

Natural Gas and Indoor Air Pollution: A Comparison With Coal Gas and Liquefied Petroleum Gas

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Objective The study was designed to compare the combustion products of coal gas, liquefied petroleum gas and natural gas in relation to indoor air pollution. **Methods** Regular pollutants including B(a)P were monitored and 1-hydroxy pyrene were tested in urine of the enrolled subjects. Radon concentrations and their changes in four seasons were also monitored in the city natural gas from its source plant and transfer stations to final users. To analyze organic components of coal gas, liquefied petroleum gas and natural gas, a high-flow sampling device specially designed was used to collect their combustion products, and semi-volatile organic compounds contained in the particles were detected by gas chromatograph-mass spectrograph (GC/MS). **Results** Findings in the study showed that the regular indoor air pollutants particles and CO were all above the standard in winter when heating facilities were operated in the city, but they were lowest in kitchens using natural gas; furthermore, although NO₂ and CO₂ were slightly higher in natural gas, B(a)P concentration was lower in this group and 1-hydroxy pyrene was lowest in urine of the subjects exposed to natural gas. Organic compounds were more complicated in coal gas and liquefied petroleum gas than in natural gas. The concentration of radon in natural gas accounted for less than 1‰ of its effective dose contributing to indoor air pollution in Beijing households. **Conclusion** Compared to traditional fuels, gases are deemed as clean ones, and natural gas is shown to be cleaner than the other two gases.

Key words: Natural gas; Indoor air pollution; Organic compounds; Radon; 1-hydroxy pyrene; Environmental monitoring

INTRODUCTION

China is abundant in resources of natural gas (NG), which with its high-energy value has become one of the increasingly growing sources of domestic energy in this country. Up to the end of 1993, NG users increased to 240 000 households and its supply volume reached $1.5 \times 10^9 \text{ m}^3/\text{a}$; it was expected to rise to $8.5 \times 10^9 \text{ m}^3/\text{a}$ by the year of 2000.

The health impact of NG has been widely reported in foreign literature, largely in regard with consequences of the pollutants produced in the process of its exploitation, processing and refining, for instance, acute intoxication, ill health of the population and rising morbidities of respiratory diseases and cancers.

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Biographical note of the first author: Yue ZHANG, female, born in 1953, senior technical scientist with extensive experience in toxicological and genetic assessment of environmental pollutants.

The health consequences of NG as a domestic energy source might well be focused on its impacts on living environment, or indoor air pollution. Over the recent years, both national and international scientific communities have taken great interest in indoor air pollution as a major public health problem. However, no impacts of NG on indoor air pollution have so far been systematically reported in either national or international literature.

MATERIALS AND METHODS

Monitoring of Indoor Air Pollution

Twelve families living in apartment houses were selected and each four of them used NG, coal gas (CG) and liquefied petroleum gas (LPG) respectively. They were monitored for 3 successive days in 3 separate groups in summer (between June 24 and July 4, 1992) and winter (between December 10 and 19, 1992). To avoid interference with cooking smoke in kitchens, samples were collected twice in the morning and in the afternoon by closing all doors and windows and turning off kitchen ventilators. During the sampling kitchen stoves were turned on at an angle of 45° and kettles with water were then placed on them for half an hour to keep the water at an un-boiling state.

Inhalable particles (IP), CO, SO₂, NO₂, CO₂ and B(a)P were used as indicators of indoor air pollution, and their respective sampling methods are shown in Table 1^[1].

TABLE 1
Methods of Sampling and Analysis

Indicators	Sampling Devices	Method of Analysis
IP	KC 8301 Sampling Device	Conventional Method
CO	Plastic Bags	Analyzed by Ecolyzer
SO ₂	Absorbent Sodium Mercuric Tetrachloride	Spectrophotometry With Para-rosaniline Hydrochloride
NO ₂	Ethylene Naphthalene Hydrochloride	Spectrophotometry
CO ₂	Portable Infrared Analyzer GXH-3010C (Only in Winter)	
B(a)P	Air and Particles Were Collected by PUF and Glass Filter (GF) Collectors	PAHs Were Analyzed After Extraction by Inverse Phased-high Pressure Liquid Chromatography (HPLC) and Fluoroscopy

Organic Components of Gas Combustion Products

Air samples in the households exposed to the three respective types of gases were collected in large quantity by high flow collectors. The samples containing particles produced in the process of combustion of the 3 gases, controls and blank filter membranes were separately placed in 250 mL soxhlet extractors. To improve extraction rates all the samples were immersed in dichloromethane for 20 h at room temperature before they were extracted at 70°C-80°C for 12 h. The extracts were dried to constant weight by anhydrous calcium chloride after they were processed by K-D concentrator. The contents of organic compounds on the particles as products of gas combustion were calculated according to the weight of the samples, blank filters and extracts (gravimetric method). The extracts which

had been solved into dichloride methane to reach an appropriate concentration were analyzed by GC/MS (Finnigan-MAT 4510)^[2].

Determination of Radon Concentrations in NG in Beijing

Natural gas in Beijing is originated largely from Renqiu Oil Field located in north China. To have a clear picture of radon (Rn) concentrations in different oil wells and their possible changes in transportation processes, NG samples from 5 source wells, as well as from its exits, entries to Beijing and final users were detected respectively. To identify changes of Rn concentrations over time, NG samples were collected from final users at 7:00-7:30 a.m., 11:30-12:00 a.m. and 6:00-6:30 p.m. for a total of 8 days (two days per week) beginning from March 1992, thereafter from March 1992 to February 1993, gas samples were collected from final users at 11:30-12:00 on one day randomized in the middle of each month, and Rn concentrations were determined by FD-125 indoor Rn analyzer.

Determination of 1-hydro Pyrene in Subjects' Urine

From each of the respective 4 households using different types of gases as their cooking fuel, the family members doing most cooking were defined as study subjects and concentrations of 1-hydro pyrene were determined in their urine samples.

Pretreatment of urine samples: Urine samples of 10 mL were added into 5 mL 0.5 mol/L acetate buffer (pH 5) containing 1 000 units of glucuronidase (type H-1 Sigma Corp. USA). The mixtures were then kept at 37°C for 1 h and isolated after hydrolyzation by SEPPAK C 18 column (Wates Corp). They were concentrated at 60°C up to 0.5 mL after elution by methanol and then tested for 1-hydro pyrene by HPLC.

Expression of 1-hydro pyrene in urine: as the samples tested were not the whole urine within 24 h, the following two correcting methods were adopted to eliminate possible errors derived from urine samples themselves: creatine correction- corrected by creatine values in the same sample and expressed by $\mu\text{mol/L}$ 1-OH-pyrene/mol creatine; and gravity correction-corrected by the Chinese standard gravity of urine (1.020).

RESULTS

Monitoring of Indoor Air Pollutants

As shown in Table 2, IP and B(a)P concentrations in the NG, CG and LPG were higher in winter than in summer, but not significantly different in summer. IP concentrations were all around $150 \mu\text{g}/\text{m}^3$, being little higher in the CG group. In winter IP concentrations exceeded hygienic standards in all the three groups, being highest in the LPG group ($317 \mu\text{g}/\text{m}^3$). As for IP concentrations, there was generally no significant difference in the 3 groups, either in winter or in summer. In contrast, great seasonal differences were observed for B(a)P concentrations on particles, by several times and even dozens of times higher in winter than in summer, while the lowest was in NG and the highest in LPG.

As shown in Table 3, CO concentrations were higher in winter than in summer, and higher in kitchens than in living rooms. They were much higher than the standard either in winter or in summer, notably in the former. If the 3 types of gas were compared, CO concentrations in NG and CG were similar while those in LPG were markedly higher than in the others. SO₂ concentrations were quite low, at the normal range either in the 2 seasons or in all the 3 groups, and were only slightly higher than the standard in winter in the CG group possibly due to sulfur contents in coal gas. NO₂ concentrations were determined only in

winter and were low in all the 3 groups, indicating that indoor NO₂ came merely from kitchen stoves and its differences in winter and summer were insignificant and irregular. Although the highest NO₂ concentrations were found in kitchens using LPG in summer, they were relatively higher even in kitchens using NG in winter. They were somehow lower in the CG group, but were higher than the standard in the 3 groups. Since nitrogen is always present in natural gas, it should be given serious attention in the studies concerning the relationship between indoor air pollution and natural gas.

TABLE 2
IP and B(a)P Indoor Concentrations in NG, CG and LPG

Types of Gas	Households	IP (mg/m ³)		B(a)P (ng/m ³)	
		Summer	Winter	Summer	Winter
NG	1	0.185	0.253	0.81	10.10
NG	2	0.165	0.272	0.99	10.31
NG	3	0.098	0.298	1.17	13.00
NG	4	0.134	0.145	2.16	21.74
$\bar{x} \pm s$		0.14 ± 0.033	0.242 ± 0.058	1.28 ± 0.52	13.79 ± 4.73
CG	1	0.089	0.121	1.151	12.02
CG	2	0.178	0.209	2.71	19.86
CG	3	0.261	0.266	1.97	17.00
CG	4	0.159	0.189	1.90	15.91
$\bar{x} \pm s$		0.17 ± 0.061	0.196 ± 0.052	2.02 ± 0.43	16.20 ± 2.81
LPG	1	0.097	0.250	2.46	18.40
LPG	2	0.091	0.356	3.67	36.19
LPG	3	0.167	0.328	2.64	14.37
LPG	4	0.153	0.334	4.79	41.25
$\bar{x} \pm s$		0.12 ± 0.033	0.317 ± 0.040	3.39 ± 0.93	27.55 ± 11.40

TABLE 3
CO, SO₂ and NO₂ Concentrations in Indoor Air in the 3 Groups

Type of Gas	Sampling Sites	CO		SO ₂		NO ₂	
		Summer	Winter	Summer	Winter	Summer	Winter
NG	K, Average	7.1	10.8	0.10	0.04	0.64	0.87
NG	K, Range	3.8-12.5	1.9-16.3	0.09-0.13	0.02-0.20	0.23-1.43	0.15-3.04
NG	L.R., Average	3.5	5.2		0.03		0.10
NG	L.R., Range	1.4-10.0	1.9-12.5				
CG	K, Average	6.5	10.9	0.11	0.74	0.38	0.23
CG	K, Range	4.4-10.6	6.3-32.5	0.07-0.22	-0.07-2.69	0.10-0.39	0.07-0.38
CG	L.R., Average	2.7	6.9		0.15		0.04
CG	L.R., Range	1.9-5.0	3.8-15.0				
LPG	K, Average	20.3	21.8	0.11	0.02	0.93	0.42
LPG	K, Range	5.0-56.3	8.8-48.8	0.10-0.34	0.10-0.07	0.15-2.19	0.13-0.85
LPG	L.R., Average	5.4	10.3				0.04
LPG	L.R., Range	2.5-6.3	5.0-31.3				

Note. K: Kitchens; L.R.: Living rooms.

As shown in Table 4, CO₂ concentrations were lowest either in living rooms or in kitchens before the stoves were turned on, but they were steadily rising above the standard along with stove operation (0.1% in public places). They were the highest in the NG group and declined gradually after the stoves were turned off.

TABLE 4

CO₂ Concentrations in Indoor Air (%)

Time of Testing (min.)	Sites of Testing	LPG			CG			NG		
		X	N	Range	X	N	Range	X	N	Range
Before Fire on	K.	0.091	3	0.072-0.120	0.055	2	0.042-0.069	0.138	5	0.070-0.210
Before Fire on	L.R.	0.071	5	0.064-0.083	0.060	1	0.060	0.166	4	0.085-0.240
5-15 min on Fire	K.	0.125	3	0.055-0.220	0.130	2	0.119-0.142	0.254	4	0.120-0.420
15-30 min on Fire	K.	0.228	5	0.090-0.230	0.182	3	0.160-0.199	0.431	7	0.250-0.720
30-45 min on Fire	K.	0.336	5	0.200-0.500	0.198	9	0.180-0.240	0.607	9	0.250-0.790
30-45 min on Fire	L.R.	0.136	3	0.119-0.170	0.078	4	0.058-0.101			
5-15 min off Fire	K.	0.207	4	0.170-0.280	0.145	3	0.122-0.168	0.350	7	0.130-0.700
15-30 min off Fire	K.	0.123	3	0.120-0.128	0.118	3	0.105-0.134	0.333	5	0.118-0.590
30-45 min off Fire	K.	0.137	2	0.103-0.170	0.104	3	0.101-0.106	0.218	9	0.077-0.460
30-45 min off Fire	L.R.	0.120	1	0.120	0.080	2	0.064-0.096			
1 h After Fire Off	K.	0.074	2	0.068-0.080	0.067	2	0.054-0.080	0.145	8	0.056-0.250
Total Samples			36			34			58	

Note. K: kitchen; L.R.: living rooms.

Method of Analysis

CO₂ concentrations were determined by portable infrared analyzer GXH-3010C. In the tightly closed kitchens the operator analyzed CO₂ concentrations (%) during the combustion of NG, CG and LPG and at different periods of time after turning off the stoves, and living rooms were used as controls. The CO₂ standard in public places is 0.1% or 0.15%.

Organic Compounds in Combustion Products of NG, CG and LPG

Findings in the studies are illustrated in Tables 5-8.

TABLE 5

Contents of Organic Compounds on Particles as Combustion Products of NG, CG and LPG

Types of Gas	NG	LPG	CG
Contents of Organic Compounds (%)	0.21	0.34	0.88

TABLE 6
Organic Compounds as Combustion Products of NG Identified by GC/MS

No.	Molecular Formula	Molecular Weight	Chemical Names
1	C ₁₃ H ₁₈	184	3,3-dimethyl Hendecane
2	C ₁₃ H ₂₈	184	2,6,8-trimethyl Decane
3	C ₁₇ H ₃₆	240	Heptadecane
4	C ₁₉ H ₄₀	268	2,4,10,14-tetramethyl Pentadecane
5	C ₁₈ H ₃₈	254	Octadecane
6	C ₂₁ H ₄₄	296	2,6,10,14-tetramethyl Heptadecane
7	C ₁₉ H ₄₀	268	Nonadecane
8	C ₁₉ H ₃₂	260	(1-methyl dodecyl)-benzene

TABLE 7
Organic Compounds as Combustion Products of LPG Identified by GC/MS

No.	Molecular Formula	Molecular Weight	Chemical Names
1	C ₁₃ H ₂₈	184	3,7,7-trimethyl Decane
2	C ₁₃ H ₂₈	184	2,6,8-trimethyl Decane
3	C ₁₇ H ₃₆	240	Heptadecane
4	C ₁₉ H ₄₀	268	2,4,10,14-tetramethyl Pentadecane
5	C ₁₄ H ₁₀	178	Phenanthrene
6	C ₁₈ H ₃₈	254	Octadecane
7	C ₂₁ H ₄₄	296	2,6,10,14-tetramethyl Heptadecane
8	C ₁₉ H ₄₀	268	Nonadecane
9	C ₁₉ H ₃₂	260	(1-methyl dodecyl)-benzene
10	C ₂₀ H ₄₂	282	Larane
11	C ₁₆ H ₁₀	202	Pyrene
12	C ₂₁ H ₄₄	296	Heneicosane
13	C ₂₂ H ₄₆	310	Docosane
14	C ₂₃ H ₄₈	324	Tricosane
15	C ₂₄ H ₅₀	338	Tetracosane
16	C ₂₅ H ₅₂	352	Pentacosane
17	C ₂₆ H ₅₄	336	Hexacosane
18	C ₂₇ H ₅₆	380	Heptacosane
19	C ₂₈ H ₅₈	394	Octacosane
20	C ₂₉ H ₆₀	403	Nonacosane

TABLE 8
Organic Compounds as Combustion Products of CG Identified by GS/MS

No.	Molecular Formula	Molecular Weight	Chemical Names
1	C ₁₃ H ₂₈	184	2,6,8-trimethyl Decane
2	C ₁₂ H ₈	152	Acenaphthene
3	C ₁₃ H ₂₈	184	3,3-dimethyl Undecane
4	C ₁₆ H ₃₄	226	5-propyl Tridecane

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No.	Molecular Formula	Molecular Weight	Chemical Names
5	C ₁₇ H ₃₆	240	Heptadecane
6	C ₁₄ H ₁₀	178	Phenanthrene
7	C ₁₈ H ₃₈	254	Octadecane
8	C ₁₉ H ₄₀	268	Nonadecane
9	C ₁₉ H ₃₂	260	(1-methyl dodecyl)-benzene
10	C ₂₀ H ₄₂	282	Eicosane
11	C ₁₆ H ₁₀	202	Pyrene
12	C ₂₁ H ₄₄	296	Heneicosane
13	C ₂₂ H ₄₆	310	Docosane
14	C ₂₃ H ₄₈	324	Tricosane
15	C ₂₄ H ₅₀	338	Tetracosane
16	C ₂₅ H ₅₆	352	Pentacosane
17	C ₂₆ H ₅₄	336	Hexacosane
18	C ₂₇ H ₅₆	380	Heptacosane
19	C ₂₈ H ₅₈	394	Octacosane
20	C ₂₉ H ₅₀	408	Nonacosane
21	C ₁₆ H ₁₀	202	Fluoranthene
22	C ₁₈ H ₃₅ ON	281	9-octadecene Amide
23	C ₃₀ H ₆₂	422	triacontane

Radon Concentrations in Natural Gas Fields in Beijing

Radon concentrations were determined in natural gas fields in Beijing, and the findings are illustrated in Tables 9-10.

TABLE 9

Daily Changes of Rn Concentrations in Natural Gas in Beijing (Bq/m³)^a

Times of Sampling	Mar. 4	Mar. 5	Mar. 7	Mar. 10	Mar. 11	Mar. 17	Mar. 18	Mar. 24	Mar. 25	Average values	SD
Morning		104	82	79	122	67	112	101	68	92	21
Noon	126		100	88	101	118	107	114	93	106	13
Evening	82		71	116	95	117	112	118	120	104	19
Daily Averages	104		84	94	106	100	111	111	94	101	
SD	22		15	20	14	29	3	9	26	9	

Note. ^a:1992

TABLE 10

Monthly Changes of Rn Concentrations in Natural Gas in Beijing (Bq/m³)^a

Months of Sampling	Mar.	Apr.	May	July	Aug.	Sept.	Oct.	Nov.	Dec.	Jan. ^a	Feb. ^a	Yearly Averages
Monthly Averages	101	157	120	103	137	106	128	92	121	97	70	112
SD	8	1	2	74	4	13	7	13	23	18	12	24

Note. ^a:1993

As shown in Tables 9-10, Rn concentrations in the noon and evening, i.e. the peak time of daily gas use, were basically similar, being 106 and 104 Bq/m³ respectively, while the average Rn concentrations in the morning were 92 Bq/m³, lower than that in the noon and in

the evening. This might be explained by the fact that after staying in the NG pipe over night Rn went partially decayed in the morning. In the measurement for 9 successive days in March, the daily Rn concentrations in NG ranged from 84 to 111 Bq/m³, or lower than relative changes of RN concentrations within one day. The Rn concentrations ranged from 70 to 157 Bq/m³ within a year, with an average value of 112 Bq/m³ and no evident seasonal change was observed.

The average Rn concentrations in the Renjiu gas field and the final users determined from March 1992 to February 1993 was 112 Bq/m³, which was higher than the concentration of 73 Bq/m³ in the gas fields of north China as reported in 1986, but was lower than that in the coal gas used in Beijing and higher than that in the locally used liquefied petroleum gas^[4]. Nevertheless, this figure proved much lower than mathematic averages of Rn concentrations in 15 gas fields in Sichuan Province^[5].

Concentrations of 1-hydroxy Pyrene in Urine of the Study Subjects

As shown in Table 11, 1- hydroxy poyrene concentrations in subjects' urine were positively correlated to B(a)P concentrations in the air of kitchens. The highest B(a)P concentrations were found either in subjects' urine or in the air exposed to LPG, the second high concentrations were found in those exposed to CG and the lowest in those exposed to NG.

TABLE 11

Concentrations of 1- hydroxy Pyrene in Subjects' Urine and in the Indoor Air

Types of Gas	B(a)P in Indoor Air (ng/m ³) $\bar{x} \pm s$	Hydroxy Pyrene in Urine ($\mu\text{mol/mol}$ creatine) $\bar{x} \pm s$	Number of Samples
LPG	3.39 \pm 0.93	0.277 \pm 0.237	4
CG	2.02 \pm 0.43	0.247 \pm 0.157	4
NG	1.28 \pm 0.52	0.147 \pm 0.173	5

DISCUSSION

Comparison of Combustion Effects of NG, CG and LPG

Among the three gases, NG showed the best combustion effects, which was followed in order by LPG and CG. This is closely associated with their respective chemical composition (89% CH₄, for instance, in NG, 50% C₃H₈ and 50% C₄H₁₀ in LPG and 27% CH₄ in CG). The components of the combustion products were directly related to composition of the gases themselves, as well as to conditions of combustion. The organic matters contained on the particles as the combustion products of the 3 gases all included three normal alkanes, three branched-chain alkanes and one alkyl substitute of benzene, suggesting that despite some differences in their original composition the products of combustion in them are partially identical.

Comparison of Organic Components of NG, CG and LPG

Both LPG and CG had rather complicated organic components, and the particles as their combustion products all contained polycyclic aromatic hydrocarbons in addition to several

normal alkanes. The CG particles differed from the LPG ones by containing one more normal alkanes, two additional polycyclic aromatic hydrocarbons (namely, acenaphthene and fluoranthene) and also octadecene amide.

Particles produced in the combustion process of the three gases all contained B(a)P as a result of indoor air monitoring (Table 2). However, B(a)P was absent in their combustion products according to the findings of GS/MS, and pyrene was not detected in NG combustion products by GS/MS (Tables 6, 7 and 8). For B(a)P, HPLC with a sensitivity of 10^{-12} g was applied while for pyrene GS/MS with a sensitivity of 10^{-9} g was used. It is important to note that though containing lower hydrocarbons as its chief components, NG was still able to polymerize into polycyclic aromatic hydrocarbons under a special combustion process.

Estimation of Internal Radiation Doses of Rn in Beijing Inhabitants

Both Rn and its daughter elements of short shelf life will produce internal radiation doses once they are inhaled. According to the model of dose estimation described in the ICKP report (No.30), the aerodynamic diameter equals to $0.2 \mu\text{m}$, suppose Rn's daughter elements of short shelf life have f_p equivalent to 0.025. Assuming that F is exaggerated as 1, or the equilibrium ratio between Rn and its daughter elements equals to 1, and that the inhabitants stay in indoor conditions all a year round, then the equivalent effective doses of Rn and its daughter elements for the inhabitants in that year will be 0.035 and $1.84 \mu\text{Sv}\cdot\text{a}^{-1}$, respectively, and $1.88 \mu\text{Sv}\cdot\text{a}^{-1}$ in total. These doses only represent 1.8‰ of the yearly equivalent effective doses to which Beijing inhabitants are exposed in indoor and outdoor conditions, or 0.8‰ of the yearly equivalent effective doses ($2.43 \mu\text{Sv}\cdot\text{a}^{-1}$) to which they are exposed from all natural radiation sources in the city.

CONCLUSIONS

In general, the indoor air pollution caused by NG, CG and LPG will be more severe in winter than in summer and more evident in kitchens than in other rooms of the house. The concentrations of traditional pollutants such as particles, NO_2 , CO, SO_2 are all within normal range, but they are lower in NG than in CG and LPG.

Combustion products of all the three gases contained lower organic components, no more than 1%, and of them NG was at the lowest level.

The NG from the Renqiu Oil Field which is currently used in Beijing has low Rn and its daughter elements, and their yearly effective radiation dose to which Beijing inhabitants are exposed is less than 1‰; nevertheless, in case the NG from Shaanxi Oil Field is used in the future, further monitoring of Rn will be necessary.

To reflect biological effect of B(a)P, the concentration of 1-hydroxy pyrene was detected in urine samples of the subjects and the findings indicate that it is a fairly sensitive indicator.

Considering the difficulty in sampling combustion products of the gases and a large quantity of samples required to test bioactivity of these products, micro Ames test was not established in the present study, but according to the findings in organic matters and available reports in the literature, it could be estimated that combustion products of NG will not display high bioactivity, or their bioactivity will be lower than that of CG and LPG.

Sample size in the present study was relatively small due to physical, technical and manpower limitations and further study will be needed to explore the health impact of natural gas, an ever widely used domestic energy source in China.

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