Ultrafine particles and nitrogen oxides generated by gas and electric cooking

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Abstract

Objectives—To measure the concentrations of particles less than 100 nm diameter and of oxides of nitrogen generated by cooking with gas and electricity, to comment on possible hazards to health in poorly ventilated kitchens.

Methods—Experiments with gas and electric rings, grills, and ovens were used to compare different cooking procedures. Nitrogen oxides (NOx) were measured by a chemiluminescent ML9841A NOx analyser. A TSI 3934 scanning mobility particle sizer was used to measure average number concentration and size distribution of aerosols in the size range 10–500 nm.

Results—High concentrations of particles are generated by gas combustion, by frying, and by cooking of fatty foods. Electric rings and grills may also generate particles from their surfaces. In experiments where gas burning was the most important source of particles, most particles were in the size range 15–40 nm. When bacon was fried on the gas or electric rings the particles were of larger diameter, in the size range 50–100 nm. The smaller particles generated during experiments grew in size with time because of coagulation. Substantial concentrations of NOx were generated during cooking on gas; four rings for 15 minutes produced 5 minute peaks of about 1000 ppb nitrogen dioxide and about 2000 ppb nitric oxide.

Conclusions—Cooking in a poorly ventilated kitchen may give rise to potentially toxic concentrations of numbers of particles. Very high concentrations of oxides of nitrogen may also be generated by gas cooking, and with no extraction and poor ventilation, may reach concentrations at which adverse health effects may be expected. Although respiratory effects of exposure to NOx might be anticipated, recent epidemiology suggests that cardiac effects cannot be excluded, and further investigation of this is desirable.

Keywords: cooking fuels; nitrogen oxides; ultrafine particles

Many studies have now shown associations between the airborne concentrations of particles and cardiorespiratory ill health. Most have focused on the mass of particulate matter less than either 10 or 2.5 µm in aerodynamic diameter (PM10 and PM2.5). It has been hypothesised that these effects may be due to the nm sized particles (ultrafine particles (UFPs)) comprising the largest number of particles, rather than the mass which is principally determined by larger, greater than 1 µm sized particles. This implies that particle numbers could be a better metric than particle mass for predicting health effects and for control purposes. This ultrafine hypothesis was based on animal studies that showed nm sized particles to be toxic, whereas larger particles of the same material were not. It was recognised that most studies relating health effects to particulate pollution are based on PM10 measured in urban air, where UFPs generated by combustion are an important component. Relatively few studies have so far tested the ultrafine hypothesis, although there is toxicological evidence in support of the earlier studies showing that both small size and adsorbed transition metals may contribute to toxicity. Epidemiologically, to date studies of associations between the number and mass concentrations of particles in the outdoor air and health effects have given conflicting results, suggesting that number of UFPs may be more relevant to cardiac effects.

As well as this there is evidence of associations between exposure to nitrogen dioxide (NO2) and adverse health effects. At high concentrations NO2 may increase sensitivity to allergens in asthmatic people, whereas at ambient concentrations it also has been associated with exacerbations of asthma and increased death rates. Most recently, it has been associated with episodes of cardiac arrhythmia in subjects with implanted defibrillators. This evidence is not wholly consistent, and as there are close correlations between NO2 in the general environment and other pollutants derived from traffic, a question remains as to whether these low level associations may be due to confounding.

Most people spend at least 90% of their time indoors and the question therefore arises as to how relevant air pollution generated indoors is to adverse health effects. The most important indoor activities generating particles are smoking and cooking. Several studies have investigated relations between cooking and respiratory symptoms, and some have shown that children who live in houses where gas is used have more respiratory symptoms than children who live in houses where other cooking fuels are used. Jarvis et al also showed an association between gas cooking and aggravation of asthmatic symptoms. These effects have generally been attributed to increases in concentrations of NO2. However, Garrett et
al\textsuperscript{21} concluded that exposure to gas cooking was an important risk factor for respiratory symptoms even after adjusting for concentrations of NO\textsubscript{x}, suggesting an additional hazard associated with the use of gas stoves. Recently we reported in this Journal that PM\textsubscript{e}, generated by gas cooking has the potential to cause proinflammatory effects in lung cells.\textsuperscript{20}

In this paper, we present the results of studies of the generation of UFPs and nitrogen oxides (NO\textsubscript{x}) during different cooking procedures, and discuss the possibility that such pollutants may contribute to the health effects found in people who use gas for cooking.

Methods

LABORATORY
The study was conducted in a laboratory (volume 70 m\textsuperscript{3} and internal surface area about 130 m\textsuperscript{2}) in Aberdeen. No mechanical ventilation was used during the studies and the windows were closed during all measurements. The only potential sources of particles present in the laboratory were a gas and an electric cooker, both with four rings, an eye level grill, and an oven. The rings of the electric cooker were closed metal plates.

NITROGEN OXIDES
The concentrations of nitric oxide (NO) and NO\textsubscript{x} (NO+NO\textsubscript{2}) were measured with a chemiluminescent ML9841A NO\textsubscript{x} analyser. The NO\textsubscript{x} was obtained by subtracting NO from NO\textsubscript{2}. Concentrations of NO\textsubscript{2} were measured at face level in front of the cookers. The lower detection limit of the monitor was 0.5 ppb. The instrument was calibrated with certified gas probes for NO\textsubscript{2}. Data were stored every 5 minutes in an internal memory, downloaded, and exported into Microsoft Excel.

ULTRAFINE PARTICLES
A TSI 3934 scanning mobility particle sizer was used to measure average concentrations of numbers and size distribution of aerosols in the size range 10–500 nm. Particles were electrically charged to a known charge distribution with a bipolar charger in a Model 3071A electrostatic classifier. The particles were then classified according to their ability to traverse an electric field, and counted with a Model 3022A condensation particle counter. Sampling was through straight copper tubing, a method that we had shown previously to result in minimal loss of particles.\textsuperscript{12} The system is automated with a personal computer and data were downloaded and exported in a Microsoft Excel spreadsheet format. Samples were taken in two 2.5 minute scans and the scanning mobility particle sizer system calculated 5e minute averaging periods, which were used in processing the data. The scanning mobility particle sizer system was calibrated by Bristol Industrial Research Associates. All test and calibration data have been obtained with standards, the accuracies of which are traceable to a nationally recognised standards laboratory, or have been verified by instrumentation, the accuracy of which is traceable to a nationally recognised standards laboratory, or is derived from accepted values of physical contents. Particle number concentrations were measured at the same place as the NO\textsubscript{x}, at face level in front of the cookers. The UFP concentration in this paper is presented as the number of particles between 10 nm and 100 nm. The particle numbers in the size range 10–500 nm (which are divided in 53 size ranges by the scanning mobility particle sizer system) were used to assess the ranges of the particle sizes.

COOKING EXPERIMENTS
All cooking experiments were carried out between March and December 1999. The experiments with the gas and the electric cooker were all performed the same way, comparing effects of the two cooking fuels. For every cooking experiment (use of rings, oven, and grill) both fatty (bacon) and low fat (bread, potato, or vegetable) foods were cooked. The different methods carried out are presented in table 1.

Table 1 The cooking experiments

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Time (minutes)</th>
<th>Power</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rings:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 Ring</td>
<td>15</td>
<td>Full power</td>
<td></td>
</tr>
<tr>
<td>4 Rings</td>
<td>15</td>
<td>Full power</td>
<td></td>
</tr>
<tr>
<td>Boil water</td>
<td>15</td>
<td>Full power</td>
<td>Water 1 l</td>
</tr>
<tr>
<td>Stir fry</td>
<td>5</td>
<td>Full power</td>
<td>Vegetables 500 g</td>
</tr>
<tr>
<td>Fry bacon</td>
<td>7</td>
<td>Full power</td>
<td>Bacon 4 rashers</td>
</tr>
<tr>
<td>Oven:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bake cake</td>
<td>40</td>
<td>180°C</td>
<td>Sponge cake</td>
</tr>
<tr>
<td>Roast meat</td>
<td>75</td>
<td>180°C</td>
<td>Turkey joint 500 g</td>
</tr>
<tr>
<td>Bake potatoes</td>
<td>75</td>
<td>180°C</td>
<td>Potatoes 2, total 500 g</td>
</tr>
<tr>
<td>Grill:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Grill only</td>
<td>15</td>
<td>Full power</td>
<td></td>
</tr>
<tr>
<td>Toast</td>
<td>5</td>
<td>Full power</td>
<td>Bread 2 slices</td>
</tr>
<tr>
<td>Grill bacon</td>
<td>10</td>
<td>Full power</td>
<td>Bacon 4 rashers</td>
</tr>
</tbody>
</table>

LOSESSES OF OXIDES OF NITROGEN AND PARTICLES FROM THE ROOM AIR
The loss of NO\textsubscript{x} and UFPs was estimated from the decay curve for each pollutant at the end of a period of cooking and presented as the time in minutes it took for the concentration to fall by 50%. Loss rates were only estimated if there was a clear rise in the concentration of pollutant during the cooking experiment together with a decay curve that did not show any irregular pattern. For NO\textsubscript{x}, the loss was from a combination of air exchange and adsorption onto surfaces.\textsuperscript{24} Loss of UFPs was assumed to be due to exchange with the outdoor air, deposition on room surfaces, and coagulation. Information on the deposition velocity for UFPs was not available, but it was assumed that this makes a relatively small contribution. The loss of concentration of particle numbers from coagulation is dependent on the square of the concentration, which means that it will be relatively more important at the end of a period of cooking when the UFP concentration is highest.\textsuperscript{25}

STATISTICAL ANALYSES
The results of the concentrations of particle numbers and NO\textsubscript{x} during cooking are presented as the maximum 5 minute concentration that occurred during the cooking method.
above the baseline concentration. Baseline concentrations were calculated by taking the average concentration of eight measurements; four just before and four just after the experiment and four after the experiment when the rise in particles or NO was gone back to a steady level. For comparison between concentrations of particle numbers and NO, generated during different cooking methods, the non-parametric Mann-Whitney test was used. A p value 0.05 or less was taken as significant.

Results

COOKING EXPERIMENTS

Numbers of UFPs and concentrations of NOx

The peak concentrations of number of UFPs for the different cooking methods are presented in table 2. In general, cooking with gas generated more particles than did electricity. Table 3 presents the NOx concentrations generated during gas cooking. When the electric cooker was used, there was no rise in NOx. When gas rings were fully turned on, concentrations of both number of UFPs and NOx rose rapidly (fig 1). Four gas rings burning resulted in a rapid rise in UFPs to on average 150 000 UFP/cm³ and a rise in NOx and NOx respectively on average to about 1000 ppb and about 2000 ppb. The gas grill alone did not generate detectable concentrations of NO or NOx. One electric ring without anything on it resulted in a rise in numbers of UFPs; however a pot with water on the ring never resulted in a rise in numbers of UFPs. Four electric rings on full power did result in all occasions in a rise in UFPs to about 110 000 UFPs/cm³.

Frying bacon on the gas rings caused the highest peak concentration of numbers of UFPs, 500 000 UFPs/cm³. The peak concentration of UFPs generated by frying bacon on gas was significantly higher than that by frying vegetables on gas (p=0.006). Frying bacon on gas also resulted in a significantly higher peak concentration of UFPs than frying bacon on an electric ring (p=0.006).

Use of the gas oven generated more UFPs than use of the electric oven. The gas oven also generated high peaks of NOx. For example, cooking potatoes for 75 minutes resulted in NO peak concentrations of about 370 ppb and NOx peaks of about 1000 (table 3). There was a progressive rise in NOx and NO until the oven was turned off (fig 2) and a steady concentration was not reached. The particle concentration did, however, reach a plateau at about 120 000 UFP/cm³.

To investigate whether the rises in UFPs and NOx in these short term experiments were caused mainly by ignition (which was electronic) or would continue with continued combustion, four gas rings were kept on full power for 2 hours. After an initial rapid rise in particles, the concentration declined. However, particles were still being generated after this initial rise while the gas was still burning, as a much steeper fall in concentration of particle numbers occurred after the rings were turned off (fig 3). The NOx reached steady concentrations after the rings had been on for 75 minutes, with NOx concentrations of 2200 ppb and NO concentrations of 5800 ppb. In our experiments the NO concentrations were always higher than those of NOx, during the use of the gas rings and oven. This lasted for at least 2 hours after the cooking experiments were finished (fig 3).

Loses of oxides of nitrogen and particles from the room air

Loses of NOx and UFPs were measured for 23 experiments, where both showed a clear rise due to the experiments. The measured mean (SD) 50% loss rate for NOx was 67 (27) minutes. The mean (SD) 50% loss rate for UFPs was twice as fast, at 30 (10) minutes. The reason for the higher loss rate for UFPs is likely to be coagulation of the particles.

PARTICLE SIZES

The peak particle sizes described below are the mode of the UFP number distribution.

Table 2 The maximum 5 minute UFP numbers above baseline, and the particle size at peak concentration for each cooking method

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Cooking fuel</th>
<th>n*</th>
<th>Peak concentration above baseline of UFPs/cm³</th>
<th>Particle size (nm) at peak concentration mean (SD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rings:</td>
<td></td>
<td></td>
<td>Mean (SD) ×10⁴</td>
<td>Mean (SD)</td>
</tr>
<tr>
<td>1 Ring Gas</td>
<td>4</td>
<td>2.6 (1.2)</td>
<td>16 (13)</td>
<td></td>
</tr>
<tr>
<td>4 Rings Electric</td>
<td>5</td>
<td>9.4 (7.2)</td>
<td>32 (14)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>14.6 (8.3)</td>
<td>26 (12)</td>
<td></td>
</tr>
<tr>
<td>Boil water Gas</td>
<td>4</td>
<td>13.3 (8.1)</td>
<td>17 (5)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>—‡</td>
<td>—‡</td>
<td></td>
</tr>
<tr>
<td>Stir fry Gas</td>
<td>5</td>
<td>13.7 (12.6)</td>
<td>41 (8)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>11.1 (10.4)</td>
<td>37 (13)</td>
<td></td>
</tr>
<tr>
<td>Fry bacon Gas</td>
<td>5</td>
<td>15.9 (5.0)</td>
<td>72 (20)</td>
<td></td>
</tr>
<tr>
<td>Oven:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bake cake Gas</td>
<td>2</td>
<td>9.8 (0.9)</td>
<td>34 (1)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>3.0 (0.1)</td>
<td>38 (6)</td>
<td></td>
</tr>
<tr>
<td>Roast meat Gas</td>
<td>2</td>
<td>12.4 (2.0)</td>
<td>31 (7)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>2.4 (1.4)</td>
<td>23 (6)</td>
<td></td>
</tr>
<tr>
<td>Bake potatoes Gas</td>
<td>3</td>
<td>12.5 (5.5)</td>
<td>39 (2)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>4.6 (1.2)</td>
<td>46 (4)</td>
<td></td>
</tr>
<tr>
<td>Grill:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Grill only Electric</td>
<td>5</td>
<td>10.3 (12.4)</td>
<td>24 (6)</td>
<td></td>
</tr>
<tr>
<td>Toast Gas</td>
<td>5</td>
<td>7.7 (11.1)</td>
<td>20 (7)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>13.8 (13.9)</td>
<td>28 (6)</td>
<td></td>
</tr>
<tr>
<td>Grill bacon Gas</td>
<td>4</td>
<td>13.4 (4.0)</td>
<td>27 (5)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>41.3 (10.8)</td>
<td>39 (7)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>53.0 (3.8)</td>
<td>53 (9)</td>
<td></td>
</tr>
</tbody>
</table>

*Fewer replicated oven experiments were able to be carried out owing to a temporary failure of the SMPS system.
†Based on three of the five experiments, as two of the experiments did not show a rise in particle numbers.
‡No change in concentration due to the cooking method.
§Not possible to give one size (multiple peaks).
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Discussion

Many epidemiological studies have now shown relations between exposure to outdoor particulate air pollution and ill health. Similar relations have also been shown with NO₂. In both cases these associations seem to be present even at what are, in conventional toxicological terms, very low concentrations. Moreover, they occur despite the fact that most people spend at least 90% of their time indoors. These findings raise two important issues. Firstly, if the associations are causative, other sources of particles and NO₂ would be expected to influence the health of exposed people. We therefore asked ourselves what concentrations might arise in kitchens relative to those outside. Secondly, if total exposure to the toxic substances is an important determinant of effects, a proportion of this is likely to occur indoors. Further refinement of epidemiological studies is likely to require modelling of individual exposures, and it will therefore be necessary to know the contribution to these of indoor exposures. The present study was therefore also planned as a first step towards such modelling and also as part of a study to determine whether particles and gases generated indoors have the same toxicity as those outdoors.

We found that many UFPs are produced not only by gas combustion but also from heating electric plates and grills. Cooking with electricity or gas has the potential to add to particle numbers, mainly through frying and grilling of fatty foods and through the use of fats for frying. Particles generated by gas combustion are smaller in size than particles generated by cooking fatty food. Two recent studies have also investigated concentrations of particle numbers indoors. Wallace identified the gas oven, gas burners, and the electric toaster as being the most important sources of UFPs. Abt

measured during the experiments (table 2). Particles produced by the gas rings alone and by boiling water on gas had a peak diameter in the range 15–40 nm. The peak was in the same size range for the particles that were produced when the gas grill alone was turned on and when electric rings were turned on without a pan. When bacon was fried on the gas or electric rings the particles were larger, most particles being in the size range 50–100 nm. Immediately after the gas rings were turned on, the highest numbers of particles were in the very fine size range, but thereafter the particles grew in size with time. Figure 4 shows the size range of particles generated while four gas rings were on for 2 hours. Five, 15, and 30 minutes after the rings were turned on, the peak concentration was respectively at 15, 30, and 40 nm diameter. Thus, while the gas rings were burning, the particles grew up to a peak of about 50–70 nm. The only situation in which particles did not grow with time was in some of the bacon frying experiments, when the initial particle size was already relatively large, up to 80 nm. The most plausible explanation for the findings is coagulation of the very small particles into larger agglomerates.

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et al concluded that cooking, including grilling or baking, toasting, and barbecuing, contributed primarily to particles with diameters 20–500 nm. Frying was also found to release particles in this size range. Although no experimental studies of human exposure to indoor UFPs have been undertaken, the concentrations found indoors are as high as indoor UFPs have been undertaken, the experimental studies of human exposure to release particles in this size range. Although no experimental studies of human exposure to indoor UFPs have been undertaken, the concentrations found indoors are as high as those found outdoors during episodes of high air pollution, when people have been shown to demonstrate health effects.

The tiny particles produced during burning of gas are probably carbonaceous. We have shown evidence that the original very small particles are likely to agglomerate, thus reducing the overall number as time passes. Small particles are also generated when the electric rings are turned on without a pan. These might have been deposited matter burnt off the ring, metal from the rings themselves, or both. This type of emission does not occur when a pot of water is put on the ring, and we assume that this is a consequence of the heat of the ring being conducted to the water so that material on the ring does not burn off.

Any activity involving heating of fatty foods, not surprisingly, gave rise to high concentrations of particles, although the higher temperatures reached quickly by gas seemed to be responsible for a greater rise with this mode of cooking. Siegmann et al have measured the size distribution of the aerosol produced from heating rape seed oil at different temperatures. They showed that diameter and concentrations of numbers increased with increasing temperature. The mean droplet diameter ranged between 30 nm at 223°C and 100 nm at 256°C. The concentration of numbers of particles with a diameter less than 100 nm rose rapidly with temperature, the oil at 256°C releasing about twice as many particles than the oil at 223°C.

Our studies do not represent the personal exposures of people working in a kitchen, as the sampling took place at a fixed place at head height in front of the cookers, and concentrations measured here are likely to overestimate the exposures of a cook. Moreover, we did not actively ventilate the laboratory during the experiments and there was no extractor fan present. However, when frying bacon and stir frying vegetables, the concentrations measured during the cooking experiments should be close to the actual exposure of a person cooking.

The current United Kingdom standard for ambient air NO2 is based primarily on experimental exposure studies in humans, which have shown subtle effects on inflammatory markers and increased susceptibility to allergens at concentrations around 200 ppb. Epidemiological studies have shown effects on populations at lower concentrations, and a recent panel study has suggested that vulnerable people with cardiac disease may have effects at ambient concentrations below 40 ppb. If NO2 or conceivably NO, is responsible for these effects, a kitchen where concentrations of NO2 can be relatively high might be an important source of exposure. Concentrations of NO in homes have been investigated in several studies and internal sources tend to be the dominant contributor to personal exposure, the most important being environmental tobacco smoke and gas cooking. In this study we have shown that using the gas rings and oven generated substantial amounts of NO2, certainly of a concentration that can cause ill effects in humans, for example by increasing susceptibility to allergens and causing airway inflammation. During gas cooking background concentrations increase dramatically and short term indoor concentrations are higher than are reached outdoors. In Aberdeen, during 1999, the highest hourly outside NO2 and NO concentrations were respectively 63 ppb and 357 ppb, and such concentrations were only reached rarely. Concentrations of NO2 are monitored at 83 national network sites across the United Kingdom and in 1998 the highest 99.8th percentage value of hourly concentrations was 140 ppb at a kerbside site in Glasgow. We found hourly NO2 concentrations of more than 300 ppb with two gas rings on for 15 minutes. This compares with a concentration of 260 ppb for 30 minutes associated with increased response to allergen inhalation in the study of Strand et al. Other short term studies have shown similar effects.

It is known that nitric oxide in the outdoor air rapidly disappears due to its reaction with ozone to form NO2. Indoors, however, we found that concentrations of NO exceeded those of NO2 for at least 2 hours after our experiments on the gas cooker and oven (fig 1 and 2). Farrow et al hypothesised that diarhoea (that was significantly associated with NO2 concentrations) might in fact have been due to exposure to high concentrations of NO. It is even possible that prolonged exposure to NO by influencing natural production of NO by endothelial cells, could be responsible for some of the cardiac effects associated

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epidemiologically with NO$_2$; no studies are known to have investigated this hypothesis further.

As well as carbon particles and NO$_2$, cooking is associated with the generation of fatty particles, which might themselves be a health hazard. Several studies have shown aerosols from cooking oils to be mutagenic and genotoxic, possibly contributing to the raised incidence of lung cancer found among Taiwanese and Chinese women, not entirely explicable by exposure to cigarette smoke. Recently, Ko et al. published details on cooking conditions and habits of 131 non-smoking incident cases of newly diagnosed lung cancer in women and found that the risk of lung cancer increased with the number of meals cooked each day, with not using a fume extractor, and with waiting until fumes were emitted from the oil before they began cooking. In our study we found generation of UFPs to be higher for gas cooking, probably due to the higher immediate temperature the oil is exposed to.

In summary, cooking in a poorly ventilated kitchen may give rise to potentially toxic concentrations of particles. If gas is used, high concentrations of NO$_2$ are also generated, which if not adequately extracted could result in risks to respiratory and possibly cardiac health of exposed people. Further studies of the relations between indoor exposure to NO$_2$ and cardiac events are necessary.

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