (for present purposes, taken as 0.30);
- V=wind velocity (half the average seasonal velocity, m h⁻¹) [24].

was used for calculating the quantity of air forced through openings and cracks by the wind. The reasons for this choice are complex, and their discussion would occupy too much space. Equation (2) gives an air flow per unit area that is slightly too low for windows not advantageously placed and slightly too high for openings that are normal to the prevailing wind.

Ventilating openings were assumed to have a flow coefficient slightly greater than that of a square-edge orifice. Inlets and outlets, including crack area, should be equal to maximise air exchange; where they were unequal the smaller area was



Figure 3: Fourteen Australian sites rank-ordered in relation to their mean interior air alpha radiation readings (showing air exchange rate, mean outdoor alpha and mean soil alpha radiation results).

- Key: 'sa' = standard above-ground houses with cavity-brick walls, including those with on-grade, as well as elevated timber floors.
 - 'ew' = mud-brick, rammed earth houses. This category includes standard above-ground houses with earth-walls and on-grade, as

adopted, and Figure 2 was used to adjust for the area differential, and to derive the increase to be added.

Figure 3 has been constructed to indicate relationships, where they exist, between the various levels of mean alpha activity at any site and the average air exchange rate per hour considered on a yearly basis.

Postal Survey

There was no incidence of any type of

well as elevated, timber floors.

'tt' = terratecture houses with concrete floor, with walls and roof earth-covered.

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- 'es' = earth-sheltered houses with concrete floor, with walls only earth-covered.
- 'lt' = lithotecture dwellings excavated into self-supporting rock ('dugouts').

Note: Scale C is not an absolute scale; it facilitates assessments of relative air exchange rates within the set of 14.

cancer in the respondent group; although the period of residency varied from 2 to 25 years with an average of 9 years, only one respondent had been in occupation for 25 years.

Discussion

The results of indoor samples show a distribution of radon activity in a great range from 19 Bq m⁻³ to 388 Bq m⁻³. There is an increase of about 20-fold from Site 38 to Site 36. From Figure 3, which gives the preliminary survey sample for the 14 sites, it can be seen that the

expected inverse relationship between air exchange rate and mean indoor alpha activity did not occur as a general trend. At Sites 55 and 29, very high air exchange rates were accompanied by low levels of internal air alpha concentration; at Site 11, the interior alpha level was relatively high despite a high air exchange rate.

Table 4 gives the result of a statistical correlation. The mean value of the interior alpha activity concentration and the air exchange rate were calculated for each building type. The standard errors of the mean are also given in this table.

Table 4				
Statistical Correlation				
Interior Alpha Activity Versus Air				
Exchange Rate				

Building Type	Interior Alpha Activity Bq m-3	AirExch- ange Rate*
Earth-Sheltered	50	59
Standard above- ground	33 ± 2	68 <u>+</u> 10
Terratecture	74 <u>+</u> 8	16±8
Lithotecture	141 <u>+</u> 42	55 ± 20
Earth-wall, above-ground	149 <u>±</u> 82	32 <u>+</u> 4

*Air exchange rate in arbitrary units, indicating proportional relationahip within the set only.

The earth-shelterd, standard aboveground and terratecture buildings have an approximately inverse relationship between air exchange rate and indoor alpha activity. The lithotecture and above-ground earth-wall buildings deviate from the inverse law. Both these types have a large variation of activity concentration. In the latter case, the activity concentration varies from the low of 19 Bq m-3 found at Site 38 to the extremely high of 388 Bq m-3 found at Site 36. This range gives rise to a standard error of more than 50% of the mean.

Lithotecture also gives rise to a large range of mean activity concentration. While the cause of the discrepancy is still unknown, it seems some reasons other than ventilation have affected the activity concentration in these buildings. Anthony Nero Jr. has reported a similar result in an article: 'Indoor Air' in the 1986 Yearbook of Science and the Future, where he wrote:

In a number of studies both the radon concentration and the ventilation rate were measured in groups of houses, including ordinary houses and some that had unusual energy-conservation features. The surprising result was that, although there were great differences in concentrations among the houses, they had no apparent correlation with the ventilation rates.

The above-ground building at Site 19 has its indoor and outdoor air readings very close together because it is located in the opal-mining township of Coober Pedy, South Australia, and is equipped with a room airconditioner. Hence, interior build-up is unlikely to occur. Other sites with higher exchange rates are Sites 29 and 11. Site 29 is a standard aboveground structure but has excessive ventilation as it is an office, not a bedroom as in all other cases. At Site 11, a Coober Pedy 'dugout', varying the proportion of inlet to outlet may reduce the number of air exchange per house, but would produce interior alpha build-up.

In general, lithotecture 'dugouts' ('lt' on Figure 3) at Coober Pedy gave mean interior air alpha readings that varied from the highest to the middle of the range in the complete set. Standard above-ground residences ('sa') grouped themselves towards the low end of the range, whilst earth-wall houses ('ew') ranged from the lowest to the highest. Terratecture ('tt') and earth-sheltered ('es') samples were grouped around the middle of the range.

An anomolous situation was found to exist at Site 36, where a very high alpha level prevailed together with a high ventilation rate. However, the existence of raw mud-brick walls could possibly explain this, as it was the only case of raw surfaces in the test rooms of any earthwall dwelling in the sample of 14. The outdoor reading, that registers the effects of inversions, etc. was not high; the owner of the building was contacted and he established that the source of the soil for his walls had been an alluvium quarry containing a high proportion of metamorphics in the river sand. In this instance it would be prudent to seal the wall surfaces.

The samples in the rest of the dwellings at Sites 19, 29, 31 and 55 registered an average level of 38 Bq m⁻³. While all these are above-ground houses, the average level obtained is not inconsistent with the average found in any Australian home.

Most of the soil and water samples were unreliable because dust and water condensation had collected on their surfaces. The relative values of these samples given in Figure 3 should be used for qualitative comparison only. Nevertheless, a large range of values can be observed which may be correlated to the high activity concentrations registered by the indoor samples. The extremely high indoor level found at Site 26 suggests a high concentration of radionuclides such as uranium or thorium. This is consistent with the report that bore water is used at this site.

An estimation shows that the risk of cancer due to lifetime exposure to airborne nuclides at Site 36, which has an extraordinarily high level of 388 Bq m⁻³ for indoor activity, is close to one in 10 000, while the average risk to background radon in indoor air is about one in 1 000 000, if extrapolation from uranium mine data is valid.

Problems Associated with the Study

Selection of the sample could not be randomised; the problem was to identify a large enough sample of residents in a reasonable range of building types who were sufficiently cooperative to undertake the tasks involved in putting the detectors in place and returning them some 12 months later with the completed questionnaire. However, the major problem associated with this study involved the estimation of an air-exchange rate from the survey data on degree of

wind exposure, the factors concerning window and door types outlined previously, as well as the presence or absence of gaskets and caulking, and the extent dimensions and of wall/floor/ceiling cracks. Such estimates are, at best, indicative of relative air exchange rates within the set, and cannot be considered as hard figures. The only solution to this problem would be to subject each dwelling to the type of airleakage test that is under development by Associate Professor John Ballinger at the University of New South Wales. However, on the basis of cost, the application of this test to such widely scattered survey sites would be prohibitive.

Conclusion

The postal survey of this small sample of 14, produced no information concerning the health of the occupants with respect to lung cancer. The buildings at Sites 39 to 42 are at the top of the range of indoor readings for the remaining sites, although nowhere near the level of Site 36, but thus have painted earth-wall surfaces that may have inhibited radon exchange. The building at Site 11 requires a higher ventilation rate, and all rock surfaces should have painted hardwall plaster added. It appears that ventilation rate may not affect radon concentration in the direct inverse way expected, an observation borne out by overseas research.

Although the data from only 14 sampling sites were available at this time, the results are interesting. They suggest that the average level of radon in lithotecture is about 30 percent higher than in terratecture. In addition, the highest level measured in the present study indoors, in a house of earth-wall construction with unsealed mud-brick walls, was found to be approximately 20 times higher than the lowest level of 19 Bq m^{-3} in a house of cavity-brick construction. Such high levels could increase the risk of lung cancer by a factor of more than 10, if the extrapolation of data obtained from uranium mines is valid. These results apparently reveal a health hazard, and a comprehensive

survey for radon activity in Australian buildings is indicated to determine if such high levels exist elsewhere.

Appendix A 🕑

Nuclear track detectors: The detectors were mounted on a plastic or cardboard backing. Each sensor plate contained four detectors; two were made of CR-39 plastic and the remainder were made of LR-115. The working energy ranges of these two types of detectors are different. The LR-115 detectors are sensitive to an energy of up to approximately 4 MeV, while the CR-39 detectors are sensitive than LR-115 detectors and therefore, will give rise to more accurate counting because high statistical counting can be achieved for the CR-39. On the other hand, the counting procedure for LR-115 detectors gives less ambiguity because the tracks are well defined in the red dye of these detectors. Any defects that occur on CR-39 detectors could strongly affect the result of counting, and further, the plate-out effect is more serious for CR-39 detectors than for LR-115 [9].

Appendix B

Detector processing: The detectors were etched in a concentrated KOH solution at elevated temperatures for suitable periods. The CR-39 detectors were etched at 65° C for 5 hours and the LR-115 detectors were etched at 50° C for 75 minutes. The track density register on the surface of the detectors was determined in track cm⁻²/day⁻¹. At least 5% accuracy in statistics was maintained in the counting procedures. The track density recorded by an unexposed detector that had been processed in the same way was subtracted from the measured track density to correct for etched pits due to defects on the surface of the detectors.

Calibration: A radon chamber [11] was used for calibration. Four detectors prepared from both types of detecting materials were mounted in the chamber on the inside surface of the perspex housing. To provide a known activity of radon for calibration, a radon capsule (seed) with an activity between 30 and 40 MBq (chosen for convenience) was assayed accurately, that is, to within ± 1 percent, by comparing it with a secondary standard radium-226 reference source, using an ionisation chamber in conjunction with an electrometer, and then ageing it to a convenient residual activity.

The seed was opened in the chamber using a method described elsewhere [20]. The detectors were protected by a plastic covering for the six-hour period immediately after the opening of the seed. It allowed the gas in the chamber to reach radioactive equilibrium and adequately mix (*ibid*.). The detectors were then exposed for the required period of time. The exposed detectors were etched, as previously described, and the track density was used to relate to radon activity.

Glossary

Becquerel (Bq) - the SI unit of activity, equal to one nuclear transformation per second. It replaced the Curie (Ci). 1Bq = 2.7 x 10⁻¹¹ Ci. 1 Ci = 3.7 x 10¹⁰ Bq.

Dose Equivalent - product of absorbed dose and quality factor (QF). The SI unit of dose equivalent is the Sievert.

Half Life - the period of time in which half the nucleii in a given sample of a particular radionuclide undergo radioactive decay.

Natural Background - ionising radiation received by the body from natural sources, such as cosmic radiation, terestrial radioactivity, and naturallyoccurring radionuclides within the body.

Quality Factor (QF) - a non-dimensional factor used in the context of radiation protection to express the relative biological effectiveness of different kinds of ionising radiation.

Radon Daughters - the decay products of radon gas, Po-218 (Ra-A), Pb-214 (Ra-B), and Bi-214, Po-214 (RaC-C').

Sievert (Sv) - the SI unit of dose

equivalent. It replaces the rem. 1 Sv = 100 rem.

Working Level (WL) - radon daughter concentration in air that results in an inhaled potential alpha energy of $2.08 \times 10^{-5} \text{ Jm}^{-3}$.

Working Level Month (WLM) - product of Working Level and exposure time. 170 WL hours = 1 WLM.

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