# The Ten Pollutant Study in Jacksonville, Florida

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## ABSTRACT

In 1997 Jacksonville's Environmental Quality Division began ambient air monitoring for air toxics. In addition to air toxics monitoring Jacksonville began calculating hazardous air pollutant (HAP) emission inventories for point, area and mobile sources. Air monitoring data indicated ten HAPs were continually present at the majority of the air toxics monitoring sites and have consistently high average annual concentrations in comparison with other monitored pollutants. These HAPs were chosen for further study. HAP inventory data from the year 2000 was compiled based on Source Classification Code to determine which source categories were emitting these HAPs. Results indicate the majority of emissions appear to be from mobile sources and the least emissions from point sources. On-road mobile sources are a significant contributor to at least five HAPs found from air monitoring. Non-road mobile sources are also a significant contributor. At least seven of the HAPs are emitted from point and area source surface coating operations. Degreasing and consumer/commercial solvent use are significant contributors for at least six of the monitored HAPs. Other significant contributors include dry cleaning and gasoline use. This study has aided Jacksonville in determining which types of facilities are contributing to ambient levels of particular HAPs. This data can be used to develop local regulatory efforts to minimize emissions and for risk analysis modeling. Future HAP emission inventories and air toxics monitoring will aid in establishing trends over time, which can be used to determine the success of regulatory efforts.

## INTRODUCTION

In the late 1980's the air section of the City of Jacksonville's Environmental Quality Division (EQD) began developing an air toxics program. The program consisted mostly of tracking Toxic Release Inventory (TRI) data and conducting risk assessments, which included comparing air pollution dispersion modeling results with health benchmarks[1]. In 1997 Jacksonville began ambient air monitoring for air toxics using EPA Method TO-15 for measurement of thirty-eight volatile organic compounds[2]. Twenty-six of these compounds are identified on EPA's list of 188 Hazardous Air Pollutants (HAPs) in Title III of the 1990 Clean Air Act Amendments. Ambient monitoring was conducted utilizing a mobile monitoring laboratory. Two stationary air toxics monitoring sites were added to the air toxics monitoring network in 1999 and three additional stationary sites were added in 2002. After several years of monitoring, it became apparent that fifteen of the monitored pollutants were continually present in the ambient air. These pollutants also have consistently high average annual concentrations in comparison with the other monitored pollutants. Ten of these fifteen pollutants are HAPs and were chosen for further study. These ten HAPs are: toluene, benzene, xylene, methyl chloride (chloromethane), ethyl benzene, styrene, carbon tetrachloride, methylene chloride (dichloromethane), methyl chloroform1(1,1,1-trichloroethane) and tetrachloroethylene (perchloroethylene).

In 1998, Jacksonville conducted its first emission inventory for HAPs, using 1997 data. Prior to 1998, a criteria pollutant emission inventory was conducted every three years as well as an ozone inventory, a requirement of the State Implementation Plan (SIP). For the HAP inventory, point, area and mobile source emissions were calculated for the 188 pollutants on EPA's HAP list. A second HAP inventory was conducted using 2000 data. These inventories were developed for several uses, one of which was to determine the origin of the ten HAPs being found in the ambient air from air monitoring. This paper discusses the sources of emissions for the ten HAPs as identified in the 2000 HAP inventory.

## METHODOLOGY

The City of Jacksonville's mobile monitoring laboratory began collecting ambient air samples at various locations around Jacksonville in July of 1997. These samples are collected in 6-liter stainless steel Summa canisters and are analyzed in accordance with EPA Method TO-15. The mobile laboratory is equipped with an Agilent Technologies 5973N Gas Chromatograph/Mass Spectrometer, Entech Instruments Cryogenic Concentrator and computer. Samples enter the system through a stainless steel probe line and are collected in stainless steel canisters for a four-hour period prior to analysis. In addition to measuring pollutant concentrations, the system monitors meteorological conditions including wind speed, wind direction and temperature. The mobile laboratory monitors the air at approximately five different locations per year and remains at each location for approximately ten weeks. Monitoring locations were chosen to characterize pollutant concentrations over as large an area as possible while also maintaining security for the equipment, because the mobile laboratory is left unattended over night. Also sites were selected which meet the criteria for distance from obstacles and unrestricted airflow as required by 40 CFR Part 58 for criteria pollutant monitoring. In 1997 through 2000, an onboard generator supplied electricity for the mobile lab. Due to continual malfunctions and down time with the generator, permanent power supplies were recently constructed at five locations. The mobile lab now operates within these locations. In 1997 & 1998 the laboratory was located in mostly suburban neighborhoods to establish background data or determine concentrations which citizens may be exposed to in and around their homes. In 1999 locations were chosen closer to industry in an effort to monitor pollutants, which may be emitted by industrial facilities. Beginning in 2000, the mobile lab operated mostly in the vicinity of industry or within high motor vehicle traffic areas.

The stationary air toxics monitoring site network also includes five monitoring sites. These sites are separate from the sites used for mobile laboratory monitoring. Two stationary air toxics monitoring sites were established in January of 1999 and three additional sites were added in October of 2002. All five monitoring sites were co-located with criteria pollutant monitoring sites in an effort to reduce the expense of purchasing addition shelters. Sites were chosen to provide an adequate representation of the entire county. Several of the sites are downwind or in the vicinity of industrial facilities or high motor vehicle traffic areas. One twenty-four hour canister sample is collected one day per week at each site and is analyzed using EPA Method TO-15 at the EQD laboratory. Each week the day of sample collection is rotated to the next day so that samples are collected on a variety of days.

The air toxics monitoring data is evaluated at the end of each calendar year and various statistics are compiled from the data. For each pollutant, annual average concentrations are calculated along with maximum and minimum concentrations. Another of the parameters determined is the percentage of samples containing a particular pollutant as shown in Table 1.

**Table 1. Percentages of air samples found to contain particular pollutants 1997 – 2000.**

| Pollutant | 1997 | 1998 | 1999 | 2000 |
|---|---|---|---|---|
| Dichlorodifluoromethane-F12 | 99 | 84 | 96 | 85 |
| Chloromethane | 91 | 1 | 61 | 85 |
| Trichloromonofluoromethane-F11 | 88 | 87 | 94 | 85 |
| Benzene | 96 | 97 | 96 | 83 |
| Toluene | 99 | 98 | 97 | 83 |
| Carbon Tetrachloride | 1 | 1 | 78 | 74 |
| Styrene | 81 | 93 | 58 | 67 |
| 1,2,4-Trimethylbenzene | 75 | 46 | 72 | 66 |
| Xylene | 65 | 74 | 96 | 63 |
| Ethyl benzene | 31 | 47 | 80 | 56 |
| 1,1,1-Trichloroethane | 41 | 19 | 40 | 54 |
| 1,3,5-Trimethylbenzene | 8 | 31 | 33 | 50 |
| 1,1,2-Trichloro-1,2,2-trifluoroethane-F114 | | 3 | 22 | 47 |
| Dichloromethane | 28 | 45 | 41 | 33 |
| 1,2-Dichloro-1,1,2,2-tetrafluoroethane-F113 | | 7 | 3 | 25 |
| Chloroform | | | 7 | 25 |
| 1,4-Dichlorobenzene | | | 5 | 12 |
| Tetrachloroethene | 14 | 7 | 18 | 11 |
| 1,3-Dichlorobenzene | 1 | | 1 | 3 |
| 1,1-Dichloroethene | | | | 1 |
| Bromomethane | 2 | 1 | 1 | 1 |
| Vinyl chloride | | 1 | | |
| Chloroethane | | | | |
| 1,1-Dichloroethane | | | | |
| cis-1,2-Dichloroethene | | | | |
| 1,2-Dichloroethane | | | | |
| 1,2-Dichloropropane | | | 3 | |
| Trichloroethene | 2 | 1 | | |
| cis-1,3-Dichloropropene | | | 1 | |
| trans-1,3-Dichloropropene | | | | |
| 1,1,2-Trichloroethane | | | | |
| 1,2-Dibromoethane | | | | |
| Chlorobenzene | | | | |
| 1,1,2,2-Tetrachloroethane | | | | |
| a-Chlorotoluene | | | | |
| 1,2-Dichlorobenzene | | | | |
| 1,2,4-Trichlorobenzene | | 1 | 6 | |
| Hexachlorobutadiene | | 1 | 6 | |

Table 1 indicates that fifteen of the pollutants were present in at least one percent of the samples collected each year during this four-year period. The table includes data for 782 samples. An additional twelve pollutants were present in at least one percent of the samples collected for at least one year of the four-year period, and another eleven pollutants were not present in any of the samples. Of the fifteen pollutants, which were most prevalent in the samples, ten are HAPS and are highlighted in red in Table 1. These ten HAPs also consistently had high average concentrations in comparison to the other pollutants. For these reasons, these HAPs were targeted for further study.

HAP inventory data from the year 2000 was used to further evaluate the ten HAPs chosen for this study. The Jacksonville HAP emission inventory included emissions calculated for point, area and mobile sources. Point sources are large industrial facilities that are considered major sources. Activity level data for these sources was obtained from the pollution sources and emissions were calculated individually for each emission point at each facility. Fifty-eight point sources were included in this inventory. Area sources are smaller air pollution emitting facilities such as small print shops and automobile repair shops. Area source emissions were calculated based on actual process rates for some categories such as dry cleaning facilities. Other categories were calculated using aggregated activity level data such as census and county employment data. Emissions were calculated for thirty-seven area source categories. Mobile sources include both on-road and non-road sources of air pollution. On-road sources include automobiles and trucks. Non-road sources include trains, marine vessels, airplanes and agricultural equipment. On-road mobile source emissions were calculated using vehicle miles traveled for automobiles and utilizing the Highway Performance Monitoring System (HPMS) and the MOBILE6 model[3]. Non-road mobile sources were calculated using aggregated activity level data from such sources as the Waterborne Commerce Statistics Center for Marine Vessels and emission factors were obtained from EPA's National Toxics Inventory (NTI) documentation[4].

The HAP emissions inventory was calculated for the 188 pollutants on EPA's HAP list. HAP inventory data was compiled based on Source Classification Code (SCC) to determine which source categories were emitting the ten HAPs targeted in this study. Pie charts were created for each of the ten HAPs and used to demonstrate the various sources of emissions for each.

## RESULTS AND DISCUSSION

Emission inventory data has many uses, which include determining which pollutants may be of concern in a particular geographical area, which pollution sources are contributing to emissions of a particular pollutant and in establishing trends. Several of the pollutants which were calculated in the largest mass in the inventory, are also pollutants monitored in consistently high concentrations at mobile laboratory and stationary monitoring sites.

The 2000 HAP inventory was used to determine the origin of the ten HAPs included in the study. HAP emissions were segregated into point, area and mobile sources. Figure 1 shows 2000 emission inventory data for the ten HAPs included in the study in total and by inventory section. According to the inventory, of the ten HAPs studied, the majority of emissions appear to be from mobile sources and the least emissions from point sources. Toluene is the HAP with the greatest mass emissions, mostly from mobile sources. Toluene is also emitted by area sources and a small amount is emitted by point sources. Toluene is both a significant source of emissions in the inventory as well as in the monitoring data. Four other HAPs are mostly emitted by mobile sources, they include: xylene, benzene, ethyl benzene and styrene. Xylene is the HAP with the second greatest mass emissions according to the inventory and benzene has the third highest mass emissions. Both methyl chloroform and perchloroethylene are mostly emitted by area sources and methylene chloride contributes a small portion of emissions to the area source sector. Of the ten HAPs studied, both xylene and toluene make the most significant contribution to the point sector, although the point source sector is meager for these ten HAPs.

HAP inventory data was further used to determine which source categories or types of industry were emitting each of the ten HAPS included in the study. On-road mobile source emissions account for almost sixty-percent of emissions for five HAPs. These HAPs include: toluene, xylene, benzene, ethyl benzene and styrene, see Figures 2 through 6. The second greatest source of emissions for xylene, benzene and ethyl benzene is non-road mobile sources, accounting for approximately thirty percent. Non-road mobile sources are the third highest source of emissions for toluene, accounting for seventeen percent and surface coating is the second highest source of emissions at eighteen percent. The second greatest source of emissions for styrene is area source miscellaneous organics. Miscellaneous organic emissions data were derived from the 1999 TRI. The organic chemical manufacturing facilities that reported information to the TRI but were not included in the point source inventory were classified as miscellaneous organics. Other sources of emissions for toluene, xylene, and benzene include surface coating and degreasing. Aircraft emissions account for two percent of benzene emissions and four percent of styrene emissions.

**Figure 1. Jacksonville 2000 HAP inventory data for ten HAPs.**

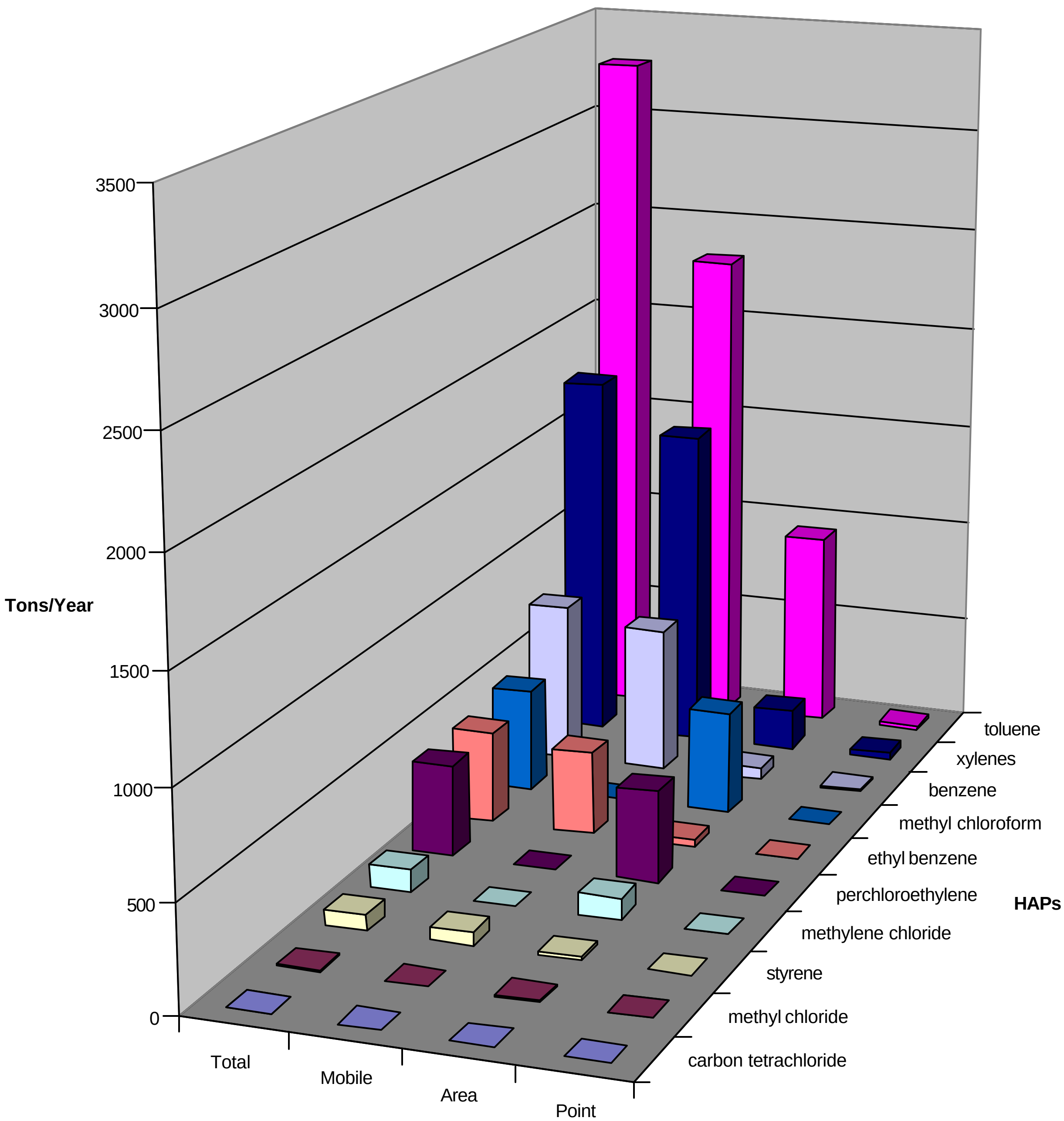


**Figure 2. Sources of toluene emissions.**

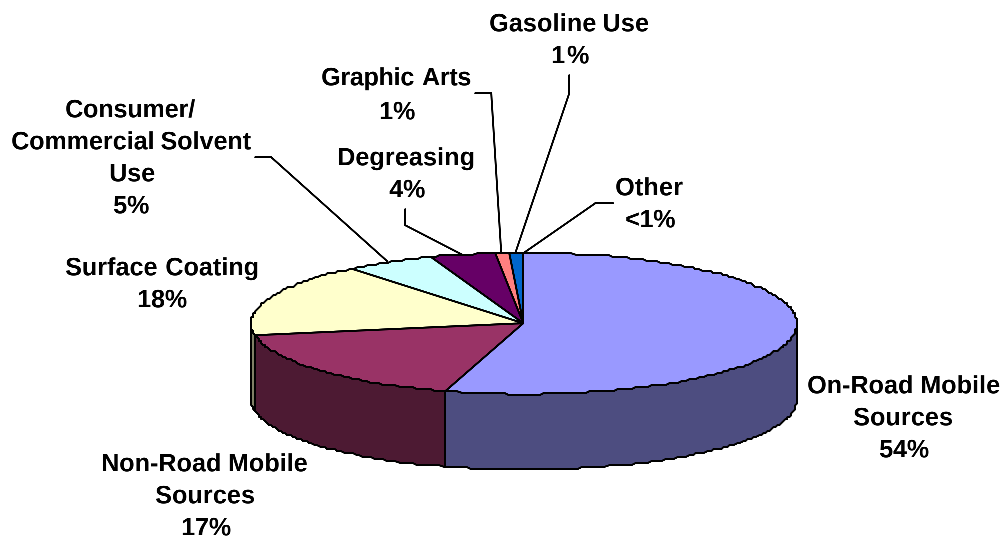



**Figure 3. Sources of xylene emissions.**

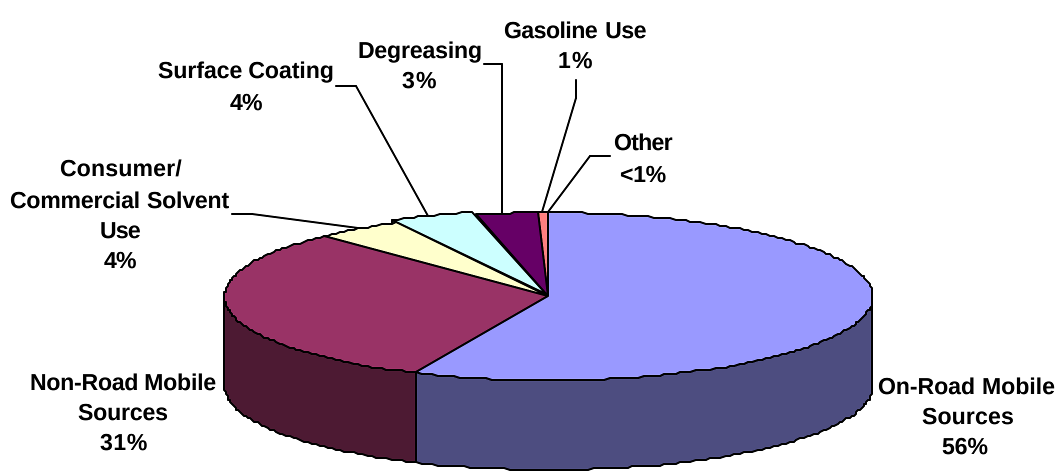


**Figure 4. Sources of benzene emissions.**

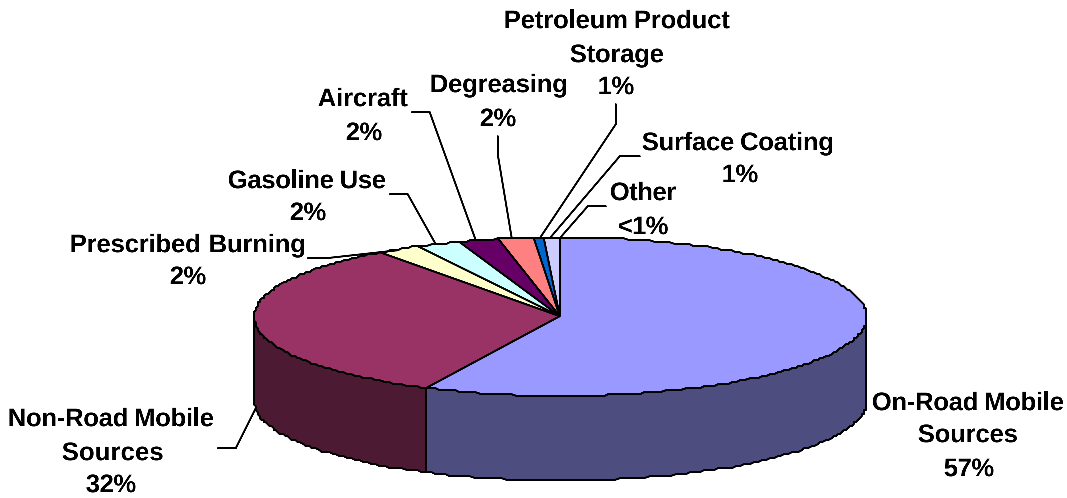



**Figure 5. Sources of ethyl benzene emissions.**

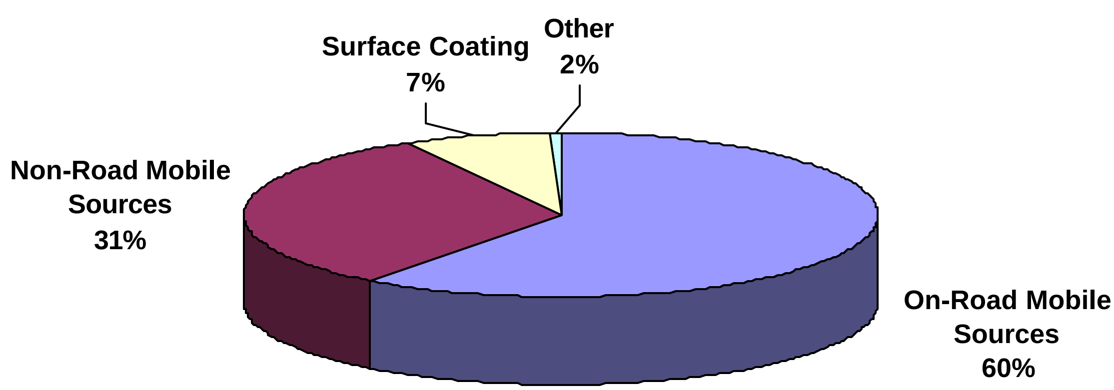
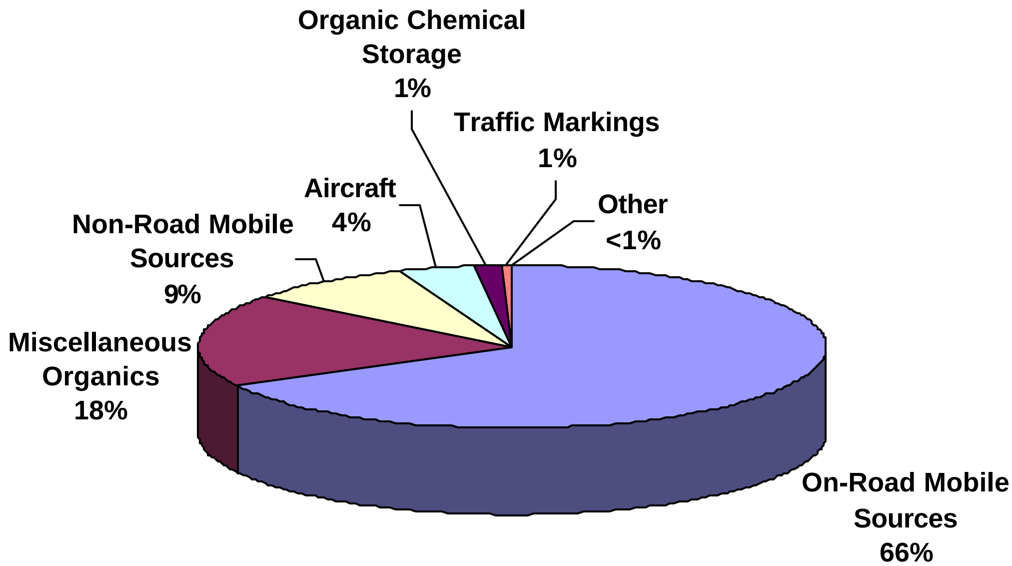


**Figure 6. Sources of styrene emissions.**




For two of the remaining HAPs in the study, methylene chloride and methyl chloroform, degreasing accounts for the majority of emissions, comprising close to sixty percent of the total emissions for each of these HAPs, see Figures 7 and 8. Commercial/consumer solvent use is a significant source of emissions for both HAPs, comprising thirty-one percent of the emissions for methyl chloroform and thirteen percent of the emissions for methylene chloride. Landfills account for a very small part of the emissions for these two HAPs and surface coating also is a source of emissions for both HAPs. Twenty-seven percent of methylene chloride emissions are from surface coating, which is the second largest source of emissions. Coal combustion from electric generation, Publicly Owned Treatment Works (POTWs) and industrial/commercial fuel combustion also are small sources of emissions for methylene chloride.

Two additional HAPs included in the study, carbon tetrachloride and methyl chloride, are emitted by various sources. POTWs account for the majority of emissions for carbon tetrachloride (96%) and five percent of the emissions for methyl chloride, see Figures 9 and 10. Landfill emissions are close to one percent of emissions for both of these HAPs. Traffic markings are the second highest source of emissions for carbon tetrachloride. Methyl chloride is mainly emitted by surface coating operations (42%), prescribed burning (32%) and coal combustion from electric generation (18%). One other minor source of emissions for methyl chloride is wild fires (3%) and another minor source of emissions for carbon tetrachloride is commercial/consumer solvent use (<1%).

The last HAP included in the study is perchloroethylene. According to the emissions inventory, emissions from perchloroethylene are mostly from dry cleaning facilities. Degreasing accounts for the second highest source of emissions at twenty-six percent and commercial/consumer solvent use contributes a small amount of emissions at three percent. See Figure 11.

**Figure 7. Sources of methylene chloride emissions.**

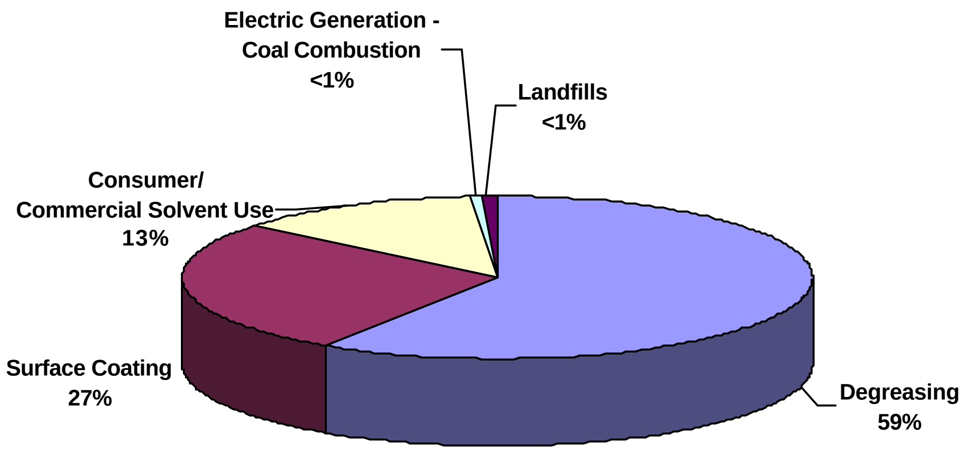



**Figure 8. Sources of methyl chloroform emissions.**

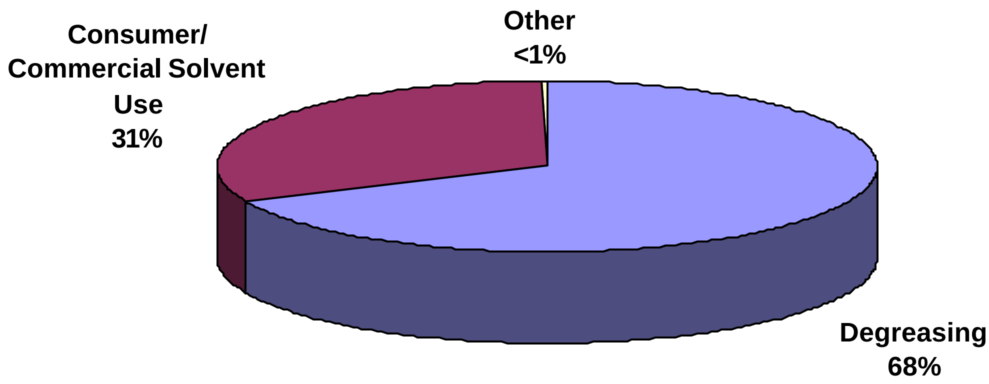


**Figure 9. Sources of carbon tetrachloride emissions.**

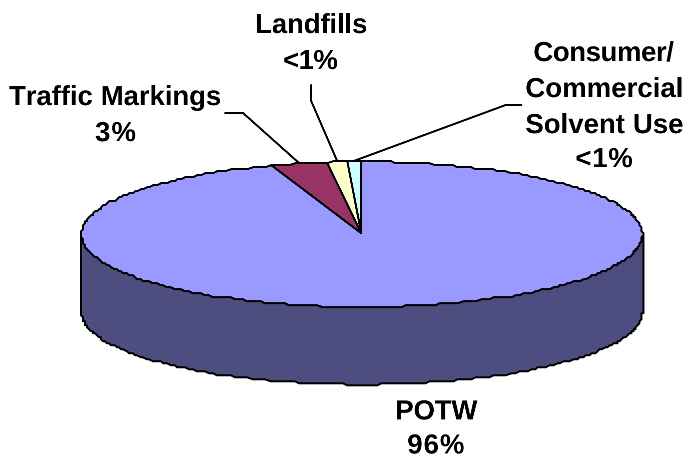



**Figure 10. Sources of methyl chloride emissions.**

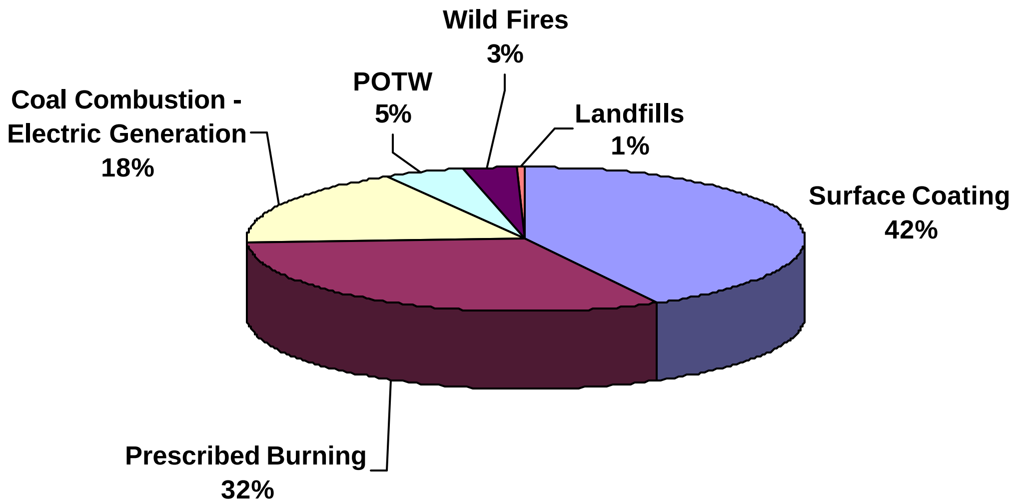
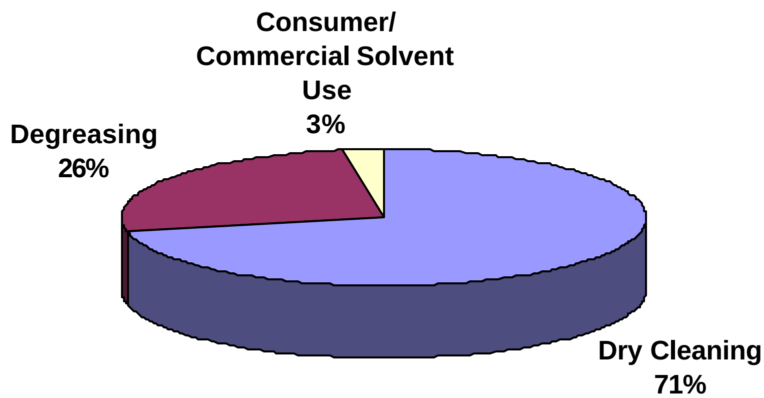


**Figure 11. Sources of perchloroethylene emissions.**



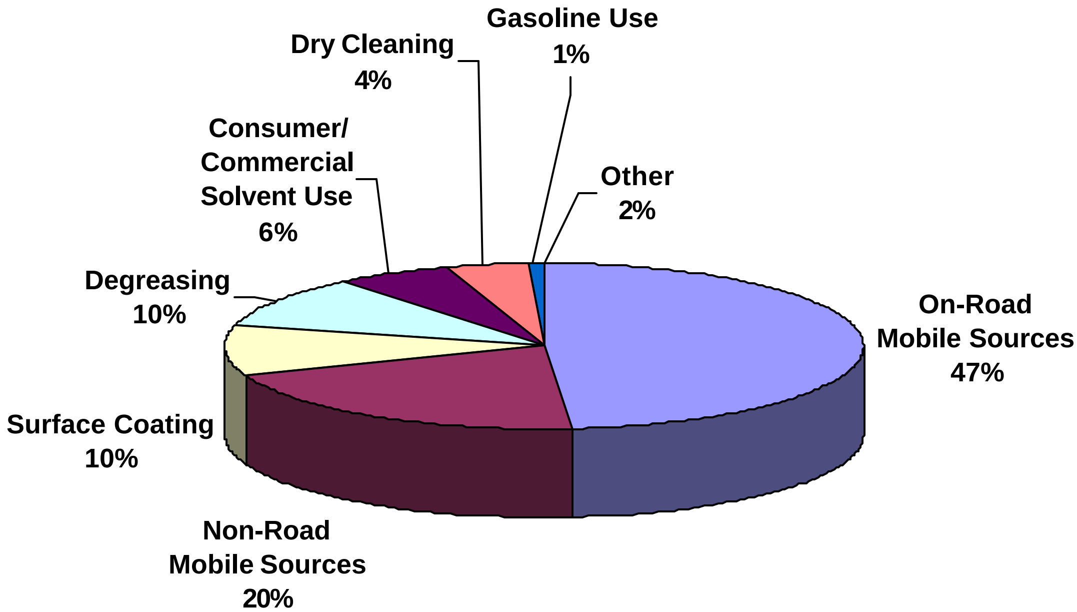

## CONCLUSIONS

The 2000 HAP emissions inventory has been a valuable tool in quantifying emissions from HAPs that were targeted by air toxics monitoring. The inventory indicates on-road mobile sources are a significant contributor (50-60%) to five HAPs found from air monitoring. Non-road mobile sources are also a significant source of emissions, contributing approximately thirty percent of the emissions for three of the ten HAPs included in the study. Seven of the HAPs are emitted from surface coating operations. Degreasing and consumer/commercial solvent use contribute to the emissions for at least six of the HAPs included in the study. Other significant contributors include dry cleaning and gasoline use.

Figure 12 indicates the sources of emissions, according to the 2000 HAP emissions inventory, for all ten HAPs included in the study. Almost half of the total emissions for all ten HAPs are from on-road mobile sources. Non-road mobile sources are the second largest source of emissions. Combining both on-road and non-road emissions, mobile sources account for the largest source of emissions for the ten HAPs included in the study, with a combined contribution of sixty-seven percent. Other sources contributing a significant amount of emissions to all ten HAPs are surface coating (10%) and degreasing facilities (10%), most of which are area sources. Dry cleaning emissions account for four percent of the emissions from all ten HAPs. Many other sources such as gasoline use, graphic arts, aircraft, prescribed burning, miscellaneous organics, traffic markings, electric generation coal combustion, wildfires, landfills, POTWs, printing/publishing, mineral products, etc contribute less than one percent to the total emissions for all ten HAPs included in the study.

The ten pollutant study has aided Jacksonville in determining which types of facilities are contributing to ambient levels of particular HAPs. This data can be used for local regulatory efforts, air pollution dispersion modeling and risk assessment. A 2002 HAP emissions inventory is currently in progress for Jacksonville. Future HAP inventories will aid in establishing trends over time, which can be used to determine the success of regulatory efforts.

**Figure 12. Sources of emissions for ten HAPs.**

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## Key Words

Jacksonville
Toxic Release Inventory
Hazardous Air Pollutants (HAP) Emission Inventory
Hazardous Air Pollutants (HAPS)
Air Toxics Monitoring
EPA Method TO-15
Mobile Monitoring Laboratory
Toluene
Xylene
Benzene
Methyl Chloroform
Ethyl Benzene
Perchloroethylene
Methyl Chloride
Styrene
Carbon Tetrachloride
Methylene Chloride
Point Sources
Area Sources
Mobile Sources