SURVEY OF RADON CONCENTRATIONS IN VIRGINIA AND MARYLAND WELL WATER

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ABSTRACT

In northern Virginia and southern Maryland, approximately 15% of the private homes use private water wells for potable water. A survey of drinking water consumed in 200 homes with private water wells revealed that the dissolved radon in water wells ranged from less than 100 pCiL⁻¹ to about 10,000 pCiL⁻¹, and that the average well water radon was about 2,500 pCiL⁻¹. Average waterborne radon in geological units ranges from about 1000 pCiL⁻¹ in quartzite (somewhat higher in sandstone) to over 3000 pCiL⁻¹ in granitic rocks.

A comparison between the age of study participants who have not developed cancer and their waterborne radon shows no age-versus-radon correlation, indicating that low-radon through high-radon water wells are randomly distributed among the without-cancer study population (i.e., no tendency for older people to consume higher concentrations of waterborne radon). However, a similar age-versus-radon comparison for people who have developed cancer shows that low-radon through high-radon water wells are not randomly distributed among the with-cancer study population. For these people who have consumed their present well water for at least 10 years, there is a positive correlation between the age of cancer development and the waterborne content of their drinking water.

Recent suggestions by the U.S. Environmental Protection Agency encourage the operators of municipal water supplies (and by inference, homeowners with private water wells) to limit their distribution-site waterborne radon to less than 300 pCiL⁻¹. In northern Virginia and southern Maryland, most of the water wells exceed 300 pCiL⁻¹. Consequently, considerable private and public expense may be associated with a national attempt to maintain potable water supplies at less than 300 pCiL⁻¹ of waterborne radon.
INTRODUCTION

The number of deaths from cancer in the U.S. has doubled since 1950, from about 200,000/year to 400,000/year, and has increased an estimated 10-fold since 1900 (Moolenaar, 1988). This observation is largely responsible for the upsurge in public fear and environmental regulation. However, most of the increase in cancer mortality is due to the growth in the U.S. population. Also, cancer is largely a disease of old age, and the only major cause of cancer mortality that has increased in cases per 10,000 population since 1950 is lung cancer, which is believed to be largely due to smoking. Cancer mortality, adjusted for increasing population and age, and exclusive of respiratory cancer, has decreased by about 10% since 1950, and now stands at about 10/year per 10,000 population. The mortality is about equally divided between breast, digestive and gland cancer (Moolenaar 1988).

The identification of natural and industrial-age carcinogens has proven successful when high carcinogen levels are present. The currently used no-threshold model of chemical carcinogenesis, in which mortality at high exposure levels is extrapolated to estimate mortality at lower levels, has resulted in public health policies implemented in the absence of certainty on the scientific judgements. Current U.S. Environmental Protection Agency policy on indoor radon is currently the most widely known example of high-to-low extrapolated risk estimates.

The biological significance of radon is related to the fact that radon and its radioactive daughter products comprise the major portion of the natural internal radiation dose to man. For the general population, excess amounts of these radionuclides can be inhaled when in buildings built over soils with high radon emanation properties, or ingested by consuming well or spring water.

The airborne radon inside a home originates from the geological material beneath the home, as does the radon in the water of homes with private water wells. The only difference is that the radon in well water originates from somewhat deeper geological material beneath the home. Differences in weather, home use and home design strongly influence indoor radon derived from the soil, but these factors probably do not significantly influence the quantity of radon in the home water supply, nor the rate at which dissolved radon escapes the home water supply.

Although the ingestion of radon-enriched water supplies radionuclides to the blood, the possibility that cancer can occur due to drinking naturally radioactive water has not been studied nearly as extensively as might be expected. Private and public water supplies are not regulated for radon. The alpha dose from drinking naturally radioactive water is primarily due to the presence of uranium decay chain nuclides, including Ra-226, Rn-222, Po-218 and Po-214. Of these radionuclides, radon most easily escapes from the surfaces of cracks in rocks and from the surfaces of sediment grains to enter ground water, because radon is a noble gas. For this reason, its concentration is usually many times greater than the other alpha emitters in drinking water.

Few of the radionuclides occurring in natural water are sufficiently studied to facilitate the adoption of regulatory guides. The interim Maximum Contaminant Level for the sum of Ra-226 and Ra-228 has been set by
the U.S. Environmental Protection Agency at 5 pCiL$^{-1}$ (Cothren 1987a). US-EPA drinking water standards have not yet been set for uranium or radon. In the summer of 1991, the U.S. Environmental Protection Agency proposed a Maximum Contamination Level of 300 pCiL$^{-1}$ for radon in municipal water supplies. As will be discussed below, most of the public and private wells in northern Virginia and southern Maryland greatly exceed 300 pCiL$^{-1}$.

HEALTH CONSIDERATIONS

Radon is a noble gas, chemically inert and highly soluble in groundwater. Alpha decay recoil and diffusion are thought to be the mechanism by which radon moves from grain and fracture surfaces into groundwater. In much of the United States, municipal water supplies derived from well water are usually low in radon, and the distribution site waterborne radon is even lower because the half-life of radon (3.8 days) is less than the time between removal of groundwater and delivery to homes (Brutsaert et al. 1981, Smith et al. 1961). However, our experience in the central Appalachians indicates that well water is commonly well above 300 pCiL$^{-1}$, and that the interval between well water removal and arrival at the distribution sites is usually less than the radon half-life. Of course, for private wells the pumping-to-consumption interval is virtually always less the radon half-life.

The geochemical behavior of radium and radon are rather different (Graves 1988), but radium is the parent radionuclide of radon, so radium enriched water can be enriched in radon (Smith et al. 1961). This close association of radium and radon in terms of both their origin and their carcinogenic behavior serves to complicate health considerations of natural radionuclides. One example, which is probably not atypical, is that internally deposited radium and/or radon induces brain carcinomas (Wrenn et al. 1987). Oech et al. (1987, 1988a, 1988b) noted a correlation between radium and radon in domestic water and the development of lung cancer in a study of several Texas counties. Hess et al. (1982, 1983) noted a correlation between radon in home water supplies in Maine and the incidence of lung cancer. In both the Texas and Maine studies, the cancer producing mechanism is thought to be an outgassing of radon from water, which thereby increases the airborne radon concentration and presumably the rate of lung cancer.

A few studies have been directed toward the possibility of developing cancer by the ingestion of radon enriched water (Cross et al. 1985, Cothren 1987b). Early work indicated that radon in water at a level of about 20,000 pCiL$^{-1}$, assuming an average consumption of about 300 ml/day, would lead to the maximum stomach dose of 0.5 rem/year which was specified by the International Commission on Radiological Protection (Hens 1966). Since levels at or above 20,000 pCiL$^{-1}$ apparently are very rare (Cothren 1987b, Michel and Jordan 1988, Dixon and Lee, 1988), and because airborne radon can be measurably enriched by the outgassing of the home water supply (e.g., Cothren 1987b, Prichard 1987), it has been thought that the cancer risk from the inhalation of radon released from water would almost always exceed the cancer risk that results from the ingestion of radon enriched drinking water.
In contrast to these observations, the following report will show that in a population consuming well water with between 100 and 10,000 pCiL$^{-1}$, the development of cancer is more common as the radon concentration in the drinking water increases.

APPROACH

The Center for Basic and Applied Science has conducted a study of regional indoor radon since 1987 (Mose and others, 1990, 1991). Most of the approximately 1500 study homes are in Prince William and Fairfax County in northern Virginia, and the immediately adjacent Montgomery County in southern Maryland. About half of the participants of our indoor radon project joined our radon-verses-cancer study, and additional homeowners subsequently joined only our radon-verses-geology study.

Arrangements were made with Dr. Ed Vitz of the Waterborne Radon Survey at Ritztown University in Pennsylvania for the radon analyses using liquid scintillation. To gather the measurements of radon in drinking water, the homeowners were provided with an inexpensive syringe, a pair of capped vials containing 5 mL of toluene based liquid scintillation fluid, along with directions about how to collect two 10 mL samples of drinking water from a commonly used water tap. The homeowners filled out a pre-test survey about their family health which asked, in part, the age of each home occupant, the time spent in the home (i.e., when did the person come to the home), and a few questions about cancer (e.g., did any type develop, and if so, when was it discovered and what type).

Although only a single pair of water samples was analyzed for waterborne radon in each home, radon concentration in groundwater is thought to usually show variation below a factor of two (Prichard and Gesell 1981). Precipitation and recharge rates affect the radon concentration in ground water, so radon tends to decrease during high precipitation periods and increase during drought periods. However, precipitation in the study area tends to occur throughout the year. Studies now in progress will serve to determine if the level of groundwater radon variation in the study area is small, but preliminary data suggest that the variation is indeed below a factor of two.

The homeowners who obtained the test kit also filled out a post-test questionnaire about the water supply to the home (e.g., from where is the water obtained, if from a well how deep and how far away is the well, is the water treated, and if so how, etc.). Several studies have noted that well water radioactivity increases with the depth of the well (Smith et al 1961, Bean et al 1982, Cech et al 1988a and 1988b), and we anticipate commenting upon this observation using data from our study. However, the present report is confined to a comparison between radon and cancer, without reference to the well depth and to other physical and chemical properties.

Since there had been no testing of water for its radon concentration in the study area, and there had been almost no news media coverage about radon in water, the data shown later is not biased toward homes for which the homeowner had some prior data from their drinking water. In the introductory letter to all the participants, it was noted that drinking
water can contain radon and radium, that some municipal systems contain these radionuclides, and that private well water often contains at least some radon and radium. This resulted in a data base in which about 70 percent of the homes have municipal water and 30 percent of the homes have private wells; in the study area, the actual portion of homes using private water wells is about 15 percent.

WATERBORNE RADON AND GEOLOGY

Individual geological units in the Appalachians are frequently found over vast areas which were formed by particular geological events. In northern Virginia and the immediately adjacent southern Maryland, five units can be traced over most of the terrane (Figure 1).

The oldest unit is the quartzite of the Blue Ridge, followed by the schist and phyllite. These units developed from sedimentary and volcanic strata deposited in a pre-Atlantic ocean basin about 700 to 450 million years ago. Beach sands were recrystallized into quartzite, and marine sedimentary and volcanic strata were recrystallized into schist and phyllite, and sometimes gneiss, when the pre-Atlantic Ocean disappeared during a continental collision which formed the Appalachian mountains, about 450 to 300 million years ago. One locally famous rock unit in this area is of probable volcanic origin, and now is called ultramafic rock. In Fairfax County, ultramafic rock represents an environmental hazard because it contains asbestos which formed during the recrystallization event. To a great extent, the recrystallization of the terrane was facilitated by intrusions of pre-, syn- and post-recrystallization granitic rocks now found throughout most of the Appalachian mountain system.

Following a long interval of uplift and erosion, narrow basins developed about 200 to 150 million years ago, just prior to the opening of the modern Atlantic Ocean. One of these basins, locally known as the Culpeper Basin, accumulated mainly red terrestrial sandstone and shale. Approximately 150 million years ago, and continuing today, the Atlantic Ocean opened and the western margin known as the Coastal Plain accumulated sand and clay.

Using the questionnaires supplied by participants in our radon studies, approximately 160 homes could be located with enough precision to determine the underlying geological unit. As shown in Table 1, some units appear on average to be twice as radioactive in terms of waterborne radon, compared to other units. Ongoing more detailed studies may provide conclusive answers, but the explanation for this variation may be straightforward. Water with the least radon concentration comes from Blue Ridge quartzite, whose protolith is sandstone, and the Culpeper Basin sandstone. The protolith for sandstone and quartzite is sand, which tends to accumulate in river and beach environments. The high-energy and high-oxygen characteristics of river and beach environments tend to remove uranium, and accumulate quartz which is low in uranium and radium, the precursors of radon. Consequently, the quartzite and sandstone are low in waterborne radon. Sand and clay of the Coastal Plain appears on average to have low to intermediate waterborne radon, perhaps because the sand tends to have lower uranium and radium, and the clay tends to have higher
Figure 1. Geological sketch map of Montgomery County (southern Maryland), the District of Columbia, and Fairfax and Prince William Counties (northern Virginia).
uranium and radium.

Table 1

WATERBORNE RADON CONCENTRATIONS ACCORDING TO GEOLOGICAL UNITS: RADON MEASUREMENTS ARE IN pCiL⁻¹ OF WATER

<table>
<thead>
<tr>
<th>Geological Unit</th>
<th>Number of Wells</th>
<th>Average</th>
<th>Median</th>
<th>Minimum</th>
<th>Maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Granitic Rocks</td>
<td>21</td>
<td>3292</td>
<td>2912</td>
<td>805</td>
<td>6946</td>
</tr>
<tr>
<td>Ultramafic Rocks</td>
<td>3</td>
<td>3045</td>
<td>3434</td>
<td>1415</td>
<td>4287</td>
</tr>
<tr>
<td>Schist/Phyllite</td>
<td>111</td>
<td>2541</td>
<td>2379</td>
<td>&lt; 100</td>
<td>7402</td>
</tr>
<tr>
<td>Sand and Clay</td>
<td>3</td>
<td>2359</td>
<td>1571</td>
<td>&lt; 100</td>
<td>5507</td>
</tr>
<tr>
<td>Sandstone</td>
<td>20</td>
<td>1787</td>
<td>1540</td>
<td>129</td>
<td>4699</td>
</tr>
<tr>
<td>Quartzite</td>
<td>4</td>
<td>1017</td>
<td>860</td>
<td>720</td>
<td>1630</td>
</tr>
</tbody>
</table>

Schist and phyllite have intermediate to high waterborne radon, probably because these units are the host rocks for the granitic rocks. Granite tends to be among the most radioactive of rock types, primarily because it is usually derived by partial melting in the lower part of the mountain system during the recrystallization episode. During the intense recrystallization in zones of partial melting, elements like uranium tend to accumulate in the partial melt, and subsequently are disseminated throughout the molten granite, prior to its crystallization. As the uranium-enriched granite melt moves to higher crustal levels, its radioactive component can escape as vapors which enrich the granite host rock. The reducing environment of the ultramafic rocks tends to accumulate uranium and radium during recrystallization, and this may account for the relatively high waterborne radon concentration in the ultramafic rock.

Table 2

INDOOR (AIRBORNE) RADON CONCENTRATIONS ACCORDING TO GEOLOGICAL UNITS: RADON MEASUREMENTS ARE IN pCiL⁻¹ OF AIR

<table>
<thead>
<tr>
<th>Geological Unit</th>
<th>Number of Homes</th>
<th>Average</th>
<th>Median</th>
<th>Minimum</th>
<th>Maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Granitic Rocks</td>
<td>61</td>
<td>2.9</td>
<td>2.4</td>
<td>0.9</td>
<td>10.1</td>
</tr>
<tr>
<td>Ultramafic Rocks</td>
<td>49</td>
<td>3.7</td>
<td>2.3</td>
<td>0.6</td>
<td>15.8</td>
</tr>
<tr>
<td>Schist/Phyllite</td>
<td>532</td>
<td>4.6</td>
<td>3.5</td>
<td>0.6</td>
<td>68.2</td>
</tr>
<tr>
<td>Sand and Clay</td>
<td>74</td>
<td>2.5</td>
<td>2.2</td>
<td>0.6</td>
<td>7.4</td>
</tr>
<tr>
<td>Sandstone</td>
<td>58</td>
<td>3.7</td>
<td>2.6</td>
<td>0.9</td>
<td>14.8</td>
</tr>
<tr>
<td>Quartzite</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
Figure 2. People without cancer that consumed well water for at least 5 years.

Figure 3. People without cancer that consumed well water for at least 10 years.
As described above, granitic rock tends to be enriched in uranium, and consequently enriched in radon. In most cases, the uranium atoms (and therefore the radium and radon) are mainly located on grain boundaries. When groundwater moves through the granitic rock, radon is relatively easily removed from its grain boundary locations. In the other rock types of the study area, the waterborne radon is less concentrated because the intergranular radioactivity never accumulated (e.g., the sandstone and quartzite), or the radioactivity is chemically held (e.g., schist and phyllite, ultramafic rock). In granite, the radioactivity is present, but easily removed by percolating groundwater.

It is unfortunate that indoor radon of the geological units (Table 2) does not correlate well with waterborne radon (Table 1). Prior to this study, we had hypothesized that since indoor radon mainly originates from the home’s underlying soil and rock, and since the immediately underlying soil and rock are similar to the deeper zone where well water originates, we would expect to find a correlation between indoor radon and waterborne radon. Were the correlation to exist, we could perhaps use aeroradioactivity to predict waterborne radon as we did to predict indoor radon (Mose et al 1990).

Unfortunately, indoor radon does not correlate well with waterborne radon (compare Tables 1 and 2). Continuing studies are designed to confirm or to disprove this observation. However, the correlation may simply not exist. It may be that variations caused by weather and home construction may obscure potential correlations between indoor radon and waterborne radon. It may be that natural chemical changes which occur as rock decomposes to form soil may be important: indoor radon is mainly derived from the upper 10 meters of the Earth’s surface; waterborne radon in Virginia and Maryland wells is probably mainly related to radon concentrations at depths approaching 100 meters.

WATERBORNE RADON AND CANCER

In order to examine the possible relationship between radon in water and cancer, we made the assumption that the body reaches an equilibrium with its normal level of radon (and other radionuclides) consumed by drinking water. We assumed, as discussed in Prichard and Gesell (1981), that radon and other radionuclides act in concert with other carcinogens to produce the cancers. We assumed that radon and other dissolved radionuclides move throughout the body, and while it is perhaps not possible to know what fraction of these radionuclides decay before being removed from the body, this natural radiation could contribute in a measurable way to the development of cancer.

The radon-in-water data reported in this paper are measurements that were accumulated in 1988-1991, using water taps in area homes. In these more than 1000 homes, there about 2,500 occupants. However, data from about 35 homes were not used due to problems in the sampling procedure or in the questionnaire accuracy. All individuals who had lived in their present home for less than 5 years were not counted. Individuals who had smoked tobacco for more than 2 years and had developed lung cancer were
Figure 4. People without cancer that consumed well water for at least 15 years.

Figure 5. People with cancer that consumed well water for at least 5 years.
not counted. Finally, this report is not concerned with the effect of potential carcinogens in treated water supplied by municipal water treatment systems. Consequently, of the approximately 2,500 individuals who participated in our radon-in-water study, the following observations are based on 523 people who used well water. The correlations between cancer and radon level in drinking water are based on tabulations of all types of cancers taken together.

Of the many variables involved in a study of this type, the two most significant are the effect of increasing age, which is associated with higher rates of cancer, and increasing radon in drinking water, which may be also related to higher rates of cancer. To estimate the effect of increasing age, the 523 person study group was divided into subsets, composed of individuals who consumed their present well water for at least 5 years (523 people, of which 3.8% developed cancer), individuals who consumed their present well water for at least 10 years (331 people, of which 3.9% developed cancer), and people who consumed their present well water for at least 15 years (157 people, of which 7.0% developed cancer). For people who had not developed cancer, the at-least-5, at-least-10 and at-least-15 year subsets refer the interval between the present and when the person moved into the home; for people who had developed cancer, the time intervals are between the time the cancer was detected, and when the person moved into the home.

Diagrams presented later in this discussion will show that cancer seems to occur more often in people who consume higher concentrations of radon in their well water. However, to more clearly express the significance of this observation, it is first necessary to determine if a simpler relationship occurs. It is generally known, and seen in our study, that older people tend to develop cancer more frequently than younger people. The "simpler relationship" which should be examined is the possibility that in the study area, older people tend to have more waterborne radon in their well water. For example, it could be hypothesized that since northern Virginia and southern Maryland have been rapidly urbanized over the past 50 years, older citizens tend to consume well water with higher radon levels because, perhaps, the older wells are less efficient (lower productivity wells tend to accumulate water that passes through soil and rock more slowly, and might thereby become more radioactive). If this were true, the cancer- verses-radon relationship presented later might simply caused by the tendency for older citizens to develop cancer, and only coincidentally do these citizens happen to consume well water with higher waterborne radon.

To test the possibility that older citizens might tend to consume well water with higher waterborne radon concentration, a subset was developed using all the study participants who have lived in their present home consuming well water for at least 5 years, and have not developed cancer (Figure 2; 504 people). There does not appear to be any correlation between age and waterborne radon, nor is there any correlation for the without-cancer individuals who have lived in their present home for at least 10 years (Figure 3; 318 people), or for those who have lived in their present home for at least 15 years (Figure 4; 146 people). Our conclusion is that there is no correlation between the radon concentration in well water and the age of the general population that consumes well water.
Figure 6. People with cancer that consumed well water for at least 10 years.

Figure 7. People with cancer that consumed well water for at least 15 years.
A very different picture emerges for the people who have consumed well water, and have developed cancer. The cancers that were counted in this study include many types: bladder, bone, breast, colon, esophagus, gastric, kidney, liver, lung (in non-smokers), lymph nodes, pancreas, prostate, thyroid, skin, and uterus. This approach, in which all cancer types are compiled in radon-vs-cancer compilations, is similar to that of Hess and others (1982, 1983), who found a significant correlation for all cancers together verses county averages for radon in water.

The group of people in our study who have lived in their present home for at least 5 years and have developed cancer also does not show a positive radon-vs-age relationship (Figure 5; 19 people). However, there is a positive radon-vs-age relationship for the group of people who lived in their present home for at least 10 years (Figure 6; 13 people), and this correlation is even more apparent in the group that has lived in their present well-water home for at least 15 years (Figure 7; 11 people). Although the number of individuals is small, these observations are compatible with the hypothesis that waterborne radon is a carcinogen with a measurable effect only after exposure intervals of about 10 years or more, at least at the levels of waterborne radon encountered in the study area.

The unexpected correlation between radon-in-water and the development of cancer may relate to observations made about the rate at which radon is removed from the body, following its inhalation or ingestion. Pohl and Pohl-Ruling (1967) and Lytkken and Ong (1989) have shown that inhaled radon is able to move through the lungs, where it is transported by the blood throughout the body. It is this mechanism which led Henshaw et al (1990) to compare cancers of many types with regional indoor radon levels in several states and cities. Even more recently, Gosink et al (1990) showed that radon consumed in well water has a surprisingly long biologic residence time (e.g., a sedentary or sleeping person would take about 12 hours to eliminate the radon consumed in a drink of Virginia or Maryland well water).

CONCLUSIONS

Our study of indoor radon in Virginia and Maryland shows that the indoor radon concentrations are almost all between 1 and 100 pCiL⁻¹ (Mose and Mushrush 1988). Municipal water supplies obtained from reservoirs provide potable water at less than 100 pCiL⁻¹; radon in drinking water from most private wells ranges from about 100 pCiL⁻¹ to about 10,000 pCiL⁻¹ (Mose et al 1990).

Waterborne radon ranges from an average of about 1000 pCiL⁻¹ to an average of over 3000 pCiL⁻¹ in geological units. A review of radon levels in people who consume well water but have not developed cancer indicates older people, who are generally know to more commonly develop cancer, do not tend to have high-radon water wells. However, the effect of higher radon-in-water concentrations is shown in the subset that developed cancer.

It now appears that the ingestion of radon enriched water is a health concern, but much remains unclear. Both radon and radium carried by ingested water can move throughout the body, to produce an effect at many
cancer prone sites. Studies cited earlier indicate that radium can be a
cause of many types of cancer, and that radium and radon are often both
found in groundwater. It is possible that radon is merely a "flag" for
radium or some other groundwater component, but a useful flag because the
measurement of dissolved radon is quick and inexpensive. On the other
hand, it may be that the carcinogen is simply radon. Presumably the radon,
as well as the other carcinogens, are all at some equilibrium
concentration in the body, but the radon can more easily enter into well
water, and is often 10 to 1000 times more concentrated than radium.

As in most investigations, thoughtful precautions and additional data
are often useful. In our study, we provide indoor radon monitors and we
encourage homeowners with private water supplies to obtain a measurement
of the radon concentration in their water. In homes with an indoor radon
concentration that the homeowner views as representing an unreasonable
risk, we suggest that they reduce the airborne radon with the use of
sub-slab ventilation. Similarly, in homes with unreasonably high radon
concentrations in the drinking water, we suggest the installation of a
charcoal water purification system. Unfortunately for the general
population, the perception of what represents an unreasonable risk depends
on many factors, only one of which is advice. At this stage of public
awareness policy, airborne radon is clearly more discussed than waterborne
radon. We suggest, pending the gathering of additional data of the type
compiled in this report, that radon in both air and water be equally cited
by public health officials.

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