Adjustment of Urinary Mercury in Health Risk Assessment of Mercury

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The determination of adjustment method of urinary mercury in spot urine is one of the important issues in assessing the health risks of mercury workers. But there have been debates about whether creatinine or other forms of correction for urinary concentration are better in reducing the variation of urinary mercury. We evaluated four adjustment methods - specific gravity, creatinine, log creatinine and excretion rate - by correlation between values adjusted by the four methods and individual exposure levels which were the geometric mean of daily air mercury level for 2 or 5 days, and mercury concentrations in 24 hour urine were also investigated to compare the results of spot urine. The correlation between values of spot urine and mercury exposure level was over 0.8 in all adjustment methods for workers who worked over 1 year. All four adjustment methods for urinary mercury were found to be similar in assessing the exposure, log creatinine and excretion rate method however were not practical to use due to lack of reference values, and variable standard values of specific gravity. And the creatinine adjusted values were more sensitive in low mercury exposure level. We therefore recommend the creatinine adjustment method for adjustment of urinary mercury.

Key Words : *Adjustment, Mercury, Exposure, Urine*

INTRODUCTION

Biological monitoring of exposure assesses health risk through evaluation of internal dose. It has several advantages in assessing exposure such as taking into consideration of the exposure from all sources, and all the physicochemical and biological variables(Bernard and Lauwerys, 1987). It also provides an assessment of

control technology procedures or equipment and the effectiveness of various personal protective devices (Crable and Kneip, 1988).

Urine is one of the recommended specimens for biological monitoring because of the non-invasive technique for obtaining samples. Monitoring urinary mercury is useful for assessing the risk of adverse effects and the need for preventive measures (Barregard, 1993). Urinary mercury is a valuable indicator of average long-term exposure and reflects intergrated exposure over the preceding weeks or months in workers(Barregard, 1993 ; Mason and Calder, 1994). Urine levels of mercury may be an index of renal concentration of mercury (Cherian et al., 1978).

Since urine samples obviously vary in concentration, it is usually best to normalize or compensate for dilution

by applying correction factors(Hill et al., 1988). Methods of adjusting for varying hydration have included corrections based on urinary specific gravity, timed or daily urinary excretion, and simultaneous measurement of creatinine concentration. Among them, creatinine adjustment method is recommended for most biological exposure indices, but it also has its own problems, both logistic and physiologic. A new method for assessing urinary toxic substance concentration was proposed (Greenberg and Levine, 1989). Creatinine excretion is subject to wide fluctuations due to specific internal and external factors, and the use of creatinine to correct chemical concentrations in urine would not necessarily improve the correlation to exposure dose for all chemicals(Boeniger et al., 1993).

There also have been debates about whether creatinine or other forms of correction for urinary concentration are better in reducing intra individual variation of urinary Hg and thus making a single spot measurement more closely reflect true Hg excretion(Wallis and Barber, 1982). The day-to-day variation in urinary mercury is relavitely high even when 24-hour samples are used under stable exposure conditions(Barregard, 1993). Mason and Calder(1994) reported that correction for creatinine and, perhaps slightly less satisfactory, correction for SG reduce the uncertainty of a spot urinary Hg concentraion in reflecting accurately the true Hg excretion in an individual subject. Wallis and Barber(1982) reported that correcting the concentration for specific gravity reduced the variance in daily spot urine samples, but, in a follow-up publication, Barber and Wallis(1986) reported that the correction for specific gravity and osmolality turned out to be almost identical, and that correction for creatinine was more effective than the other corrections by a small but statistically significant amount.

The study about the adjustment of mercury concentration in spot urine has been focused on the variability of urinary concentration(Wallis and Barber, 1982; Mason and Calder, 1994) but not for assessing correlation between air mercury exposure and urinary mercury. Because air mercury level also varies each day, mean air mercury for several days would be a better exposure index than daily air mercury concentration. There were few data to assess air mercury level over one day.

In this study, we concentrated mainly on which correction factors would be best for adjustment method of urinary mercury by assessment of the association of urinary mercury with exposure level which was the averaged time-weighted air mercury concentration for several days.

MATERIALS AND METHODS

1. Subjects and field research

The study subjects were 36 mercury workers in two factories. They were requested to participate in this study on a voluntary basis. Among them, 24 workers had worked for less than 1 year, and 12 workers had more than 1 year(Table 1). The air mercury of workers was sampled by the NIOSH method 6009(by personal air sampler using hopcalite during 8 hours) for 5 days (from Monday to Friday) for workers of company A and 2 days(Monday and Tuesday) in company B(Table 2). They worked in the manufacture of fluorescent lamps, and were exposed to metallic mercury vapor. The working conditions were worse and the level of air mercury was higher in company A than in company B with lack of space, older machines, poor cleaning, and numerous inexperienced workers.

All spot urines of workers were collected in polyethylene tubes for 5 days in case of A company and 2 days in company B. While at work, workers were requested to visit a research team in the company for their urine to be collected in the polyetylene tubes when they felt a desire to urinate, and also were requested to collect their urine in a box for urine collection after work and to bring back the box when they came to their factory next morining. Our study team and we followed this field research from May 23 to May 31 in 1994.

2. Analysis in Laboratory

The analysis of mercury in hopcalite was based on NIOSH method 6009. The hopcalites were dissolved by concentrated HNO_3 and HCl. The mercury was reduced by 10% $SnCl_2$ in cold vapor generator(IL-551 England) and was measured by atomic absorption spectrophotometer at 253.7 nm of radiation.

Table 1. General characteristics of subjects

Sex and age group			Work duration (yr)		Total
			<1	>=1	
Male	Age group(yr)	<20	5		5
		20–29	16	5	21
		30–39		5	5
		40–49	1		1
Female	Age group(yr)	20–29	2	2	4
Total			24	12	36

Table 2. Air mercury and 24 hour urine mercury concentration in company A and B

Day of measurement	Air mercury(Unit : mg/m³) geometric mean±S.D(Range)		Mercury in 24 hour urine(Unit : μg/l) mean±S.D(Range)	
	A company	B company	A company	B company
Monday(94.5.23 for A ; 94.5.30 for B)	0.094±0.067 (0.019-0.248)	0.030±0.022 (0.009-0.078)	206.5±178.9 (12.16-725.3)	42.03±27.14 (10.33-123.9)
Tuesday(94.5.24 for A ; 94.5.31 for B)	0.059±0.058 (0.017-0.246)	0.017±0.012 (0.005-0.036)	276.5±220.6 (11.63-754.4)	44.19±31.03 (5.11-129.5)
Wednesday(94.5.25)	0.045±0.027 (0.009-0.117)	–	176.6±187.4 (26.69-766.9)	–
Thursday(94.5.26)	0.039±0.022 (0.016-0.088)	–	213.5±181.4 (27.13-676.1)	–
Friday(94.5.27)	0.096±0.267 (0.010-1.058)	–	271.3±215.2 (46.53-750.9)	–
Geometric Mean or Mean	0.068±0.109 (0.013-0.512)	0.021±0.014 (0.007-0.051)	212.6±173.9 (21.85-650.6)	34.72±22.06 (11.8-80.0)

Urine samples were transported in icebox to our institute at each survey day and were stored in referegerator under -20℃. The analysis of the mercury in urine was carried out by the method of Japan Labor Science Institute(1979). We modified the pretreatment method by using microwave digestion.

Urinary specific gravity was measured by means of a refractometer(ATAGO, Japan), and creatinine was measured by the Jaffe method(Whelton et al., 1994).

3. Analysis of Data

The urinary excretion of mercury would be stable after the exposure to mercury over 1 year(WHO, 1976), the subjects of this study were divided into 2 groups, one of which had worked for over 1 year, and the other had worked less than 1 year. The adjustments of spot urine were made by creatinine, log creatinine, speicific gravity and timed excretion rate.

Because the air concentrations of mercury were variable each day(Table 2), we used the geometric mean of daily air mercury for survey days as the exposure level(Table 4). We were not able to use the data of urinary mercury for 3rd, 4th, 5th day in company A, as the cooperation from workers was lessened from 3rd day of survey such as they forgot the visit to survey team when they felt desire of urination.

RESULTS

The correlation coefficients of air mercury and urinary mercury for each day were variable(Table 3). The correlation·coefficient between air mercury and 24 hour urinary mercruy for workers exposed to mercury over 1 year was 0.66 and 0.69, and for workers with less than

1 year of work period was 0.55 and 0.79. In the case of spot urine, correlation coefficients were more variable in both working periods. But the correlation coefficients between mercury in 24 hour urine and in spot urine were relatively high and stable.

We defined that mercury exposure level to be the geometric mean of air mercury level for survey days. When mercury exposure level was used as the exposure status instead of air mercury in each day, the correlation coefficients changed. The correlation between mercury exposure level and mercury in 24 hour urine of workers who had over 1 year of working period were from 0.91 to 0.94 which were almost the same as in several adjustment methods(Table 4). The correlation coefficients between mercury exposure level and urinary mercury of workers who had working period less than 1 year were lower and more variable than those of workers who had a working period of over 1 year.

The correlation coefficients between mercury exposure level and mercury in spot urine were found to be lower than those in 24 hour urinary mercury, but there also was no great difference according to adjustment methods in either working period levels(Table 5). The correlation coefficients of workers who worked over 1 year were higher and more stable than those of workers who worked less than 1 year.

One of the most important reasons for measuring urinary mercury is to assess the health risk of workers by assessment of exposure. We wanted to compare two exposure indices - urinary mercury and air mercury. Ministry of Labor in Korea recommeded that level of air mercury should be below 0.05mg/m³(Minstry of Labor, 1992a), and no adverse effect level of mercury was reported 50μg/l(Rempel et al., 1990). Therefore we

Table 3. Correlation coefficients between air mercury and urinary mercury in each day

Survey 1st day

Exposure duration	Mercury concentration	in Air	in 24HU[a]	in S-U[b]			
				HgSG[c]	HgCR[d]	HgLCR[e]	HgRA[f]
≥ 1yr	in Air	–	0.73 (N=10 p=0.017)	0.33 (N=66 p=0.0064)	0.50 (N=66 p=0.0001)	0.33 (N=66 p=0.0064)	0.52 (N=63 p=0.0001)
	in 24 HU		– (N=10 p=0.017)	0.97 (N=65 p=0.0001)	0.97 (N=65 p=0.0001)	0.94 (N=65 p=0.0001)	0.95 (N=65 p=0.0001)
< 1yr.	in Air	–	0.53 (N=10 p=0.113)	0.40 (N=69 p=0.0008)	0.42 (N=69 p=0.0003)	0.34 (N=69 p=0.004)	0.44 (N=60 p=0.0004)
	in 24 HU		– (N=10 p=0.017)	0.85 (N=55 p=0.0001)	0.83 (N=55 p=0.0001)	0.83 (N=55 p=0.0001)	0.67 (N=55 p=0.0001)

Survey 2nd day

≥ 1yr	in Air	–	0.92 (N=10 p=0.0002)	0.89 (N=52 p=0.0001)	0.88 (N=52 p=0.0001)	0.87 (N=52 p=0.0001)	0.89 (N=49 p=0.0001)
	in 24 HU		–	0.91 (N=49 p=0.0001)	0.95 (N=49 p=0.0001)	0.91 (N=49 p=0.0001)	0.98 (N=49 p=0.0001)
< 1yr.	in Air	–	0.059 (N=9 p=0.88)	0.015 (N=56 p=0.91)	0.054 (N=55 p=0.70)	0.01 (N=55 p=0.94)	0.12 (N=46 p=0.42)
	in 24 HU		–	0.95 (N=50 p=0.0001)	0.94 (N=50 p=0.0001)	0.94 (N=50 p=0.0001)	0.83 (N=49 p=0.0001)

a : Mercury concentration in 24 hour urine
b : Spot urine
c : Mercury concentration adjusted by specific gravity of urine, HgSG = Hg x [(1.020-1.000) / SG-1.000],
    ※Hg = uncorrected mercury concentration
d : Mercury concentration adjusted by creatinine of urine, HgCR = Hg / creatinine concentration
e : Mercury concentration adjusted by log creatinine of urine, HgLCR = Hg / log creatinine concentration
f : Mercury excretion rate, HgRA = (Hg x urine volume) / time

Table 4. Correlation coefficients between mercury exposure levels[a] and urinary mercury in 24 hour urine

Survey day	Work duration(yr)	HgSG	HgCR	HgLCR	HgRA	HgTo[b]
1st day	≥1 (N=11)	0.92 (p=0.0001)	0.92 (p=0.0001)	0.94 (p=0.0001)	0.93 (p=0.0001)	0.93 (p=0.0001)
	<1 (N=15)	0.48 (p=0.07)	0.37 (p=0.18)	0.46 (p=0.08)	0.40 (p=0.14)	0.40 (p=0.14)
2nd day	≥1 (N=10)	0.94 (p=0.0001)	0.93 (p=0.0001)	0.94 (p=0.0001)	0.91 (p=0.0003)	0.91 (p=0.0003)
	<1 (N=13)	0.86 (p=0.0001)	0.80 (p=0.0009)	0.84 (p=0.0003)	0.86 (p=0.0002)	0.86 (p=0.0002)
3rd day	≥1	–	–	–	–	–
	<1 (N=6)	0.98 (p=0.0008)	0.98 (p=0.0007)	0.97 (p=0.0011)	0.97 (p=0.0012)	0.97 (p=0.0012)

a : Geometric mean of mercury concentration in air for survey days
b : Total mercury content in 24 hour urine

Table 5. Correlation coefficients between mercury exposure levels and urinary mercury in spot urine

Survey day	Work duration(yr)	HgSG	HgCR	HgLCR	HgRA
1st day	≥1 (N=69)	0.82 (p=0.0001)	0.84 (p=0.0001)	0.80 (p=0.0001)	0.83 (p=0.0001)
	<1 (N=85)	0.31 (p=0.0043)	0.28 (p=0.0089)	0.25 (p=0.0224)	0.24 (p=0.0359)
2nd day	≥1 (N=52)	0.83 (p=0.0001)	0.85 (p=0.0001)	0.82 (p=0.0001)	0.87 (p=0.0001)
	<1 (N=72)	0.60 (p=0.0001)	0.62 (p=0.0001)	0.57 (p=0.0001)	0.66 (p=0.0001)
3rd day	≥1	−	−	−	−
	<1 (N=72)	0.60 (p=0.0001)	0.62 (p=0.0001)	0.57 (p=0.0001)	0.66 (p=0.0001)

Table 6. Mercury concentration in 24 hour urine according to level of mercury exposure level

Work duration	Mercury exposure level(mg/m^3)	Hg-urine(μg/l)			
		50>	50-100	100<	Total
≥ 1yr.	≥ 0.05	0(0.0%)	0	1(100.0%)	1
	< 0.05	7(63.6%)	3(27.3%)	1(9.1%)	11
< 1yr.	≥ 0.05	3(42.8%)	0(0.0%)	4(57.1%)	7
	< 0.05	8(47.1%)	4(23.5%)	5(29.4%)	17

Table 7. Mercury concentration in 24 hour urine adjusted by creatinine according to mercury exposure level

Work duration	Mercury exposure level(mg/m^3)	Hg-urine(μg/l)			
		50>	50-100	100<	Total
≥ 1yr.	≥ 0.05	0(0.0%)	0(0.0%)	1(100.0%)	1
	< 0.05	10(90.9%)	0(0.0%)	1(9.1%)	11
< 1yr.	≥ 0.05	3(42.8%)	0(0.0%)	4(57.1%)	7
	< 0.05	9(52.9%)	4(23.5%)	4(23.5%)	17

Table 8. Mercury concentration adjusted by urinary specific gravity in spot urine according to mercury exposure level

Work duration	Mercury exposure level(mg/m^3)	Hg-urine(μg/l)			
		50>	50-100	100<	Total
≥ 1yr.	≥ 0.05	7(70.0%)	0	3(30.0%)	10
	< 0.05	84(76.4%)	18(16.4%)	8(7.3%)	110
< 1yr.	≥ 0.05	46(65.7%)	4(5.7%)	20(28.6%)	70
	< 0.05	129(75.9%)	20(11.8%)	21(12.4%)	170

divided air mercury level at the point of 0.05 as high and low exposure, and the concentrations of urinary mercury at the points of 50 and 100 as high, moderate and low levels to assess urinary mercury level by air exposure of mercury.

In the study of 24 hour urine of workers who worked over 1 year, 36.4% of subjects showed above 50μg/l among workers who had mercury exposure level less than 0.05mg/m^3(Table 6). When the urinary mercury concentrations were adjusted by urinary creatinine, the proportion decreased to 9.1% from 36.4%(Table 7).

In the study of 24 hour urine of workers who worked less than 1 year, 52.9% of subjects showed above 50μg/l among workers who had mercury exposure level less than 0.05mg/m^3(Table 6). When the urinary mercury concentrations were adjusted by urinary creatinine, the proportion decreased to 47.0% from 52.9%(Table 7).

In the case of spot urine adjusted by specific gravity among workers who worked over 1 year, 23.7% of samples showed above 50μg/l of urinary mercury when air mercury level was below 0.05mg/m^3(Table 8). When urinary mercury concentrations were adjusted by creatinine, the proportion decreased to 11% from 23.7% (Table 9). With workers who worked less than 1 year, 24.2% of subjects showed above 50μg/l of urinary mercury when air mercury level was below 0.05mg/m^3(Table

Table 9. Mercury concentration adjusted by urinary creatinine in spot urine according to mercury exposure level

Work duration	Mercury exposure level (mg/m³)	Hg-urine (μg/l)			
		50>	50-100	100<	Total
≥ 1yr.	≥ 0.05	7(70.0%)	0	3(30.0%)	10
	< 0.05	98(89.1%)	6(5.5%)	6(5.5%)	110
< 1yr.	≥ 0.05	49(70.0%)	2(2.9%)	19(27.1%)	70
	< 0.05	133(78.2%)	22(12.9%)	15(8.8%)	170

8). When urinary mercury concentration were adjusted by creatinine, the proportion decreased to 21.7% from 24.2%(Table 9).

DISCUSSION

The relationship between urine and air levels of mercury has been variable in several studies such as 5:4 (Roles et al., 1987), 3:1(WHO, 1976), 1:1(Bell et al., 1973). The correlation coefficients between air and urine levels of mercury were 0.81(Roles et al., 1987), 0.47 (Lauwerys and Buchet, 1973), 0.30(Park et al., 1989).

When daily air mercury concentration was used as the air level of mercury, the correlations between urine and air levels of mercury were variable both in 24 hour urine and spot urine(Table 3). The correlation coefficients of air mercury with mercury in spot urine showed big difference between the results in survey 1st and 2nd day. However, the correlation coefficients in workers with over 1 year of working period were relatively high and stable when geometric mean of daily air merury level were used as the air levels of mercury(Table 4,5). Both variation of air mercury and urinary mercury resulted in great variation of the correlation of air mercury with mercury in spot urine in table 3. Such results showed that when air mercury of factory was not stable, the correlation between air mercury and mercury in urine was also variable.

No definite or specific methods are recommended for the adjustment of values in urine sample in the Methods book by Ministry of Labor in Korea(1989, 1992b). There is an urgent need to establish the adjustment method for biological values of urine in Korea. It is not possible to establish one adjustment method for all substances.

The renal excretory mechanisms of substances were classified into four major categories - glomerular filtration, glomerular filtration and net tubular secretion, glomerular filtration and net tubular reabsorption, and no glomerular filtration(suspected tubular secretion). In the case of adjustment for urinary lead and amino-

levulinic acid which are glomerular filtration and net tubular reabsorption type, log creatinine adjustment method showed the best results among adjustment methods(Hudak and Kiss, 1991; Yu et al., 1994).

Mercury is not filtrated through glomerulus, so it may be secreted through tubules. And there was no great difference in the correlation coefficients between spot urine and mercury exposure level according to adjustment methods which were above 0.8 for workers who worked over 1 year period(Table 5). Substances of tubular secretion type may be less affected by adjustment mehods.

But specific gravity method presents a big problem when the values adjusted by it should be changed according to researcher's choice of standard values. Standard specific gravity which various authors have taken to be in the range of 1.018 to 1.024(Barber and Wallis, 1986). Smith et al.(1970) and Hill et al.(1988) used standard specific gravity as 1.024, Araki(1973) as 1.020, Mason and Calder(1994) as 1.016. There is no international or Korean standard for standard specific gravity. Correction to 1.023 gives values 33% higher than correction to 1.018(Aitio, 1988).

Adjustment by log creatinine or excretion rate is under study and could not be used as an exposure index of mercury due to lack of reference values. Among the four adjustment methods the creatinine correction method presents fewer problems and was more favorable than others in the assessment of health risk.

Only 30% of workers exposed to above 0.05mg/m³ of mercury exposure level showed over 50μg/g creatinine in both working periods(Table 9). There was no difference of proportion of urinary mercury level which is above 50μg/l or 50μg/g creatinine between the values of spot urine adjusted by creatinine and specific gravity when air mercury level was high. But it decreased that the propotion of above 50 of urinary mercury in high air mercury exposure when urinary mercury was adjusted by creatinine.

ACGIH recommended TLV-TWA of air mercury as

0.025mg/m^3 and BEI of urinary mercury as 35μg/g creatinine(ACGIH, 1995). We analysed the data in the same way with ACGIH recommeded values. The results were similar to above data(The data are not presented here).

The above results showed that despite the relative high correlation between mercury concentration in spot urine and mercury exposure level, the creatinine adjusted method may be a better estimator in assessing health risk of workers than the other methods. Log creatinine and excretion rate method were not practical to use due to lack of reference values, and standard values of specific gravity methods were variable. And Barber and Wallis(1986) and WHO(1991) recommended the creatinine adjustment method for urinary mercury, ACGIH(1995) used creatinine adjustment method for Biological Exposure Indices. So we recommend the creatinine adjustment method for correction of urinary mercury in spot urine.

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