“New Molecular Markers of Asian Air Emissions – Anthropogenic Semi-Volatile Organic Compounds”
Project Summary – Section A

The primary objective of the research activity is to identify anthropogenic semi-volatile organic compounds (SOCs) that can be used as molecular markers for Asian air emissions and trans-Pacific atmospheric transport. The outreach education activity is integrated with the research proposal in that it develops curriculum with the objective of introducing underrepresented minority high school students, and their teachers, to atmospheric chemistry and atmospheric measurements through Oregon State University’s National Institute of Environmental Health Sciences funded Hydroville Curriculum Project. A curriculum will be developed to allow students to assume the role of “Air Quality Scientist” and measure air temperature, air flow, relative humidity, CO, CO₂, O₃, and volatile organic compounds in outdoor and indoor air. The students will gain an understanding of atmospheric transport and will compare measured concentrations to recommended guidelines. In addition, the research and outreach education activities include the training of two Ph.D. students through Oregon State University’s Department of Environmental and Molecular Toxicology and Department of Chemistry and the training of up to six undergraduate students through Oregon State University’s Bioresources Research Major, during the 5-year course of the funding. In addition, the research will include undergraduates and faculty from an undergraduate university (Southern Oregon University) and members of the Makah Nation.

Anthropogenic SOCs have been chosen for this research because they are more closely tied to a variety of human activities (fossil fuel combustion, biomass burning, agriculture, and industrial development), in specific geographies, than SOCs derived from biogenic emissions. In addition, there is concern that anthropogenic SOCs (some of which are persistent organic pollutants) may undergo long-range transport and deposition to high latitudes and high elevations in North America and may impact these pristine ecosystems. The research sites chosen for this research include Cheeka Peak Observatory (CPO) on the northwest tip of the Olympic Peninsula in Washington and Mt. Ashland Observatory (MAO) in southern Oregon. CPO is a remote coastal site that has been previously used to study trans-Pacific atmospheric transport, while Mt. Ashland is a new high elevation, free tropospheric site that is approximately 90 miles from the coast. Hi-volume sampling equipment, that includes a glass fiber filter for the collection of aerosol bound SOCs and polyurethane foam for the collection of gas-phase SOCs, is being used for the collection of air samples. The sampling media is extracted using Accelerated Solvent Extraction and the extracts are analyzed by gas chromatographic mass spectrometry, with electron impact and chemical ionization, for a wide range of SOCs. Source regions are identified using five-day isentropic back trajectories and measurements of CO, O₃, and aerosol mass. The goals of this research, and intellectual merit, include 1) the identification of SOC molecular markers for incomplete combustion, agricultural, and industrial Asian sources; 2) the identification of signature enantiomer ratios of chiral SOCs that may indicate Asian source ‘age’; and 3) the ability to distinguish between fossil fuel and biomass combustion sources in Asian air masses.

The broader impacts of the proposed activity include exposure of underrepresented minority high school students, and their teachers, to atmospheric chemistry and subsequent mentoring by the PI, the training of two Ph.D. students across several academic departments and the research training of up to six undergraduate students, the inclusion of undergraduates and faculty from an undergraduate university (Southern Oregon University) and members of the Makah Nation in the research, the establishment of new partnerships and collaborations between Oregon State University and the global atmospheric science community studying trans-Pacific atmospheric transport, and the establishment of a connection to a current project in the PI’s laboratory to study the atmospheric deposition of anthropogenic SOCs from Asian and North American sources to high elevation ecosystems in National Parks located in the Western United States. The results of the proposed research have significant global environmental policy implications with regard to combustion emission controls and reductions in the use of some persistent organic pollutants in developing countries.
Project Description – Section C

RESEARCH ACTIVITY
INTRODUCTION

Although it has been recognized for almost a decade that the global transport of pollutants in the atmosphere results in surface contamination in remote locations such as the Arctic (1), it is more recent that direct contamination from Asia to the Pacific Coast of North America has been identified (2). The majority of chemical specific trans-Pacific transport research to date has focused on volatile combustion derived compounds, such as CO, NOx, O3, peroxyacetyl nitrate (PAN), and non-methane hydrocarbons, and aerosol measurements (2-6). This has been appropriate because China, and likely other parts of Asia, is expected to be a growing source of CO2, NOx, and SO2 emissions due to increased fuel combustion over the next 20 years (7). The direct result of this may be increased ground level ozone concentrations in the Western United States (8-9). This would make attaining the new U.S. ozone standard more difficult and could off-set the benefits of domestic reductions in NOx and hydrocarbons in the Western United States (8-9). Other Asian combustion sources, such as biomass burning, have been detected in the northeastern Pacific region (4). Biomass burning also has the potential to impact air quality in the Western United States. The chemical measurements to date, along with the corresponding air trajectories, show that during winter and spring seasons, when air trajectories favor direct transport from Asia to the Pacific Coast, transport across the Pacific can occur in as little as 5 to 10 days (2).

The full body of developing evidence for transport of Asian air pollutants to the Pacific coast of North America includes a growing list of computer modeling simulations, observational data (including deposition of Asian dust in North America), and direct measurements in the atmosphere (2). Relatively little research has been done to understand the importance of the atmospheric transport of anthropogenic semi-volatile organic compounds (SOCs) from Asia to the Pacific Coast of North America and the United States. The inclusion of anthropogenic SOCs into these studies is highly complementary and has the potential to add new molecular markers for distinguishing between Asian and North American combustion, industrial, and agricultural sources.

Anthropogenic SOCs, generally defined as organic compounds with vapor pressures less than about 1 Pa, have been chosen for this research because they are closely tied to a variety of human activities, in specific geographies, and may impact the health of humans and the environment. SOC behavior in the atmosphere is complex. These compounds may exist in both the atmospheric gas and particulate phases, may or may not undergo photochemical degradation in the atmosphere, and may undergo both wet and dry deposition. In addition, there is concern that anthropogenic SOCs (some of which are persistent organic pollutants) may undergo long-range transport and deposition to high latitudes and high elevations in North America and may impact these pristine ecosystems (10-13). Although SOCs derived from local biogenic emissions may impact regional air quality in North America, many of these compounds have relatively short gas-phase atmospheric life-times, are less likely to undergo long-range transport, and are emitted only from biomass burning or directly from vegetation. In addition, their use as molecular markers of Asian emissions may be complicated by biogenic emissions from the Pacific Ocean, as is the case with branched hydrocarbons (14) and cholesterol (15). Potential sources of anthropogenic SOCs include combustion (polycyclic aromatic hydrocarbons), agricultural (pesticides), and industrial sources (polychlorinated biphenyls).

To date, few direct analyses of anthropogenic SOC trans-Pacific transport have been conducted. Two studies that have been conducted include a Pacific Ocean cruise as part of the SEAREX (Sea-Air Exchange) program in 1986 (14) and air samples collected in Tagish Yukon, Canada between 1992 and 1995 (16). In the Pacific cruise study, samples were collected over a period of 2-3 days and were analyzed for variety of organochlorine compounds, polycyclic aromatic hydrocarbons, organic nitrates, aliphatic hydrocarbons, and light halocarbons. This works suggests that anthropogenic SOCs, with long enough atmospheric life-times to undergo long-range transport, but short enough life-times to show enhanced concentrations during transport events, make good molecular markers (14). Potential molecular markers included polycyclic aromatic hydrocarbons, alkyl nitrate species, and trichlorobenzenes (14).
In the Canadian study, weeklong ambient air samples were collected and were analyzed for a variety of organochlorine compounds. Five-day back air trajectories were used to distinguish between weeks when Asia was the primary source and when the western United States was the primary source. The Canadian researchers found that, during winter months, increased air concentrations were primarily influenced by long-range atmospheric transport from eastern Asia (16). However, North America was a major source of hexachlorocyclohexanes and chlordane during times of the year when the air trajectories originated from North America (16). Although these researchers did not use chiral measurements to apportion sources, they recognized the utility of doing so: “…the measurement of the enantiomeric ratios (ERs) would also help to discriminate between fresh sources….and those older sources…” (16). Because Asian transport events are on the duration of 1-3 days in length, it may be problematic to attribute a longer time integrated sample, of up to a week, to a specific transport event. Ideally, if sufficient volumes of air could be collected so that the analyte concentrations were above the detection limit, shorter time periods (24 hours or less) would be sampled in order to attribute the analytes measured to a specific source region.

Some SOCs exist in biologically unmodified products or sources as racemic, chiral compounds. Specifically, SOCs that have chemical structures such that they exists in two mirror image forms, which are not super imposable on each other, have been manufactured for use in pesticides and industrial and consumer products as a racemate, where the ratio of (+) and (-) enantiomers is unity. Physical processes such as volatilization, atmospheric transport, photochemical reactions, and wet and dry deposition from the atmosphere do not differentiate between the two enantiomers because they have identical physical-chemical properties. For example, if a chiral pesticide, applied to crops in Asia, immediately volatilized into the atmosphere an enantiomer ratio (ER) of 1 would be measured in the air mass. ‘New’ sources have racemic signatures.

However, biological processes distinguish between the two enantiomers and chiral SOCs undergo enantioselective biodegradation after being deposited to soils, sediments, or water (17). The resulting SOC residue will be nonracemic, that is the ERs will be greater than or less than one. In the scenario described above, assume the sole source is no longer from immediate volatilization during pesticide use but is now volatilization from Asian soil that has been historically contaminated with the pesticide. In this case, the microorganisms are likely to have preferentially degraded one enantiomer over another (17) and the resulting ER in the soil would be greater or less than unity and the air mass would contain this same enantiomer ratio. ‘Historical’ sources have non-racemic signatures.

There is good agreement between the ER of a source (such as contaminated soil or water) and the ER of the air contaminated from that source (17-19). In addition, depending on the microbial community present, the ER for a given chiral SOC may be specific for a given source. For example, the western Arctic Ocean is depleted in the (+) enantiomer of α-hexachlorocyclohexane (α-HCH), while the Bering and Chuckchi Seas are depleted in the (-) enantiomer of α-HCH (17) and soils have different ERs than oceans (17, 18, 20). A review of the literature shows that the ERs of chiral SOCs have not been measured in Asian air masses.

There is growing interest in using the ERs of chiral organic compounds to study a wide range of environmental problems including microbial degradation in soils (20,30), surface water (28-29, 33-34), sediment (21-22) and ground water (32), air-water exchange in oceans (17,31), and biomagnification in the food chain (22-27). Although many historical and current use chiral SOCs exist, only heptachlor, heptachlor-exo-epoxide, α-hexachlorocyclohexane, cis- and trans – chlordane, o,p’-DDT, chiral PCBs and toxaphene have been studied in detail (22, 36-37).

A recent publication suggests that the use of enantiomer fraction, rather than enantiomer ratio, is a more appropriate descriptor of chiral signatures of environmental matrices (36). Historically, enantiomer ratio has been defined as:

\[
ER = \frac{A_+}{A_-} \quad (1)
\]
where $A_+$ and $A_-$ correspond to the chromatographic peak areas of the (+) and (-) enantiomers. Frequently in environmental analyses, the ER of a given sample is compared to a racemic standard with an ER = 1 (36). The ER of an environmental sample can range from zero to infinity. Not only is it difficult to represent infinity graphically, a unit change in ER toward zero is not equivalent to the same unit change toward infinity (36). ER is also difficult to use in mathematical expressions because it is not a proper fraction (36). For these reasons, the use of enantiomer fraction (EF), has been proposed, where:

\[
EF = A_+/(A_+ + A_-) \quad \text{or} \quad EF = A_1/(A_1 + A_2)
\]

(2)

\[
EF = (A_1/A_2)/[(A_1/A_2) + (A_2/A_2)] = ER/(ER + 1)
\]

(3)

and

\[
EF = 1/(1 + 1/ER)
\]

(4)

and $A_1$ and $A_2$ are the order of the eluting enantiomers on a specific chiral column when the identity of the (+) and (-) forms is not known (36). Using EF, a racemic mixture is 0.5 and the EF ranges between 0 and 1, with equivalent unit change (36). EF is more appropriate for use in mass balance calculations and can be used to apportion sources and calculate biodegradation rates (36).

The use of ERs, or EFs, has been recognized as a potentially useful tool for identifying the location and age of sources of air contamination (17, 35). Moreover, a method has been developed for using ERs for apportioning two sources of chiral compounds to the atmosphere (35). If the ERs of the two sources ($ER_a$ and $ER_b$) and the mixed air ($ER_m$) are known, the fraction ($F_a$ or $F_b$) of the chiral SOC originating from either of the two sources can be calculated (35), for example:

\[
F_a = \frac{(ER_m – ER_b)(ER_a + 1)}{(ER_a – ER_b)(ER_m + 1)}
\]

(5)

In terms of EF (36),

\[
F_a = \frac{(EF_m – EF_b)}/(EF_a – EF_b)
\]

(6)

However, this approach has yet to be used on a large, or global transport scale, to identify the chiral signatures of Asian sources or apportion multiple Asian sources.

Finally, the use of SOCs produced from incomplete combustion, specifically polycyclic aromatic hydrocarbons (PAHs), to apportion combustion sources is complex. There is some overlap in the individual PAHs that are emitted from various combustion sources, the PAH profile produced may be influenced by combustion conditions, and some PAHs may undergo gas-phase photochemical degradation in the atmosphere. Nonetheless, numerous researchers from around the world have successfully used statistical techniques, including factor analysis and multiple regression models, to quantitatively attribute the PAH signature measured in the atmosphere to various combustion sources (38-44). These combustion sources include coal, natural gas, diesel engines, gasoline engines, biomass burning, smelters, and cooking-fuels (38-44). The PAH Retene, a product from the combustion of softwood, and levoglucosan (1,6-anhydro-β-D-glucopyranose), a product of the pyrolysis of cellulose, have been used as molecular markers for biomass combustion (15, 38, 42, 45-50).

In recent years, researchers from India (51-52), Taiwan (53), China (54-58), Thailand (59-60), Vietnam (61), and Japan (62) have published on the PAH distribution and concentrations in ambient and in-door air in these countries. Atmospheric total PAH concentrations in Indian cities exceed concentrations reported internationally by 10 to 50 times (52) and PAHs are detectable in Central Pacific deep sea sediments (63) and in Pacific Ocean and Arctic air (14, 64) due to long-range transport from Asia. Published information on the PAH profiles produced from various combustion sources and the
PAH profiles measured in different countries in Asia will allow us to develop a quantitative source apportionment model for differentiating between fossil fuel and biomass combustion sources in Asia.

**RESEARCH ACTIVITY OBJECTIVES AND GOALS**

The primary objective of the research activity is to identify anthropogenic semi-volatile organic compounds (SOCs) that can be used as molecular markers for Asian air emissions and trans-Pacific atmospheric transport. Anthropogenic SOCs have been chosen for this research because they are more closely tied to a variety of human activities (fossil fuel combustion, biomass burning, agriculture, and industrial development), in specific geographies, than SOCs derived from biogenic emissions. In addition, there is concern that anthropogenic SOCs (some of which are persistent organic pollutants) may undergo long-range transport and deposition to high latitudes and high elevations in North America and may impact these pristine ecosystems.

The specific goals of this research include 1) the identification of SOC molecular markers for incomplete combustion, agricultural, and industrial Asian sources; 2) the identification of signature enantiomer ratios of chiral SOCs that may indicate Asian source ‘age’; and 3) the ability to distinguish between various fossil fuel and biomass combustion sources in Asian air masses.

**RESEARCH ACTIVITY SIGNIFICANCE**

The significance of the proposed research activities include the connection of current work by atmospheric scientists studying the trans-Pacific transport of combustion derived compounds (CO, O₃ and metals) to new molecular markers that have the potential to distinguish between fossil fuel and biomass combustion sources, as well as agricultural and industrial Asian sources. The proposed research links Oregon State University to the global atmospheric chemistry community studying trans-Pacific transport, including scientists at NOAA, Pacific Northwest National Lab (PNNL), University of Alaska, University of Washington, and international atmospheric scientists.

In addition, the PI’s laboratory is funded to participate in the U.S. Department of Interior’s Western Airborne Contaminants Assessment Project (WACAP) to study the atmospheric deposition of anthropogenic SOCs (the same compounds proposed in this study) from Asian sources to high elevation ecosystems in National Parks located in the Western United States, including two Alaskan parks, over the next 5 years. Because the WACAP sites will be located in remote locations within the parks that have no electrical power, direct atmospheric measurements of anthropogenic SOCs will not be possible. Instead, atmospheric deposition of anthropogenic SOCs will be measured in the annual snowpack and natural vegetation (lichens and tree bark (65-67)) in the parks. The WACAP sites in Olympic National Park are located approximately 200 miles from our proposed air research site at Cheeka Peak Observatory.

In the WACAP research, the SOCs will be traced through the food chain to assess the potential exposure of native people and subsistence food-eaters to Asian derived air pollutants. Sample collection in the National Parks will take place from 2003 through 2005 and will overlap in time with the research proposed herein. The proposed research and the current research in the PI’s laboratory, as part of WACAP, are highly complementary. The two programs combined have great potential to fully investigate if there is a link between the emission of anthropogenic SOCs from Asian sources and the deposition of these compounds to high elevation ecosystems of significant national importance.

**RESEARCH ACTIVITY PLAN**

**Selection of Anthropogenic SOCs.** The compounds listed in Table 1 have been selected by the PI as the target analytes for both the proposed atmospheric chemistry research and WACAP. These compounds have been chosen for the research because they represent emissions from combustion (polycyclic aromatic hydrocarbons), agricultural (pesticides), and industrial (polychlorinated biphenyls) sources. Many of the target SOCs (including PCBs and many of the organochlorine pesticides) are persistent organic pollutants. Levoglucosan has been added to the analyte list because it has been shown to be an excellent marker, along with retene, for biomass burning (15, 38, 42, 45-50). Twelve representative PCBs, two for each level of chlorination, have been chosen for the study in order to
investigate their trans-Pacific atmospheric transport based on their range of atmospheric lifetimes (see below). The target analytes that are chiral include PCB 183, o,p'-DDT, o,p'-DDD, α-hexachlorocyclohexane (HCH), cis-, trans-, and oxy-chlordanes, heptachlor, heptachlor epoxide, malathion, metolachlor, o,p'-methoxychlor, and acetochlor (see Table 1) (17-37). Table 1 is one of the most comprehensive lists of anthropogenic SOCs to be measured in ambient air. Only the USGS has attempted to measure such a wide range of anthropogenic SOCs, including organophosphate and triazine pesticides and their atmospheric degradation products, in ambient air (68).

### Table 1. Target anthropogenic SOCs.

<table>
<thead>
<tr>
<th>Polycyclic Aromatic Hydrocarbons (PAHs)</th>
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<tbody>
<tr>
<td>Analyses will also include levoglucosan as a molecular marker for biomass burning</td>
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<table>
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<tr>
<th>Polychlorinated Biphenyls (PCBs)</th>
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<tr>
<td>PCB 6 (2,3'-Dichlorobiphenyl), PCB 8 (2,4'-Dichlorobiphenyl), PCB 28 (2,4,4'-Trichlorobiphenyl), PCB 31 (2,4',5-Trichlorobiphenyl), PCB 52 (2,2',5,5'-Tetrachlorobiphenyl), PCB 74 (2,4,4',5-Tetrachlorobiphenyl), PCB 101 (2,2',4,5,5'-Pentachlorobiphenyl), PCB 118 (2,3',4,4',5-Pentachlorobiphenyl), PCB 138 (2,2',3,4,4',5'-Hexachlorobiphenyl), PCB 153 (2,2',4,4',5,5'-Hexachlorobiphenyl), PCB 183* (2,2',3,4,4',5,6-Heptachlorobiphenyl), and PCB 187 (2,2',3,4',5,5',6-Heptachlorobiphenyl)</td>
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<table>
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<tr>
<th>Organochlorine Pesticides and Degradation Products</th>
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<table>
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<tr>
<th>Other Pesticides and Degradation Products (including organophosphorus pesticides and triazine herbicides)</th>
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<tbody>
<tr>
<td>Azinphos-methyl and oxon, Chlorpyrifos and oxon, Diazinon and oxon, Dimethoate, Disulfoton, Demeton S, Ethion, Malathion*, Parathion and Parathion -methyl, Phorate, Terbufos, Disulfoton, Metolachlor*, o,p'-Methoxychlor*, Acetochlor*, Alachlor, Prometon, Triallate, Pebulate, EPTC, Carbofuran, Carbaryl, Omethioate, Trifluralin, Propachlor, Atrazine and degradation products, Metribuzin, Simazine, Cyanazine</td>
</tr>
</tbody>
</table>

*Chiral compounds

Some of the organochlorine pesticides, such as DDT and technical HCH, have been banned from use in North America but are currently used in India and other developing countries (69-70). Pesticides currently being used in North America (such as the triazine herbicides and organophosphate pesticides) are included in the target analyte list as potential molecular markers for North American agricultural emission sources. It is also possible that we may be able to link the detection of specific pesticides to the types of crops (such as rice) the pesticide is commonly used on. Knowledge about global pesticide use patterns and the crops a pesticide is used on will provide further confirmation of the source emission region for a given air mass. Some of the atmospheric and/or microbial degradation products of the pesticides (such as DDEs, oxychlordane, heptachlor epoxide, and the oxons of the organophosphates) are included as target analytes because some pesticides may undergo degradation in the atmosphere during long-range transport (see below) and some microbial degradation products may volatilize out of soil and undergo long-range transport.

In order to be suitable molecular markers, the anthropogenic SOCs must have long enough atmospheric lifetimes to undergo long-range transport in the atmosphere, but short enough atmospheric lifetimes to show a concentration enhancement in Asian air masses (14). For example, CO and O₃
atmospheric lifetimes on the order of 2-4 weeks) show enhancement in Asian air masses, while Hg (atmospheric lifetime on the order of 6 months) does not due to relatively high atmospheric background concentrations as compared to concentrations in Asia air masses (71). Data suggests that compounds with atmospheric lifetimes less than approximately 1 month show enhancement in Asian air masses (71). The estimated gas-phase atmospheric lifetimes of the target SOCs in the proposed research range from approximately 1 day to several hundred days, with the majority (80%) having estimated gas-phase atmospheric lifetimes of less than 1 month. Only the PCBs, lindane (γ-hexachlorocyclohexane), hexachlorobenzene, dacthal, and chlorthalonil have estimated gas-phase atmospheric lifetimes that exceed 1 month. The estimated gas-phase atmospheric lifetimes of the PCBs chosen for this study range from 47 days for the dichlorobiphenyls to 130 days for the heptachlorobiphenyls. Because the gas-phase PAHs have relatively short atmospheric lifetimes (on the order of 1-2 days) there is some evidence that the majority of PAHs that undergo trans-Pacific transport are the less volatile, particulate bound PAHs (63, 72).

Research Sites. Two remote research sites in the Pacific Northwest Coast region of the United States will be used to sample Asian and North American air masses for anthropogenic SOCs. The first site, Cheeka Peak Observatory (CPO), is located on Makah tribal land on the northwest tip of Washington State’s Olympic Peninsula (48.3°N 124.6°W, 480 m) (3-4). This site is operated by the University of Washington, is well established and is currently being used to study the trans-Pacific transport of CO, O₃, Hg⁰, reactive gaseous Hg, particulate Hg, and aerosols (3-4). Meteorological measurements are also made. Although this site has many advantages, including being an established, fully operational site, a drawback is that it is located close enough to the heavily traveled Strait of Juan de Fuca that NOₓ concentrations and aerosol number density measurements are influenced by the local ship traffic (4). Although BBBB and co-workers have been successful in distinguishing between the ship signal and trans-Pacific transport events (3-4), it is possible that the ship signal may complicate the interpretation of polycyclic aromatic hydrocarbon data. In addition, CPO is not a free tropospheric site and may be influenced by regional atmospheric transport. Because the historical and current measurements at CPO are well established, it is a recognized land-based site for studying trans-Pacific transport, and it is relatively close to the WACAP sites in Olympic National Park, CPO is well suited for making atmospheric measurements of anthropogenic SOCs.

The second remote Pacific Northwest Coast research site that will be used to sample Asian and North American air masses is located in southern Oregon at the Mt. Ashland ski area (42°05′ N 122°43′ W, 2282 m), approximately 90 miles from the coast and west of Interstate-5. Mt. Ashland Observatory (MAO) has not been previously used for atmospheric research and the PI would establish the site with Mr. BBBB (UW-Bothell) and be the site manager for the period of this research. Because of Mt. Ashland’s high elevation and because it is approximately 500 m higher than the surrounding peaks, it is likely to be in the free troposphere the majority of the year. Mt. Ashland is a unique site because the summit has year round power and access, it is located west of Interstate-5, and it is not down-wind of a significant population center. A National Weather Service radar dome and building are located at the summit. Based on the results from aircraft measurements (71), it is likely that trans-Pacific transport events are more pronounced at MAO than at CPO because it is above the boundary layer. In addition, Mt. Ashland is only 200 miles North of the land based, marine boundary layer site, Trinidad Head, used by NOAA during the ITCT 2K2 campaign. Initial air trajectory analyses, using HYSPLIT 5-day isentropic back trajectories, suggest that CPO, MAO, and Trinidad Head will be highly complementary sites, at times intercepting the same air mass and at times intercepting completely different air masses.

Mr. BBBB and the PI have received permission from the National Weather Service to use their facility at the top of Mt. Ashland (see Section I). A preliminary study will be conducted in September and October 2002 by the PI, Mr. BBBB (UW-Bothell), Mr. CCCC (NOAA-CMDL), and Mr. DDDD (UC-Davis). Measurements during this preliminary study will include anthropogenic SOCs by the PI, metals (rotating drum sampler), O₃, aerosols (condensation nuclei counter), and meteorology (temperature, relative humidity, and wind speed and direction). Based on the results of this preliminary
study and comparison to O₃ sonds and meteorological measurements from Trinidad Head, CA and the Medford, OR NWS office, we will determine if Mt. Ashland is in the free troposphere for significant periods of time. If Mt. Ashland is a suitable free tropospheric site, we will establish it as the only free tropospheric site that samples clean, background air in the Western United States. If the preliminary study indicates that Mt. Ashland is not a suitable free tropospheric site for intercepting Asian air masses, we will explore the suitability of other remote high elevation sites in the Pacific Northwest, with electrical power and year-round access, to establish as a land-based free tropospheric site for the duration of this research.

In addition, it is likely that the PI’s laboratory will participate in other international campaigns focused on studying Asian air emissions and trans-Pacific transport, during the 5-year course of funding. NOAA has recently received permission from Chinese agencies to establish an air research program for mercury in Beijing, as well as permission to participate in mercury measurements during several research vessel transects from Shanghai to Antarctica and the Arctic in 2003 and 2004. Very few air measurements have been made for anthropogenic SOCs along either of these transects. John Calder, Director of NOAA’s Arctic Research Office, has agreed to assist the PI in establishing links to these international research activities, with the objective of identifying anthropogenic SOC molecular markers in Asian air masses (see Section I).

The PI’s laboratory will also collaborate with Mr. EEEE (PNNL) and Ms. FFFF (University of Alaska-Fairbanks) who are beginning to measure persistent organic pollutants in air at Barrow, AK (see Section I). This collaboration may include the coordination of sampling activities, data exchanges, or the analysis of their extracts to determine enantiomer ratios of chiral compounds in samples that show strong evidence of trans-Pacific transport and overlap with samples collected at CPO and Mt. Ashland Observatory (MAO).

Methodology and Approach. Beginning in November 2001, the PI began collecting high volume air samples for anthropogenic SOCs, in both the gas-phase and particulate-phase, at CPO with help from Mr. BBBB and his laboratory personnel. Two Hi-Vol air samplers, with flow rates of 17 m³/h, were purchased with the PI’s start-up funds and one was installed at CPO. Because these are the first air samples to be collected for these analytes in remote areas of the Pacific Northwest and the atmospheric concentrations were unknown, samples were collected for up to 48 hour periods in order to obtain good signal to noise ratios for the analytes. The sampling media consists of two glass fiber filters in series, for the collection of particle-phase analytes (the first filter) and for correction of sampling artifacts due to gas-phase sorption to the particulate during sample collection (the second filter), and two polyurethane foam (PUF) plugs in series, for the collection of gas-phase analytes (first PUF) and the determination of potential analyte break-through (second PUF). The glass fiber filters were cleaned by heating at 300°C and the PUF plugs were cleaned by Accelerated Solvent Extraction with dichloromethane and ethyl acetate prior to use. The PI’s laboratory is also investigating the use of XAD resin (styrene divinylbenzene) as a sorbent for the collection of gas-phase compounds.

From November 2001 through March 2002, samples were collected for 24 or 48-hour periods, approximately every 10 days. A sample collection interval of 7 to 10 days has been used for other anthropogenic SOC air measurements, including for the Great Lakes Integrated Atmospheric Deposition Network (73). From April through June 2002, samples were collected more frequently (every 2 to 4 days) and during time periods when trans-Pacific transport events where predicted by models. These samples were collected in conjunction with the NOAA ITCT 2K2 campaign and UW’s NSF funded PHOBEA 2 campaign. Thirty samples, including several field blanks, have been collected at CPO since November 2001. Sample collection at CPO will continue until 2006 (see Timeline).

A similar research approach will be employed at Mt. Ashland. During the preliminary study in September and October 2002, samples will be collected for 24 or 48-hour periods, every 2 to 4 days. If the preliminary study indicates that Mt. Ashland is a suitable free tropospheric site, the site will be established and longer-term measurements will begin in 2004. If it is not a suitable site, other high elevation sites in the Pacific Northwest will be investigated and an alternative site established in 2004.
Measurements for anthropogenic SOCs will take place simultaneously at both CPO and MAO from 2004 through 2006. During the majority of the year, samples will be collected every 10 days, at both sites, to better understand the seasonality of trans-Pacific transport and any variation that might occur in the SOC molecular markers on a seasonal basis. During periods when trans-Pacific transport events are most likely (Spring and Fall), intensive sampling campaigns will take place at both sites. Simultaneous meteorological, CO, O₃, and aerosol measurements will be made at these sites by the PI or other investigators (Mr. BBBB – see Section I). As with ITCT 2K2 and PHOBEA 2, the PI’s laboratory will also participate in international campaigns, including NOAA’s international research programs with China, that are focused on trans-Pacific transport, during the period of funding. During the final year of the funding, in 2007, the data analysis and manuscript preparation and publication will be completed.

The sampling schedules for CPO and Mt. Ashland will require that each site be serviced every 10 days, throughout the year. CPO is currently being serviced by University of Washington personnel and the samples collected to date have been an extension of these visits. Beginning in August 2002, Mr. GGGG, an Air Quality specialist for the Makah Tribal Council as well as a Makah, will participate in the SOC portion of the research at CPO and will collect the Hi-Vol air samples (see Section I). The Makah Nation is interested in collecting Hi-Vol samples in Neah Bay, WA for the analysis of anthropogenic SOCs and participation in this research would provide experience in sample collection. At Mt. Ashland, undergraduate students and faculty from Southern Oregon University will become involved in the research and will collect the air samples. The Makah and students will be reimbursed for their travel costs to and from the site as well as for the shipping costs. These costs have been included in the budget of the proposal (approximately $40/site visit). Changing of the sampling media by individuals close to the research sites will save on travel costs, as well as include others in the research project.

The PI’s laboratory personnel will service the Mt. Ashland site approximately once a month throughout the year (approximately $200/site visit). During periods of intensive sampling (Spring and Fall) at CPO and Mt. Ashland, the PI, graduate students, or undergraduate students will be on hand to service the sampler. Travel time from OSU to Mt. Ashland is approximately 4 hours. Filter and PUF samples will be stored in sealed containers and will be preserved in an ice cooler and transported directly back to OSU or shipped overnight to OSU. The appropriate field blanks will be used to insure sample integrity and protocol quality assurance (40). Field blanks will consist of glass fiber filters and PUF plugs that are installed in the air sampler, but the sampler will not be run.

In order to increase our air sampling capacity and flexibility, we have included in the budget the purchase of two modified Hi-Vol air samplers ($3,250 each) with increased sampling rates of 40 m³/h. With a total of four Hi-Vol air samplers and the ability to sample at higher rates, we will be able to deploy two samplers at each site, as well as collect samples for shorter periods of time (24 hours or less). The ability to deploy two air samplers at each site will allow us to conduct experiments on sampling reproducibility and will allow us to sample for two consecutive 24-hour periods without servicing the site in-between.

Analytical Chemistry. Historically, 500 mL to 1 L soxhlet extractors have been used to extract filter and PUF samples. The solvent is then rotary evaporated to yield an extract volume of 1 mL or less. These historical methods are labor intensive, time consuming, and use large volumes of solvents. Earlier this year, the PI received funding from Oregon State University’s Research Equipment Reserves Fund to purchase a Dionex Accelerated Solvent Extractor (ASE) for cleaning PUF plugs and XAD resin and for extracting the analytes listed in Table 1 from filters, PUF, and XAD used in air sampling. The PI also purchased a Zymark Turbovap with start up funds to replace the use of roto-vaps for solvent evaporation. Since February 2002, the PI’s laboratory has been perfecting the use of the ASE system and developing extraction methods. Experience to date indicates that the use of ASE to extract air sampling media for anthropogenic SOCs results in significant reductions in time (1 hour as compared to 50 hours) and solvent use (150 mL as compared to 1-2 L) and results in excellent analyte recoveries (97 ± 5%).

Because of the wide range of structures listed in Table 1, extracts will be analyzed by both electron impact and electron capture negative ionization gas chromatographic mass spectrometry
Levoglucosan has been analyzed by GC/MS with (15) and without derivatization (49). The PI’s laboratory has developed the instrumental methods for the analysis of the target analytes in Table 1 on the two Agilent 6890 GC/MS systems in the laboratory, using a 30 m J&W DB-5 GC column. These methods are being used in the validation of the ASE extraction methods. Instrument detection limits in electron impact mode are on the order of 100 pg, while the detection limits in the electron capture mode are on the order of 10 pg for chlorinated SOCs. Isotopically labeled internal standards are used as surrogates to quantify the concentration of the SOCs in extracts. The stable isotopes in use include: d10-acenaphthene, d10-fluoranthene, d12-benzo[k]fluoranthene, d14- EPTC, d10-flourene, d10-phorate, d6-dimethoate, d5-atrazine, d10-phenanthrene, d10-pyrene, d8-p,p’-DDE, d8-p,p’-DDT, d12-triphenylene, d12-benzo[a]pyrene, d12-benzo[ghi]perylene, d8-γ-hexachlorocyclohexane (d6-lindane), 13C6-hexachlorobenzene, d3-endosulfan I, d4-trifluralin, d10-parathion, d10-diazinon, d13- alachlor, d6-methyl parathion, d11-acetochlor, and d7-malathion. For the chiral compounds, a 30 m BGB Analytik 172 chiral GC column will be used to separate enantiomers.

**Data Analysis and Interpretation.** Data analysis and interpretation will rely heavily on the use of air trajectories, ancillary measurements of volatile combustion derived compounds (such as CO, NOx, O3, non-methane hydrocarbon, and aerosol measurements), and information on the current and historical use patterns of the target pesticides and various fossil fuels. Air trajectories will be calculated using data from NOAA’s HYSPLIT and will be imported into the ARC/GIS program for spatial representation. The PI will collaborate with other atmospheric scientists (including Mr. BBBB) to interpret the chemical data using the trajectories and other meteorological data. The use and limitations of air trajectories and the use of current chemical measurements for identifying trans-Pacific transport events have been described elsewhere (2-6).

**Personnel.** The proposed research will be used to educate and train two Ph.D. students through OSU’s Department of Environmental and Molecular Toxicology and Department of Chemistry as well as up to six undergraduate students involved in OSU’s Bioresources Research (BRR) Undergraduate major. Two BRR junior or senior undergraduates per year will develop, implement, and complete independent research projects directly related to the proposed research, in the PI’s laboratory. Each student will spend two years, including summers, doing research in the PI’s laboratory. Upon completion of their independent research projects, the BRR students will complete an undergraduate thesis and presentation on their research and are expected to be co-authors on publications of research supported by this funding.

**Progress to Date.** Significant progress has been made on the proposed research since the PI established her laboratory in XXXX. Using the PI’s start-up funds, state-of-the-art analytical chemistry instrumentation (two Agilent GC/MS’s, ASE, Turbopav, and two GC’s) and two Hi-Vol air samplers have been purchased and installed. Analytical chemistry methods have been developed and are being validated for the measurement of the anthropogenic SOCs listed in Table 1 in ambient air samples. Experiments are being conducted to determine the break-through volume for the target SOCs in the air samples, based on ambient air temperature and the vapor pressure of the SOC. A Hi-Vol air sampler was installed at CPO in November 2001 and 30 samples have been collected, including samples collected during ITCT 2K2 and PHOBEA 2 campaigns. In addition, the PI has done significant ground work to establish Mt. Ashland as a free tropospheric site, including three exploration trips to the site with other atmospheric scientists. The PI’s laboratory is beginning to extract and analyze the air samples collected from CPO and have confirmed that the field blank concentrations are extremely low and that PAHs and several pesticides are measurable in the air samples.

**Communication of Results.** A number of publications and presentations will result from this research over a period of five years, including:

- Identification of unique anthropogenic SOC molecular markers for Asian and North American air masses
• Concentrations and enantiomer ratios of anthropogenic SOCs in the free troposphere and relationship to measurements in high elevations of Western National Parks
• Collaborations with other international campaigns
• The use of automated solvent extraction to clean sampling media and extract air samples for SOC analysis
• Appropriate sampling media and breakthrough volumes of organophosphate and triazine pesticides in air research
• Enantioselective separation methods for chiral SOCs

Publications are likely to be submitted to *Environmental Science and Technology*, *Geophysical Research Letters*, *Journal of Geophysical Research*, *Atmospheric Environment*, and/or *Analytical Chemistry*. Presentations will be made at national and international conferences and may include American Geophysical Union, American Chemical Society, and Society for Environmental Toxicology and Chemistry.

OUTREACH EDUCATION ACTIVITY

INTRODUCTION

In September 2000, Dr. HHHH, a member of the Oregon State University’s Environmental Health Science Center, was awarded a seven-year grant by the National Institute of Environmental Health Sciences (NIEHS) to develop the Hydroville Curriculum Project (see Section I). The primary objective of this outreach education activity is to use real world environmental health science scenarios to help high school students develop content and process skills and improve overall academic performance (74). Some of the goals for students in this project include developing skills in critical thinking, problem solving, and communication, enhancing student knowledge of environmental health science concepts and issues, increasing student awareness of careers in science, and for students to interact with professionals and scientists in fields required for solving the real world scenarios (74). Some of the professional development goals for teachers in this project include increasing their knowledge of current scientific research, developing skills in team teaching, and gaining an understanding of environmental health science issues (74).

The curriculum for OSU’s Hydroville Project is being evaluated, expanded, and adapted for widespread use in classrooms using a Curriculum Development Team of high school teachers and students in OSU’s Science and Math Investigative Learning Experiences (SMILE) Program. The SMILE program was established in 1988 and provides math and science experiences to over 700 minority students and other disadvantaged students in 38 schools in ten school districts across Oregon (74). The minority students in the program primarily include Native American and Hispanic students that have been historically underrepresented in science (74). SMILE students have a high school graduation rate that is 10% higher than Oregon’s average and 80% of high school students involved in SMILE have gone on to higher education (74). In 1999, the SMILE program received the Award for Excellence in Science, Mathematics, and Engineering Mentoring from the Office of the President (74).

Following testing in the SMILE program, the Hydroville curriculum will be expanded to ten pilot schools (approximately fifteen classrooms) in the State of Oregon, including an inner city school in Portland (74). During the last two years of the project, the curriculum will be disseminated nationwide through teacher training workshops. The long-term vision is that the Hydroville curriculum will be available to students and teachers globally through the Internet.

Using simulated occurrences and real data, the Hydroville Challenge Problems deal with a variety of real-world scenarios, including an indoor air quality problem (74). In this scenario, a middle school is being renovated. Teachers, staff, and students attending the school complain of malodors and feeling sick. To solve this problem, high school students role-play forming a consulting firm to work with air quality scientists and school staff to determine if there actually is an air quality problem in the school.
When the problem is identified, the students develop a remediation plan and role-play presenting it at the next school board meeting.

OUTREACH EDUCATION OBJECTIVES, GOALS, AND SIGNIFICANCE

The primary objective of the outreach education activity is to develop the curriculum for the role of “Air Quality Scientist” for OSU’s Hydroville Curriculum Project and to expose underrepresented high-schools students and their teachers to atmospheric chemistry and atmospheric measurements that are relevant to their everyday lives. The goals of the curriculum will be to 1) introduce students to the concepts of in-door and out-door air quality; 2) introduce students to the concepts of air flow and atmospheric transport; and 3) introduce students to simple measurement techniques for measuring and evaluating air quality. In addition, the PI will serve as a mentor by email, phone, or in person to teachers and students using the Hydroville curriculum, putting a name and face on the role of “Air Quality Scientist”. The significance of this plan is that underrepresented high school students and their teachers will get first-hand experience in atmospheric chemistry and atmospheric measurements and will have personal interaction with a scientist working in the field.

OUTREACH EDUCATION PLAN

In XXXX-XXXX, the curriculum for the role of “Air Quality Scientist” will be developed by the PI and a video and website will be developed by Hydroville staff for the indoor air quality scenario. This process will begin in early XXXX, with the establishment of an in-door air quality curriculum advisory board, chaired by the PI, that will insure that the resulting curriculum is most relevant to real-world situations. Once the curriculum is established, it will be tested in teacher workshops on the OSU campus in late XXXX and XXXX. During the teacher workshops, the teachers will tour the PI’s laboratory, interact with the PI and the PI’s laboratory personnel, and will be exposed to the atmospheric chemistry research, including the methods used to sample large volumes of ambient air and resulting measurements, being conducted in the research activity of this proposal. This approach will allow the graduate students working in the PI’s laboratory to take part in the professional development of the participating teachers.

Teacher feedback will be used to refine the curriculum and it will be tested in the pilot schools during XXXX and early XXXX. Ten pilot high schools for this scenario will be selected. Based on results of the success of the curriculum, the curriculum may be further refined before dissemination to schools throughout the state and posting on the internet during XXXX and XXXX. From the beginning of the teacher workshops in XXXX, until the completion of roll out of the curriculum on the internet and in the schools in late XXXX, the PI will serve as a mentor to students and teachers by email, phone, and in person.

In order to be successful, the measurement techniques recommended and used for this curriculum must be relatively inexpensive, easy to use, and robust. In addition, the high school students must be able to make measurements that are relevant. Students should be able to make measurements that include air temperature, air flow, relative humidity, CO, CO₂, O₃, and formaldehyde or other volatile organic compounds (VOCs) in both in-door and out-door air. Air temperature and humidity will be measured with relatively inexpensive thermometers (~$25) and hygrometers (~$25). Air flow will be measured with the use of smoke tubes (set of 10 for ~ $100) or a hand held stick anemometer (~$129). We will explore the use of a manual gas pump (~$165) and colorimetric tubes from RAE Systems or Sensidyne for the measurement of CO, CO₂, O₃, and VOCs (~$3 per tube).

The experiment will involve having the students make atmospheric measurements at control sites outside of the school building and at various locations within the school building. Air temperature, air flow, relative humidity, CO, CO₂, O₃, and VOCs will be measured in the air intake for the HVAC air handlers and in the room return air. Students will calculate room air turn over rates and will compare measured concentrations to recommended guidelines. Based on the experimental data and recommended guidelines, the students and teachers will assess if there is a health risk due to their exposure to the indoor air. Part of the curriculum development by the PI and the graduate students working in the PI’s laboratory will include optimizing the measurements the high school students will make.
EVALUATION OF OUTREACH EDUCATION ACTIVITY

The success of the outreach education activity will be evaluated by the participating teachers and students. Evaluation surveys will be administered to teachers and students pre- and post-participation in the curriculum (74). These surveys will evaluate changes in the students' views about atmospheric chemistry and the importance of air quality in everyday life, students' attitudes about environmental health and decision making, and the way students approach problem solving (74). The data will include attitudinal survey-type data, problem solving responses, observational data from the classroom, and general feedback (74). A control group will consist of similar schools to the project schools that have not participated in the curriculum (74). Analysis of variance will be used to compare pre- and post-survey results and multivariate correlation techniques will be used to predict changes in pre- and post-survey changes from student characteristics such as gender (74). End points that indicate successful curriculum include: increased awareness of the impact of air quality on everyday life, greater confidence in discussing science within a group, and an increased awareness of the presence of chemicals in the environment (74).

INTELLECTUAL MERIT AND BROADER IMPACTS OF THE PROPOSED RESEARCH AND EDUCATION ACTIVITIES

The intellectual merit of the proposed activity includes the identification of anthropogenic semi-volatile organic compounds (SOCs) that can be used as molecular markers for Asian air emissions and trans-Pacific atmospheric transport. Specifically, this includes 1) the identification of SOC molecular markers for incomplete combustion, agricultural, and industrial Asian sources; 2) the identification of signature enantiomer ratios of chiral SOCs that may indicate Asian source ‘age’; and 3) the ability to distinguish between various fossil fuel and biomass combustion sources in Asian air masses. In addition, the intellectual merit of the proposed activity includes the development of a curriculum that introduces underrepresented minority high school students, and their teachers, to atmospheric chemistry and atmospheric measurements through Oregon State University’s NIEHS funded Hydroville Curriculum Project. A curriculum will be developed to allow students to assume the role of “Air Quality Scientist” and measure air temperature, air flow, relative humidity, CO, CO₂, O₃, and volatile organic compounds in outdoor and indoor air. The students will gain an understanding of atmospheric transport and will compare measured concentrations to recommended guidelines.

The broader impacts of the proposed activity include exposure of underrepresented minority high school students, and their teachers, to atmospheric chemistry and subsequent mentoring by the PI, the training of two Ph.D. students across several academic departments and the research training of up to six undergraduate students, the inclusion of undergraduates and faculty from an undergraduate university (Southern Oregon University) and members of the Makah Nation in the research, the establishment of new partnerships and collaborations between Oregon State University and the global atmospheric science community studying trans-Pacific atmospheric transport, and the establishment of a connection to a current project in the PI’s laboratory to study the atmospheric deposition of anthropogenic SOCs from Asian and North American sources to high elevation ecosystems in National Parks located in the Western United States. The two programs combined have great potential to fully investigate if there is a link between the emission of anthropogenic SOCs from Asian sources and the deposition of these compounds to high elevation ecosystems of significant national importance. The results of the proposed research have significant global environmental policy implications with regard to combustion emission controls and reductions in the use of some persistent organic pollutants in developing countries.

PRIOR RESEARCH AND EDUCATION ACCOMPLISHMENTS

The PI began her position as an Assistant Professor at Oregon State University, with a joint appointment in the Department of Environmental and Molecular Toxicology and Department of Chemistry, on XX/XX/XX. Prior to this position, the PI worked as a senior environmental and
atmospheric chemist in the consumer product industry for six years. She received her Ph.D. in Chemistry from Indiana University in XXXX, under the direction of Professor IIII.

Throughout her career, the PI has conducted research on the regional and global environmental fate and transport of anthropogenic semi-volatile organic compounds and has published in the premier journals in her field, including Nature, Science, Environmental Science and Technology, and Environmental Toxicology and Chemistry. As a graduate student in XXXX, she was awarded the American Chemical Society – Environmental Chemistry Division’s Graduate Student and Graduate Student Paper awards. In XXXX, she was an invited speaker at a Gordon Conference in her field. In XXXX, she was awarded the Society of Environmental Toxicology and Chemistry/Roy F. Weston Environmental Chemistry Award that is given each year to an international scientist that has made significant contributions to the field of environmental chemistry and is under the age of 40. The PI has organized and will chair a session on “Atmospheric Fate Processes” for the November XXXX annual meeting of the Society of Environmental Toxicology and Chemistry that will bring together scientists working on WACAP with atmospheric scientists working on trans-Pacific transport.

CAREER GOALS AND GOALS OF THE DEPARTMENT

The career goals of the PI include working collaboratively with international scientists in industry, government, and academia to understand the global atmospheric transport, deposition, and accumulation of anthropogenic semi-volatile organic compounds that have potential to impact human health and the health of the environment. The PI’s career goals also include introducing young women, and other underrepresented students, to real people (like herself) who are working in atmospheric chemistry and environmental science related fields, as well as introducing them to real-world scientific problems and solutions. The PI left a successful career in industry, not only to pursue fundamental research, but to coach, mentor, and encourage young people in exploring science related careers.

In addition, this Career Development Plan fits well with Oregon State University’s and the Department of Environmental & Molecular Toxicology’s goals. OSU aspires to stimulate a lasting attitude of inquiry, openness, and social responsibility. To meet these aspirations, OSU is committed to providing excellent academic programs, educational experiences and creative scholarship. The goals of the Department of Environmental & Molecular Toxicology include increasing the understanding of the benefits and hazards of chemical uses through quality education, research, and service. The department emphasizes advancement and application of science and technology to raise environmental quality and human health to the highest attainable level.

CONCLUSION

The Career Development Plan outlined in the field of Atmospheric Chemistry integrates atmospheric transport and atmospheric measurements into both the research and education outreach activities. This plan not only builds a firm foundation in research and education for the PI’s academic career, but also links Oregon State University to the greater atmospheric science community and introduces underrepresented high school students and teachers to atmospheric chemistry.
## RESEARCH AND OUTREACH EDUCATION PLAN TIMELINE

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References Cited – Section D


71. Jaffe, D., University of Washington-Bothell, personal communication.

