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Health Effects of MTBE Among New Jersey Garage Workers

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Methyl tertiary butyl ether (MTBE) is an oxygenated additive used in the 1993 wintertime oxyfuel program to reduce tailpipe carbon monoxide emissions. Because of complaints of acute health symptoms, particularly in the state of Alaska, this program was terminated prematurely in that state. We designed a cross-sectional cohort study of self-reported symptoms of garage workers in the state of New Jersey exposed to high and low MTBE concentration environments. Two hundred and thirty-seven participants were divided into 2 groups: 115 workers in northern New Jersey sampled during the wintertime oxyfuel program, and 122 workers in southern New Jersey 10 wk after the phase-out date for the program in that area. The outcome measures included a list of symptoms, of which some were felt to be attributable to MTBE exposure. Participants were asked to indicate the frequency of those symptoms they had experienced over the last 30 days. In addition, workers were given identical preshift and postshift questionnaires and asked to rank any discomfort they were experiencing at that time from a list of symptoms. In comparing all garage workers in northern New Jersey (high exposure) to those in southern New Jersey (low exposure), no differences were found in the reporting of symptom frequency over the last 30 days. In the pre-/postshift questionnaire analysis, both groups felt significantly worse by the end of the work day, but there was no difference between the groups across the work shift. In comparing fuelers who pump gasoline more than 5 h/day, again no differences were found in the reporting of symptom frequency over the last 30 days. In the pre-/postshift analysis, however, the northern group reported a significantly increased number of symptoms, but again no difference was found between the groups across the workshift. Because the fuelers in the north were on average 15 yr older than those in the south, this same analysis was repeated with fuelers in the north and age-, sex-, and education-matched controls in the south. In this analysis, no group differences in symptom reporting were seen. In summary, no untoward health effects clearly attributable to MTBE exposure could be demonstrated in this cohort of healthy garage workers.

Methyl tertiary butyl ether (MTBE) is an oxygenated additive to gasoline. Since 1975, high-octane unleaded gasolines have contained levels of MTBE that vary from 3 to 15% by volume (Cline et al., 1991). In addition to its use to improve octane performance, highly branched oxygenated hydrocarbons such as MTBE have been added to gasoline to lower tailpipe carbon monoxide emissions in order for communities to come into compliance with the standards set forth in the Clean Air Act (Japar et al., 1991). This act requires areas that do not meet federal carbon monoxide standards to use oxygenated fuels during the winter months, when, because of incomplete

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combustion, the carbon dioxide levels are the highest. At these times, the gasoline must contain at least 2.7% oxygen by weight or 15% MTBE by volume (Anderson, 1993). The health effects of environmental exposure to MTBE, however, remain unknown. MTBE is currently approved by the Food and Drug Administration under investigational status for use in intraductal injection to dissolve cholesterol gallstones (Thistle et al., 1989). Investigations in humans and in animals for this usage of MTBE has led to reports of a number of symptoms including nausea, vomiting, anorexia, hypotension, headaches, palpitations, angina, pain, and local tissue inflammation (Tritapepe et al., 1989; Adam et al., 1990; Holl et al., 1991; Murray et al., 1988; DiPadova et al 1986; Neoptolemos et al., 1990).

During the winter of 1992–1993 (November 15, 1992 to April 30, 1993), MTBE was added to gasoline stores to up to 15% by volume. From gasoline, MTBE is volatilized into the air. In the state of Alaska, health concerns and complaints by residents forced the end of the use of MTBE in fuels in December 1992, in advance of the March 31 termination date (*Anchorage Daily News*, February 10, 1993). The primary aim of this study was to test the hypothesis that the prevalence of acute health effects due to exposure to MTBE is higher among those people who report having the greatest occupational exposure to MTBE.

METHODS

A cross-sectional study of symptoms and exposure was done among New Jersey garage workers. The state of New Jersey maintains a series of garages for maintenance and fueling of the state transportation fleet. All garages that employed more than five people and did at least half of their fueling as gasoline (not just diesel) were sampled. The entire state of New Jersey participated in the oxygenated fuel program during the winter of 1992. However, the part of New Jersey considered to be in the Philadelphia airshed area (southern New Jersey) was only required to have oxygenates in the fuel from November 15, 1992 to February 28, 1993, while that part of New Jersey in the New York airshed area (northern New Jersey) was required to participate in the oxygenated fuel program from November 15, 1992 to April 30,1993. All data were collected from northern New Jersey during April 1993. Because stores of gasoline may get used up over varying periods of time, in order to ensure the least amount of misclassification of exposure, data were not actually collected in southern New Jersey (our low-exposure population) until May 1993.

Garages were visited after employees had been at work for approximately 1 h. Employees were asked to fill out several questionnaires, including one that inquired about symptoms and their severity over the last 30 days. (Symptoms thought to be attributable to MTBE exposures were compiled from the reported acute side effects after its use in gallstone dissolution, animal toxicology studies, and reports from the studies in Fairbanks and Anchorage.) The questions about MTBE symptoms (headache, nausea, daytime sleepiness while driving and at other times, cough, lightheadedness, and eye irritation) were encouched in a number of other questions about non-MTBE symptoms (diarrhea, fever, sweats or chills, and muscle aches). These non-MTBE questions provided a basis for determining if one group had a higher incidence of acute viral illness, which might make the findings difficult to interpret. In addition, employees were asked to fill out a questionnaire inquiring about preshift symptoms (although this was usually done after the employee had been at work for about 1 h). These symptoms included headache, nose and mouth irritation, dizziness, nausea, sleepiness, cough, and disorientation, thought to be attributable to MTBE exposure, and fever, diarrhea, sweats or chills, and muscle aches, considered to be non-MTBE symptoms. The employees were given an identical questionnaire and asked to fill this out postshift and return it in the self-addressed stamped envelope provided. The participants were also asked to fill out some questions regarding their likelihood for sensitivity to chemicals in general, which had been used in other studies by one of us. The participants were also asked to fill out the Wahler Physical Symptoms Inventory (Wahler, 1983) and the Amplification Scale (Barsky et al., 1990). The instrument by Wahler is a standardized inventory that assesses overall health and physical symptom reporting over a year's period of time. The Amplification Scale assesses the tendency to attend to or amplify bodily sensations. Since some symptoms on the Wahler overlapped with acute MTBE symptoms (i.e., headaches, dizzy spells, nausea), these symptoms were excluded from analyses involving both questionnaires. Finally, the participants were asked to rate the smell of the gasoline in their garage on that day on a scale from 1 to 5 with 5 being the most objectionable.

Active air sampling was done for 1 h at 1 garage in northern New Jersey and in the garages in southern New Jersey. In addition, passive samplers were given to 20 subjects. They were asked to wear one sampler on each of three subsequent work days and then return the samplers in the prestamped envelope provided. A fourth trap was included to serve as a field blank. The traps consisted of 0.2 g carboxen 569, which retains MTBE, in a 1/4-in ID stainless steel tube purchased from Perkin Elmer for use with a PE automated thermal desorption unit (ATD400). A diffusive cap design for the trap was deployed to maintain a constant flow rate. Solid endcaps were placed on the traps during transport and storage. The traps were initially cleaned by heating them for 12 h at 250°C with a constant flow of purified nitrogen passing through them.

After return to the laboratory, each trap was analyzed by thermal desorption coupled to gas chromatography/mass spectroscopy (GC/MS). Identification was based both on retention time and mass spectrum. The amount of MTBE measured in the samples and blank adsorbent traps were characterized into one of five groups: <5 ng, not detected; 5–15 ng, low; 15–25 ng, medium; 25–100 ng, high; and >100 ng, very high. Each participant was asked to wear the trap all day on the outside of clothing and to take it off only at bedtime. The time that each trap was exposed was recorded. However, the exact volume of air sample was not known, since the diffusion coefficient of MTBE for these traps is not known. An estimate of the volume collected was made based on diffusion coefficients of other ethers. The diffusion coefficients for diethyl ether, isopropyl ether, and *n*-butyl ether are 0.092, 0.068, and 0.054 cm^2/s, respectively. Since MTBE has a similar molecular weight and structure to isopropyl ether and since diffusion coefficients are known to be a function of these two properties, a value of 0.068 cm^2/s was used as the best estimate. To calculate the sampling rate of the passive sampler the diffusion rate is combined with the physical parameter of the sampler and used in Fick's law: $U_m = (60\ D_1 A)/Z$ where U_m is the uptake rate in milliliters per minute, D_1 is the diffusion coefficient through air of the vapor under study in square centimeters per second, A is the cross-sectional area of the sampling tube in square centimeters, and Z is the pathlength of the air gap in centimeters. For the traps used, $A = 0.2\ cm^2$ and $Z = 1.5$ cm. Using these values, the expected uptake rate of MTBE is 0.074 ml/min. For an 8-h sample the concentration is calculated as follows: Concentration (mg/m^3) = [amount on the trap (ng) × (10^{-6} mg/ng) × 8 h] / [0.0044 L/h × (10^{-3} L/m^3)]. The exposure categories would correspond to these 8-h time-weighted averages: <1 mg/m^3, not detected; 1–3 mg/m^3, low; 3–6 mg/m^3, medium; 6–22 mg/m^3, high; and >22 mg/m^3, very high.

All statistical analyses were performed in SAS/PC from the SAS Institute Inc., Cary, N.C. All independent variables were tested individually using the chi-square statistic. In addition, summary scores were created for each individual over the entire range of MTBE and non-MTBE symptoms. These were scored for the 30-day questionnaire as 0 = never had the symptom, 1 = had the symptom less than once per week, 2 = had the symptom 2–3 times per week, and 3 = had the symptom more than 3 times per week. The maximum summary scores for the 30-day questionnaire were 21 for the MTBE symptoms and 12 for the non-MTBE symptoms. For the pre- and postshift questionnaires, a summary score was obtained by scoring 0 = does not have the symptom, 1 = notices the symptom a little bit, 2 = has moderate discomfort from the symptom, 3 = has quite a bit of discomfort from the symptom, and 4 = has extreme discomfort from the symptom. The maximum summary scores for the pre- and postshift questionnaire were 28 for MTBE symptoms and 16 for non-MTBE symptoms. The differences between group summary scores on the 30-day questionnaire were evaluated using Student's *t*-test. The differences between summary scores on the pre- and postshift questionnaires were analyzed using repeated-measures analysis of variance across the shift (time effect), between the northern and southern groups (group effect), and between groups across the workshift (time × group interaction).

All parts of this study were approved by the Investigational Review Board of the Robert Wood Johnson Medical School prior to its inception.

RESULTS

There were 115 garage workers who participated in the northern part of New Jersey, which represented a 79% participation rate, and 122 workers in southern New Jersey at an 81% participation rate. Reasons given for not participating were primarily those of not wanting to take the time to fill out the questionnaires and also lack of interest. Eighty percent of participants in both the north and the south remembered to fill out and send back their postshift questionnaires. Based on the assumption that the background rate for headache in the unexposed is 10%, this study had a 92% power to detect a 2.5 increase in this symptom with a one-sided alpha error of 5%. Headache was chosen for a single-variable power study since it was the most prevalent reported symptom in the Alaska studies. In Fairbanks, 9/101 university students reported headache but 5/16 taxi drivers reported this symptom (Middaugh, 1992).

Overall, the workers in the north and workers in the south were similar with the exception that workers in the north had slightly more years of education than those in the south (Table 1). There was no difference in symptom reporting between the two groups for non-MTBE symptoms, making it unlikely that any acute illness prevalent in one or several garages might account for differences in reporting. There was slightly more nausea reported among southern garage workers, but this did not reach statistical significance in the chi-square stratified over all categories (p = .10) but did in an ever versus never analysis (p = .03). The only other difference between the two groups was in reporting daytime sleepiness at times other than when driving; the rate of reporting both of these symptoms was higher among southern workers than in those in the north (Table 2). Analysis of pre- and postshift questionnaires revealed that all workers felt significantly worse at the end of the work day regarding those symptoms thought to be attributable to MTBE exposure (p = .004), but there was no difference between the groups (p = .72) and no difference in reporting rates between groups across the work shift (p = .44) (Table 3).

Garage workers constitute a heterogeneous group of fuelers, mechanics,

TABLE 1. Demographic Characteristics of the Overall Study Population

Characteristic	North (high exposure)	South (low exposure)	p
n	115	122	
Age (mean, yr)	41.6	40.4	0.36
Male gender (%)	98.8	95.9	0.76
Years of education (mean)	12.6	11.9	0.002
Race (% white)	81.1	90.4	0.11
Wahler Inventory (mean)	37.4	41.9	0.29
Amplification Scale (mean)	2.16	2.19	0.73
Sensitivity to chemicals (%)	0.12	0.14	0.81
Response to gas smell (mean)	3.9	4.0	0.20

TABLE 2. Symptoms Over the Last 30 Days Among the Study Population

Symptoms	North	South	*p*
MTBE symptoms (% ever)			
Headache	36%	39%	0.82
Nausea	27%	35%	0.03
Cough	12%	17%	0.23
Lightheadedness	10%	13%	0.41
Sleepiness driving	20%	24%	0.37
Daytime sleepiness	13%	24%	0.001
Eye irritation	22%	27%	0.39
Summary score (mean)	4.3	5.1	0.09
non-MTBE symptoms (% ever)			
Diarrhea	14%	20%	0.08
Fever	4%	4%	0.90
Chills	9%	8%	0.56
Muscle aches	34%	35%	0.91
Summary score (mean)	1.8	1.9	0.60

mechanic helpers, and others. In some garages certain employees were dedicated fuelers, while at other locations this job was rotated among many mechanic helpers. Because fuelers may have a much higher exposure than some other garage workers, separate analyses were done for those individuals who indicated that they pumped gasoline more than 5 h/day. There were 13 employees in the north and 15 in the south who indicated that they spent on average more than 5 h/day pumping gasoline. These two groups differed significantly in age (Table 4). Thirty-day symptom reporting for both MTBE and non-MTBE symptoms and the associated summary scores were

TABLE 3. Pre-/Postshift Symptom Reporting of the Study Population

Report	North	South	*p*
MTBE symptoms			
Pre-shift summary score	1.73	1.74	
Post-shift summary score	2.05	2.30	
Time effect			0.004
Group effect			0.72
Time × group effect			0.44
non-MTBE symptoms			
Pre-shift summary score	1.01	0.98	
Post-shift summary score	0.92	0.89	
Time effect			0.29
Group effect			0.88
Time × group effect			0.99

TABLE 4. Demographic Characteristics of Workers Who Pump Gas >5 Hours Per Day

Characteristic	North (high exposure)	South (low exposure)	p
n	13	15	
Age (mean, yr)	51.5	35.5	0.001
Male gender (%)	92.3	93.3	0.92
Years of education	12.1	11.6	0.50
Race (% white)	72.7	80.0	0.91
Wahler Inventory (mean)	42.5	23.9	0.22
Amplification Scale (mean)	2.01	1.82	0.48
Sensitivity to chemicals (%)	0.08	0.07	0.90
Response to gas smell (mean)	4.5	3.9	0.17

not significantly different. Analysis of the pre- and postshift questionnaires did show a higher rate of symptom reporting in the north than in the south that reached statistical significance for MTBE symptoms (p = .03) and nearly so for non-MTBE symptoms (p = .07), but there was no difference in reporting by time (at the end of the work day) and no difference between the groups across the work shift (Table 5). Since these two groups varied by an average of 15 yr in age and had large differences in their Wahler Inventories for overall symptom reporting, age- (±2 yr), gender-, and education-matched southern controls were found for 11 of the 13 northern fuelers. These southern controls were chosen among those who wore the 3-day passive samplers and had readings of "nondetectable" or "low" levels of MTBE exposure or from those southern garages where the active air sampling was nondetectable (Table 6). When northern fuelers were compared with age-, sex-, and education-matched controls, the differences in pre- and postshift questionnaire analyses disappeared (Table 7).

TABLE 5. Pre-/Postshift Symptom Reporting Workers Who Pump Gas >5 Hours Per Day

Report	North	South	p
MTBE symptoms			
Preshift summary score	2.44	0.69	
Postshift summary score	3.22	1.00	
Time effect			0.12
Group effect			0.03
Time × Group effect			0.49
non-MTBE symptoms			
Preshift summary score	1.33	1.22	
Postshift summary score	0.23	0.54	
Time effect			0.68
Group effect			0.07
Time × Group effect			0.38

TABLE 6. Demographic Characteristics of Northern Workers Who Pump Gas >5 Hours Per Day and Matched Southern Controls

Characteristic	North (high exposure)	South (low exposure)	*p*
n	11	11	
Age (mean, yr)	50.5	49.1	0.66
Male gender (%)	100	100	
Years of education	12.1	12.3	0.67
Race (% white)	78	73	0.43
Wahler Inventory (mean)	46.4	44.3	0.93
Amplification Scale (mean)	2.02	2.33	0.40
Sensitivity to chemicals (%)	9.1	18.2	
Response to gas smell (mean)	4.36	4.00	0.43

Active air sampling done for 1 h in the single garage for which this test was available in the north yielded high MTBE levels. Active air sampling at garages in the south varied from low to nondetectable levels. Passive samplers were given to 20 subjects, 14 in northern New Jersey and 6 in southern New Jersey. One of the set of 3 samples in the north had high levels in the blank sample, so only 13 individuals had valid samples at 9 separate sites. All nine sites in the north had at least one individual whose samplers were categorized as high. For seven of the sites it was categorized as high or very high on all of the days samplers were collected. There was one individual in the north whose samplers were in the medium range and one whose samplers were low or medium who was employed as a supply parts supervisor. Two individuals in the north had samples on 1 of 3 days with nondetectable levels, but those individuals had high levels on other days. Of the six individuals who participated in the south, two sets were returned

TABLE 7. Pre-/Postshift Symptom Reporting of Northern Workers Who Pump Gas >5 Hours per Day and Matched Southern Controls

Report	North	South	*p*
MTBE symptoms			
Preshift summary score	2.62	2.45	
Postshift summary score	3.37	2.00	
Time effect			0.81
Group effect			0.66
Time × Group effect			0.33
non-MTBE symptoms			
Preshift summary score	1.25	1.64	
Postshift summary score	1.12	0.91	
Time effect			0.28
Group effect			0.94
Time × Group effect			0.44

with contaminated blanks. The four sets of samplers were given to individuals at four different locations. Two of these locations had high levels on two of the days the samples were collected. One of the locations had low concentrations on 1 day and nondetectable levels on the other 2 days. The final location had nondetectable levels on all 3 days.

CONCLUSIONS

This study was conducted to contrast symptom reporting among two groups of garage workers: one group actively exposed to MTBE, and another group several weeks after MTBE was scheduled to be phased out of the wintertime oxyfuel program. No increased rates of symptom reporting were found among northern garage workers who worked in the area that was still required to participate in the oxyfuel program. Although both groups reported feeling worse by the end of the work day, there were no differences between the groups across the work shift. Because workers required to refuel vehicles with gasoline may be at risk for the highest exposures, this group was looked at separately. Fuelers in the north reported more symptoms as a group than those in the south, but this difference disappeared when they were compared to age-, sex-, and education-matched controls, and no difference was present between the groups across the work shift.

The findings in this study are in contrast to those reported elsewhere. In Fairbanks, AK, studies were done on taxi drivers, health care workers, and university students, and in Anchorage, AK, on taxi drivers and health care workers utilizing the amount of time these groups averaged in their cars to stratify exposure. These studies report the number of persons meeting a case definition of oxyfuel-related illness as higher among taxi drivers than among the comparison groups (Middaugh, 1992, 1993).

Symptoms thought to be due to MTBE (headache, mucous membrane irritation, nausea, cough, lightheadedness, and drowsiness) are not uniquely attributable to this exposure. Gasoline by itself, which was present in all of the garages, may account for any of these complaints. Likewise, not all of the garages were of standard design, and some variation in ventilation between the workplaces is bound to occur. Workers by and large are a healthier group than the population as a whole. In addition, garage workers may be a self-selected group who are better able to tolerate exposures to these types of fumes. This study was done at the end of the oxyfuel season across the phase-out period. It may be that some degree of discomfort is noticed when an exposure is added (at the beginning of the season), but after workers have developed some tolerance to exposure to the chemical, no lessening in symptoms is appreciated with its withdrawal. Finally, some degree of misclassification of exposure is present in this study. Overall, we believe that at the times of sampling, those workers in the north were more exposed than those in the south. Nevertheless, despite the precaution of waiting 10 wk after the phase-out date of oxyfuels in southern New Jersey, our measurements clearly indicate that some workers still received a significant exposure, most likely due to changing fuel filters and pumps in vehicles that still contained the wintertime oxyfuel. In addition, the southern group, even outside of the oxyfuel season, continued to have some environmental exposure to MTBE due to its presence in premium-grade high-octane fuels.

Disregarding any problems with exposure misclassification, we found overall symptom reporting to be quite low. On the postshift questionnaire we found scores ranging from 1.0 to 3.22 among the various groups studied on our symptom-severity score index out of a maximum of 28. This does not exclude the possibility that certain subgroups of the population may exhibit an increased sensitivity to MTBE and that future studies should look at these groups separately.

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