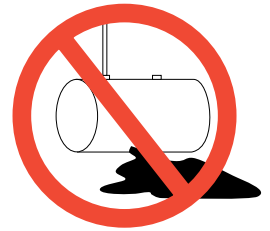
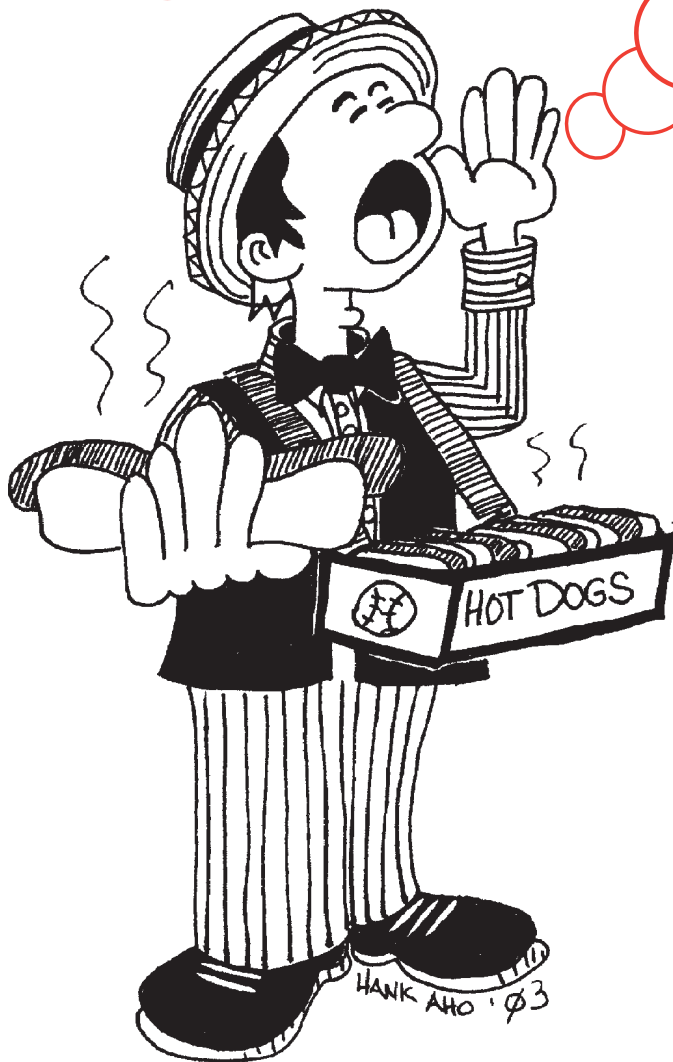


L.U.S.T.LINE

A Report On Federal & State Programs To Control Leaking Underground Storage Tanks



A **Hot Dog** by Any Other Name Could Be Your Drinking Water



by Patricia Ellis

"Hot dogs, getcher hot dogs!"

The cry of the hot dog vendor at the ballpark. The steaming hot frank with your choice of mustard, ketchup, relish...the captivating aroma and the even more satisfying taste! But let's not stop to think about what's actually in a hot dog. Sure, some of us take comfort in consuming only hot dogs that are "all beef" or "chicken" or Kosher. But what's really in a hot dog? Do we really want to know? And just what has the composition of hot dogs got to do with an article that is ostensibly about leaking underground storage tanks, anyway? Well, it has to do with this propensity to not want to know about those ingredients...even the ingredients in our own drinking water.

■ *continued on page 2*

Inside

- 7 NEIWPCC Survey on Oxygenates at LUST Sites: Part 1
- 10 An Overview of Treatment Technologies for MTBE
- 14 Part 2: Surface Flux-Chamber Method
- 19 Taking On Today's Challenges
- 20 DE's Systemwide Approach to Preventing Releases
- 25 Baffled by a Leak? Check the Inventory Records
- 29 Thoughts on the Tortoise and the Hare Revisited
- 30 ME's Dispenser and Submersible-Pump Sump Study
- 33 PEI's 2003 Edition of RP200
- 34 CA Water District Sues Oil Companies
- 34 CA Updates Guidance on ELD and SIR
- 36 EPA HQ Update

■ Hot Dogs from page 1

Is Ignorance Bliss?

The drinking water supply systems in the United States are unquestionably the best in the world. Most people can simply turn on the faucet and draw a glass of fresh, clear water that they can put unflinchingly to their lips and drink. Yet, a growing segment of the population uses a filter of some sort, and increasing numbers of people buy bottled water. In fact, in the last 40 years, it is estimated that the U.S. drinking water industry has lost nearly 60 percent of its customers to competitors (currently unregulated) who are "bottled water and point-of-use/point-of-entry providers." (Means et al., 2002) Why?

The reasons are many—taste, odor, color, fad/style, fear (justified or imagined). For those of us on public water supplies, our water suppliers provide us with annual Consumer Confidence Reports (CCRs) that show us that our water has been tested for

a variety of contaminants and is safe to drink. But most water suppliers analyze for a couple dozen contaminants at most. The CCRs tell us whether or not these contaminants were detected and at what concentrations.

When these contaminants are detected, even when their concentration may from time to time exhibit a spike above a regulatory threshold, this water is still distributed to us. Generally an accounting gimmick, such as 30-day average concentration, is employed so that it can be claimed that although detected above the limit, the concentration did not exceed "permissible" levels and the water is safe to drink.

For example, if the analytical report for a sample indicates that each of the BTEX compounds is present but at concentrations below their MCLs (5 ppb, 1,000 ppb, 700 ppb, and 10,000 ppb, respectively), is water with up to 11,705 ppb of BTEX really safe to drink? Do we want to drink it knowing that although the levels are reportedly safe, these contaminants are present at all? Do we want our children drinking it? And, health concerns aside, how does it taste? What about other contaminants that are not on this list of only a couple dozen? Are some of them present and, if so, what do we know about them?

Petroleum Cocktail Hour

Petroleum (and the various fuels distilled from petroleum) is composed of hundreds to thousands of individual organic compounds. (Although this article focuses on gasoline, much of the discussion is applicable to other fuels as well.) "Gasoline" is a complex blend of several hundred hydrocarbons (i.e., compounds that contain only hydrogen and carbon atoms) and other organic compounds that typically contain nitrogen, oxygen, or sulfur. The specific composition of any particular blend of gasoline is a function of the petroleum source, refining and blending processes, and additives (Kreamer and Stetzenbach, 1990). The composition also varies with geographic location and from season to season to maintain performance specifications and comply with regulatory requirements.

The primary groups of hydrocarbons in gasoline are the paraffins, olefins, naphthenes, and aromatics (Youngless et al., 1985). Table 1 lists some representative examples for each of the various classes of these organic compounds. Additive packages (which are generally proprietary) vary considerably and typically include compounds that function as antioxidants, antiicers, metal deactivators, detergents, and corrosion inhibitors, among others (Youngless et al., 1985). Some of these compounds are extremely large, complex molecules.

Some components of gasoline may also contain metal species. The most familiar of these, but not the only ones, are the organic lead compounds, which are no longer used in modern unleaded gasolines. In the past, especially with leaded fuels, a wide variety of dyes were incorporated into gasoline blends as well. Table 2 lists a few of the many gasoline additives.

In addition, a significant number of the compounds in gasoline are *unknown* (or unidentified), except for the number of carbon atoms they contain (Kreamer and Stetzenbach, 1990). What do we know about the toxicity of each of the compounds in gasoline? How do they behave in the environment? Which ones are in our drinking water and at what levels?

For an organic contaminant to show up in a water sample, it must be water soluble. It is well known that aromatic hydrocarbons (of which BTEX is probably the best recognized) are the most soluble constituents of gasoline. Table 3 lists 43 common gasoline constituents with solubility greater than 1 mg/L. Two of the nonaromatic compounds in this table have a higher solubility than ethylbenzene (the "E" in BTEX). This list isn't comprehensive, and there are undoubtedly other compounds with similar properties and, hence, significant water solubility.

While these constituents represent pure compound solubility, and individual solubilities from a mixture would be somewhat lower, the point is that there are lots of soluble constituents in gasoline that can appear in groundwater. If a sample is only analyzed for the aromatic fraction, how do we know that some of these other constituents are not also present?



LUSTLine

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Toxicity of Petroleum Constituents

It should come as no surprise to anyone that exposure (e.g., through inhalation, ingestion, or dermal contact) to any of the constituents of gasoline (or any other fuel) at any concentration should be avoided. Exposure to the vapors from most gasoline constituents can cause dizziness, drowsiness, unconsciousness, and other adverse effects on the central nervous system. Prolonged exposure to low concentrations, or brief exposure to higher concentrations, may damage internal organs, cause cancer or birth defects, or may even be fatal. Ingestion of the liquid phase of neat gasoline (and most, if not all, of its individual constituents) is acutely toxic.

So where do we find information on the toxicity of specific constituents? We would expect that one of the best sources is a material safety data sheet (MSDS), and there are many places to find them on the Internet. But they are readily available only for a small percentage of the constituents of gasoline, and, as they only pertain to exposure to a single compound, the effects of exposure to dilute aqueous mixtures are entirely unknown. (This issue is likely to be one of the important public health challenges of this century, and further discussion is way beyond the scope of this article.)

One of the current ways to deal with a large number of organic compounds is to distribute them into smaller groups, each of which has a designated "surrogate." This is the approach adopted by the Total Petroleum Hydrocarbon Criteria Working Group (TPHCWG). In this method, it is presumed that all members of the group have properties that are similar to the surrogate.

But the approach has several drawbacks. First, compound toxicity isn't necessarily the same for each of the group members, and often the toxic characteristics of a significant proportion of the group are unknown. Second, the presence of the surrogate in a sample may not necessarily mean that there are any other compounds in the sample; if they are, they are probably not at the same concentration. Third, the absence of the surrogate in a sample may not

necessarily mean that all of the other compounds in the class are also absent from the sample. Fourth, many states are statutorily authorized to regulate only those contaminants that appear on EPA's list (i.e., 40 CFR 302.4, discussed in the "Regulation..." section below).

Sadly, the focus on compound toxicity has been so narrowly concentrated on human carcinogenicity that adverse effects other than cancer are usually conveniently ignored. In almost any discussion of risk management, there is no consideration of the teratogenic (birth defect) or mutagenic (mutation) effects of these toxic compounds—not to mention taste or odor!

We have no idea what contaminants are really in the water we drink (or the hot dogs we eat). Simply because a contaminant isn't listed on an analytical report does not mean that the contaminant is not present in the sample. The truth is that we just don't know, but what we don't know can potentially hurt us.

The issue of exposure to multiple toxicants is likewise given short shrift—exposure to multiple toxic compounds is limited to presumed simple additive effects, if it's considered at all. Yet, it is well recognized that the toxicity of a chemical may be increased (or in some cases even decreased) by simultaneous or consecutive exposure to another chemical (Lu, 1991). There is no consideration of synergistic (multiplicative) effects, or whether mixtures may contain procarcinogens, cocarcinogens, or cancer promoters.

And then there's the issue of whether or not a specific compound is a human carcinogen or just an animal carcinogen. Too often an animal carcinogen is touted as being a human noncarcinogen simply because there isn't any confirmation that the compound causes cancer in humans. However, saying that a compound is a noncarcinogen, when the truth is that there isn't enough

information about it to determine whether or not it is a human carcinogen (although the compound is a known animal carcinogen), is being less than honest.

Admittedly, it is difficult (maybe even impossible) to demonstrate with 100 percent certainty that any chemical is a noncarcinogen. But for compounds that are known animal (especially mammalian) carcinogens, ordinary common sense would tell a reasonable person that these are substances with which unnecessary contact should be avoided, even at low concentrations and especially in mixtures that contain substances that may promote cancer.

In theory, a single molecule of a carcinogen can induce cancer. This means that there is no threshold dose and therefore no safe level of exposure to carcinogens. While not all cancer researchers hold this view, the opposing view (i.e., that threshold doses for carcinogens do exist) has yet to be demonstrated, even though large-scale experiments have been conducted for this purpose (Lu, 1991). Further complicating the issue is that unless a fatal quantity of pure product is ingested, most of the toxic effects are slow to develop (10 to 20 years or more in humans) and may be masked by other ailments as we age.

Regulation of Hazardous Substances

Underground storage tanks containing hazardous substances are regulated by the UST program under 40 CFR 280. Additional regulations regarding hazardous substances are found in 40 CFR 302.4 and 40 CFR 261.24. The first of these, CFR 302.4 (U.S. EPA, 2001a), is U.S. EPA's list of approximately 800 Hazardous Substances. Of these substances, only a handful are petroleum hydrocarbons found in fuels, and even fewer are fuel additives. (See Table 4.)

The second regulation, 40 CFR 261.24, is the Toxicity Characteristics (TC) rule for identifying RCRA hazardous wastes. The TC rule specifically exempts "petroleum contaminated" media and debris that fail the test for the toxicity characteristic of 40 CFR 261.24 (U.S. EPA, 2001b). Section 261.24(b) refers to 25 contami-

■ continued on page 4

■ Hot Dogs from page 3

nants (but actually lists 26) that are specifically exempt from consideration as “hazardous wastes,” provided they are subject to the corrective action regulations under 40 CFR 280 (the UST regulations). This list of 26 contaminants includes benzene and only two additional chemicals (cresol and pyridine) that *may* be present in gasoline or other petroleum fuels.

We all know that none of the components of gasoline (or other petroleum fuels) are healthy for us, so why is it that so few fuel constituents are officially designated as “toxic” or “hazardous”? Part of the answer is that there are simply too many potentially toxic substances to list; some are unidentified, and adequate toxicity testing hasn’t been conducted on others. Although not limited to organic compounds, the Chemical Abstract Service (CAS) assigns unique registration numbers (known as CAS or CASRN) to new chemicals at a rate of about 4,000 per day!!! (See <http://www.cas.org/EO/regsys.html>.)

Another part of the answer is that petroleum fuels as a whole are a critical part of the world economy. They’ve been used for close to 100 years, so we’re familiar with them, we need them, and we consider them to be relatively “safe.” Perhaps the primary reason why gasoline is considered “safe” is because UST regulations are relatively effective—at least to the extent that there aren’t daily media reports of explosions, fires, and underground rivers of gasoline flowing beneath our feet.

However, as we all know, releases from UST systems do happen, sometimes with immediate and catastrophic effect. Every day there are releases of gasoline (and other fuels) into the environment, and a significant amount of the released fuel eventually winds up in groundwater or surface water or both, some of which is used for drinking water. So how do we know what toxic compounds (if any) are actually in our drinking water?

Identification of Toxic Compounds

Let’s assume that we have a water sample that may or may not be conta-

minated with one or more of the hundreds of petroleum constituents in gasoline. What tests can we conduct to determine what contaminants are in the sample? Several analytical methods are potentially available to us to determine if any contaminants are present in the sample and at what concentrations. Though not the sole source for analytical methods, EPA’s compendium of analytical methods, SW-846, (U.S. EPA, 1997) offers us several choices of determinative analytical methods for organic compounds, including: Methods 8015, 8021, 8260, and 8270. Let’s look into each of these in ascending numerical order. (See Table 5.)

■ **Method 8015** (Nonhalogenated Organics Using GC/FID) explicitly lists 30 compounds, of which only four may be present in gasoline. Only one—methanol—is on the list of hazardous substances. This method may also be used for Gasoline Range Organics (GRO) and Diesel Range Organics (DRO), but other methods (which aren’t specified in the scope) may be more applicable. No additional guidance is provided regarding GRO or DRO.

■ **Method 8021** (Aromatic and Halogenated Volatiles by Gas Chromatography Using Photoionization and/or Electrolytic Conductivity Detectors) explicitly lists 57 compounds, of which 10 may be present in gasoline and are also on the list of hazardous substances.

■ **Method 8260** (Volatile Organic Compounds by Gas Chromatography/Mass Spectrometry) explicitly lists 107 compounds, of which about a dozen may be found in gasoline and are also on the list of hazardous substances.

■ **Method 8270** (Semivolatile Organic Compounds by Gas Chromatography/Mass Spectrometry) explicitly lists about 250 compounds, of which only a couple are likely to be found in gasoline (although many more could be present in diesel fuel and heavier fuel oils) and are on the list of hazardous substances.

U.S. EPA drinking water methods 502 and 524.2 contain a slightly different list of chemicals.

Of the more than 400 target compounds identified by the four 8000-series methods, approximately 5 percent may be present at any given petroleum release site. “Well and good,” you’re thinking, “but what’s the point of this?”

Absence of Proof Is Not Proof of Absence

Well, the first point of this is that we have no idea what contaminants are really in the water we drink (or the hot dogs we eat). Simply because a contaminant isn’t listed on an analytical report does not mean that the contaminant is not present in the sample. (Note that the converse is also true—that is, there is no proof that the contaminant is present.) The truth is that we just don’t know, but what we don’t know *can* potentially hurt us.

There are a lot of reasons why the presence of a contaminant in a sample might go unrecognized:

- There was no analysis for the contaminant.
- There was an analysis for the contaminant, but an inappropriate method was used.
- The analytical method was applied incorrectly.
- The detection limit is very high.
- Matrix interferences.

In each of these cases, a contaminant could be in a sample, but its presence (and concentration) is undetected (and undetermined). We have to do a better job than we currently do to both anticipate which potential contaminants may be present at a given site and analyze for all of them to determine whether they are in fact present or absent.

In addition to the desirability of knowing all chemicals present for the purpose of conducting a risk assessment, it is important to know all the contaminants present when developing a remediation plan.

In one of my recent projects, carbon filters used as point-of-entry treatment for domestic wells were breaking through in far shorter times than what was expected. After run-

ning Method 8260 plus requesting that all "tics" be identified by a library search, we identified a total of 45 additional chemicals, all potentially having a gasoline source, as being present in the water samples. These additional chemicals all contributed to the loading on the carbon filters and contributed to the early breakthrough. The library search gave estimated concentrations, but none of these compounds had been calibrated against a standard.

I might also have been happier if I hadn't added dissolved lead to the list of analytes because of earlier detections of EDC. Dissolved lead exceeded recommended levels in every sample (pre- and post-carbon filters), and in every well, even where no gasoline components were detected. Further analysis, this time for tetraethyl and tetramethyl lead, the organic lead that would come from leaded gasoline, was negative. Elevated lead levels appeared to be present throughout the aquifer, which would also have to be factored into a risk assessment. While the carbon filters were dealing with the gasoline contamination in the wells, albeit in an expensive manner, the filters had no effect on the dissolved lead.

Further, it isn't enough to have samples analyzed even for all potential contaminants if the samples aren't representative. Samples must be collected from locations where contaminants are most likely to be present, and they must be correctly handled during collection, transport, preparation, and analysis.

Fuel-Specific Analytical Methods

My second point is that the current analytical practices we rely on to determine whether gasoline compounds are present or absent in water (and soil) samples are incomplete and therefore inadequate. Standard operating procedures for Methods 8015, 8021, 8260, and 8270 require calibration for only a few of the many compounds that are present in gasoline, but many compounds are either not present or are unknown.

Target analyte lists must be refined so that they are more representative of the contaminants that are

Table 1 Representative Organic Compounds Found in "Gasoline"

<p>Straight Chain Alkanes propane n-hexane n-dodecane</p> <p>Branched Alkanes isobutane 2,2-dimethylbutane neopentane 3-ethylhexane</p> <p>Cycloalkanes cyclohexane n-propylcyclopentane ethylcyclohexane</p> <p>Straight Chain Alkenes cis-2-butene 1-pentene trans-2-heptane</p> <p>Branched Alkenes 2-methyl-1-butene 4,4-dimethyl-cis-2-pentene</p>	<p>Cycloalkenes cyclopentene 3-methylcyclopentene</p> <p>Alkyl Benzenes benzene toluene ethylbenzene o-xylene m-xylene p-xylene 1,2-dimethyl-3-ethylbenzene 1,2,3-trimethylbenzene 1,2,4,5-tetramethylbenzene n-propylbenzene</p> <p>Other Aromatics indan 1-methylindan phenol</p> <p>Polycyclic Aromatic Hydrocarbons (PAHs) naphthalene</p> <p><i>Source: Adapted from Cole (1994).</i></p>
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Table 2 Representative Organic Compounds Used as Additives in "Gasoline"

<p>Oxygenates methyl <i>tertiary</i>-butyl ether (MTBE) ethanol ethyl <i>tertiary</i>-butyl ether (ETBE) <i>tertiary</i>-butyl alcohol (TBA) <i>tertiary</i>-amyl ethyl ether (TAEE) diisopropyl ether (DIPE) <i>tertiary</i>-amyl methyl ether (TAME) <i>tertiary</i>-amyl alcohol (TAA) methanol</p> <p>Anti-knock compounds tetra-ethyl lead (TEL) tetra-methyl lead (TML) methylcyclopentadienyl manganese tricarbonyl (MMT)</p> <p>Anti-oxidant compounds hindered phenols phenylene diamines aminophenols</p> <p>Anti-icing compounds isopropyl alcohol amides/amines glycols organophosphate ammonium salts</p>	<p>Corrosion inhibitors carboxylic acids sulfonates amine/alkyl phosphates</p> <p>Metal deactivators disalicylidene amines phenolic amines thiourea</p> <p>Ignition controller additives tri-<i>o</i>-cresol phosphates</p> <p>Detergents aminohydroxyamide alkylphenols imidazolines</p> <p>Lead scavengers 1,2-dichloroethane (EDC) 1,2-dibromoethane (EDB)</p> <p>Dyes azobenzene-4-azo-2-naphthol benzene-azo-2-naphthol para-diethyl aminoazobenzene 1,4-diisopropylaminoanthraquinone</p> <p><i>Source: Adapted from Cummings (1977) and Irwin, et. al. (1997).</i></p>
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likely to be encountered at fuel-release sites. For example, nearly 90 percent of the analytes listed for Method 8021 are halogenated compounds that would *not* be present at fuel-release sites—why should a sample be analyzed for them and *not* for some of the few hundred other

contaminants that may actually be present? If we're going to pay for an analysis for, say, 100 compounds, wouldn't it be more cost-effective if those 100 could be reasonably anticipated to be in the sample?

■ *continued on page 6*

■ Hot Dogs from page 5

And, in order to credibly evaluate the actual risk posed by contaminants in our water, we absolutely *must* know which contaminants are in the water. In a recent series of articles by Uhler and others (2002, 2003), similar suggestions were made. They suggest a suite of 109 target analytes for the analysis of automotive gasoline using a Modified 8260 method. The list contains the PIANO compounds (Paraffins, Isoparaffins, Aromatics, Naphthenes, and Olefins), useful for recognizing peculiarities that might be inherited from refinery processes (including

various major and minor iso-alkanes), and gasoline additives, including the oxygenate additives (alcohols and ethers), lead scavengers (EDC and EDB), and methylcyclopentadienyl manganese tricarbonyl (MMT). Some of this list of compounds can be useful in fingerprinting gasoline for environmental forensic investigations, as well as a basis for conducting a risk assessment.

Just how credible, how “scientifically defensible” is a risk assessment based on omission, neglect, or wishful thinking? To only evaluate the risk posed by some, but not all, contaminants present at a site is like crossing a busy highway but only looking

in one direction as you make the attempt. Sure, you may not get hit by a car coming from the direction in which you’re looking, but one from the blind side is likely to spoil your day.

Appropriate analytical method(s) already exist in today’s marketplace. All that is lacking are appropriate calibration standards and standard operating procedures that have been optimized for analysis of these target analytes. Once it becomes routine to use these standards, risk assessments could be conducted for the contaminants to which receptors are actually exposed, rather than presumed surrogates. This will go a long way toward bolstering the credibility of risk assessment and restoring confidence in the safety of our drinking water.

Take Me Out to the Ballgame

Alternatively, we could opt to accept the status quo...we can slump down in our bleachers, hot dog in one hand, and glass of water (OK, beer) in the other, and blissfully pass away the time. ■

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Table 3
Common Gasoline Constituents
Ranked by Solubility (mg/L)

Benzene	1,780
Toluene	515
o-Xylene	220
cis-2-Pentene	203
Cyclopentane	156
Ethylbenzene	152
1-Pentene	148
3-Methyl-1-butene	130
Indan	100
1-Methyl-4-ethylbenzene	95
1,2,3-Trimethylbenzene	77
1-Methyl-2-ethylbenzene	75
Propane	62
1,2,4-Trimethylbenzene	57
Cyclohexane	55
n-Propylbenzene	52
Isopropylbenzene	50
1,3,5-Trimethylbenzene	50
Isobutane	48.9
Methylcyclopentane	42
Pentane	38.5
Naphthalene	31
1-Methyl-naphthalene	28
2-Methyl-naphthalene	25
2,2-Dimethylbutane	18.4
sec-Butylbenzene	17
Methylcyclohexane	14
Isopentane	13.8
2-Methylpentane	13.8
n-Butylbenzene	13.8
3-Methylpentane	12.8
Isobutylbenzene	10.1
Hexane	9.5
2,3-Dimethylpentane	5.25
1,2,4,5-Tetramethylbenzene	3.48
3-Methylhexane	3.3
n-Heptane	2.93
2-Methylhexane	2.54
2,2,4-Trimethylpentane	2.44
2,3,4-Trimethylpentane	2
1-Nonene	1.12

Source: Adapted from Gustafson et al. (1997).

Table 4
Hazardous Substances Listed in 40 CFR
302.4 That May be Present in “Gasoline”

SUBSTANCE	CASRN
1,2-dibromoethane	106934
1,2-dichloroethane	107062
1,3-pentadiene	504609
benzene	71432
cresols	1319773
<i>ortho</i> -cresol	95487
<i>meta</i> -cresol	108394
<i>para</i> -cresol	106445
cyclohexane	110827
ethylbenzene	100414
methanol	67561
naphthalene	91203
phenol	108952
toluene	108883
xylenes	1330207
<i>ortho</i> -xylene	95476
<i>meta</i> -xylene	108383
<i>para</i> -xylene	106423

Source: Adapted from Gustafson et al. (1997).

Table 5 Compounds Present in “Gasoline” That Appear on Target Analyte
Lists for Methods in SW-846

COMPOUND	8015*	8021	8260	8270
diethyl ether	x		x	
ethanol	x		x	
methanol	x		x	
pyridine	x		x	
benzene		x	x	
ethylbenzene		x	x	
naphthalene		x	x	x
toluene		x	x	
xylenes		x		
<i>o</i> -xylene		x	x	
<i>m</i> -xylene		x	x	
<i>p</i> -xylene		x	x	
1,2-dibromoethane		x	x	
1,2-dichloroethane		x	x	
tertiary-butyl alcohol			x	
phenol				x

*Method 8015 is also indicated to be applicable for GRO and DRO.

■ Hot Dogs from page 6

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EPA HQ UPDATE

Analytical Methods Fact Sheet Available

Analytical methods for petroleum hydrocarbons are well established; however, methods that were developed for analysis of petroleum hydrocarbons in water samples may or may not be appropriate for fuel oxygenates. A fact sheet, titled *Analytical Methodologies for Fuel Oxygenates* (EPA 510-F-03-001), outlines the potential problems of analytical methods for common fuel oxygenates and ways to address these problems. It has been distributed to states and regions and is available on the OUST Web site at www.epa.gov/oust/mtbe/omethods.pdf.


For more information, contact Hal White at (703) 603-7177.

Senate Passes UST Legislation

On Thursday, May 1, the U.S. Senate passed the Underground Storage Tank Compliance Act (S. 195) by unanimous vote. The legislation provides additional flexibility and authorization of appropriations for preventing and cleaning up releases from USTs. It also includes a dedicated authorization of appropriation for the cleanup of MTBE, mandatory inspection frequencies, additional enforcement tools, and operator training guidelines. Although the House has been working on its version of UST legislation, companion legislation has not been introduced. EPA OUST has been providing technical comments on the proposed legislation.

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