

ATMOSPHERIC POLLUTANT LOADING TO OLYMPIC, MOUNT RAINIER, AND NORTH CASCADES NATIONAL PARKS: Spatial and Temporal Variability and Linkages To Trans-Pacific Air Masses

**“PNW Snow Project”
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Abstract.

The purpose of this project was to complement the Western Airborne Contaminants Assessment Project (WACAP), a project sponsored by the National Park Service – Air Resources Division (NPS-ARD) in response to findings that toxic airborne contaminants were increasingly being found in the world's most pristine alpine and polar ecosystems, far from where such chemicals were produced or used. In the WACAP study, atmospheric deposition was measured by collecting 1-3 samples of the seasonal snowpack annually in each of 8 national parks (NPs) in Alaska, Washington, Montana, California, and Colorado. In this study, commonly referred to as "Pacific Northwest (PNW) Snow", more intense snow and precipitation sampling was done in Mount Rainier (MORA), Olympic (OLYM), and North Cascades (NOCA) NPs. Objectives were to quantify factors affecting variability in contaminant deposition, and evaluate the relative influence of local, regional, and global sources on one area of the larger study.

In MORA, the highest fluxes of both Current-Use Pesticides (CUPS) and Historic-Use Pesticides (HUPS) were found at elevations of 1500-2000m, with fluxes decreasing at higher elevations. Local meteorological conditions appear to favor both higher precipitation amounts, and higher concentrations of many contaminants in deposition at these elevations. In OLYM, concentrations of pesticides were generally higher during late winter and spring relative to late fall and early winter months. Within NOCA and MORA, no strong differences in fluxes were observed between sites on opposite sides (West-East or North-South) of the parks, suggesting that local sources of contaminants were not dominant during the sampling period. Bulk precipitation sampling proved to be a useful approach for providing information on fluxes of contaminants for discrete time periods, and for supplementing seasonal snowpack sampling at sites vulnerable to rain-on-snow or mid-winter snowmelt events.

Introduction

a. Problem statement

The enabling legislation for MORA, NOCA, and OLYM specifically recognized glaciers, snowfields, and alpine meadows as reasons to establish these parks, and as prominent resources these parks are charged to protect. Melting snow and glacial runoff are the main sources of water for streams and lakes in all three parks. The health of aquatic ecosystems in MORA, NOCA, and OLYM depends on the purity of the snowmelt. Additionally, all 3 parks are designated Class I airsheds under the Federal Clean Air Act, which specifies that no significant degradation of water quality may occur as a result of air pollution in the parks. All 3 parks contain a high proportion (up to 96%) of designated wilderness, and atmospheric contaminants are a primary threat to water quality.

Once deposited, many pollutants, particularly semivolatile organic compounds, accumulate and concentrate in food webs, threatening the viability of aquatic and terrestrial ecosystems. In addition to probable ecosystem impacts, there are specific human health concerns related to animal and plant food sources exposed to airborne pollutants. Higher trophic levels (terrestrial and aquatic predators)

bioaccumulate toxic materials, delivering concentrated doses of anthropogenic pollutants to people who consume them.

Glacial melt and snowmelt contribute to primary water sources for residents of high mountain and Arctic areas. Many communities obtain their drinking water from watersheds supplied almost entirely from snow and glacier melt water. Snowmelt contributes to primary water sources for over 5.6 million people living in the Greater Puget Sound area of Washington State, and Vancouver BC. These water sources may contain unknown quantities of airborne contaminants.

This project addressed a lack of information about precipitation chemistry at high-elevation sites in the PNW National Parks, which hampered the ability of resource managers to protect resources. In this project, a synoptic survey of snowpack chemistry in the parks was performed to provide resource managers with critical information to monitor aquatic resources in the parks, comply with Federal regulations, and inform the public and regulators regarding potential effects of atmospherically-deposited contaminants on ecosystems, park visitors and local residents.

b. Background

Mounting evidence indicates that airborne pollutants derived from human activities in Asia, Europe and the United States circulate throughout the northern hemisphere and deposit in western North America from the Arctic to central California. Donald (1999) and Blais (1998) describe the magnitude of the potential threat to North American ecosystems from these trans-Pacific contaminants. Montane landscapes intercept these air masses since they traverse the Pacific Ocean at relatively high altitudes (~ 2000 meters above sea level). Snow is efficient in removing pollutants (particularly semivolatile organic compounds or SOC_s) from these air masses and depositing them in high elevation terrestrial and aquatic ecosystems (Wania 1999; Franz 1998). This is important because snow is the dominant form of precipitation in many high elevation ecosystems in the West.

Blais et al. (1998) described a broad array of persistent organic airborne contaminants found in late winter snow pack throughout the Canadian Rocky Mountains; some of these pollutants were more concentrated at higher elevations. Identical techniques documented similar results in the Sierra Nevada (Landers, D.H., unpubl. data). Winter snows in Tagish, Yukon (western Canada) contained unusually high concentrations of HCH_s, chlordanes, and DDT. These events correlated with 5-day, trans-Pacific atmospheric transport from Asia (Bailey et al. 2000). Many contaminants attributed to trans-Pacific air masses in Bailey's (2000) work are banned from use in North America (e.g. PCB, DDT, HCH and Chlordane).

Simonich and others (1995) reported a northern hemispheric trend of increasing lighter congeners of PCB_s occurring with increasing latitude. This supports Wania and Mackay's (1993) suggestion that global distillation of organic contaminants may be occurring, through which organic compounds with low vapor pressures move toward colder areas of the biosphere such as the poles, and upward in mountainous regions. Through this "cold condensation" phenomenon, higher elevation ecosystems such as montane landscapes may become sinks for SOC_s (Wania, 1993; Simonich 1995). Through snow chemistry analysis, Donald (1999) and Blais (2001) discovered that SOC_s deposited onto glaciers can be

buried for years or decades, only to re-emerge as a threat to ecosystems as glaciers begin to melt. Airborne pollutants of U.S. origin also affect montane areas. McConnell, et al. (1998) documented regional sources of current-use pesticides deposited in remote, montane national park locations in California.

Transport of Asian air pollution across the Pacific Ocean to North America is well documented (Jaffe et al., 1999; Jaffe et al. 2003). Scientists suspect that air masses carrying these contaminants, particularly those originating in Asia, contain persistent toxic elements, organic compounds, acidic compounds and nutrients having documented adverse ecological and human effects (Bailey, et al. 2000). In the future, emissions from Asia are expected to increase dramatically as these developing industrial economies continue to grow.

There are no published inventories of the contaminants contained within Asian air masses, and little was known about the spatial and temporal deposition patterns of these materials in North America or their bioaccumulative impacts on receptor ecosystems. However, due to transport and deposition characteristics of these chemicals, it was suspected that remote, high elevation ecosystems in western North America were receiving inputs of these toxic materials that could have quantifiable, deleterious impacts.

To understand the pollutant types and quantities potentially involved with this phenomenon, we needed additional scientific information concerning deposition of trans-Pacific air toxics in ecosystems of the North America. Recognizing this as a significant threat to national parks, the NPS Air Resources Division launched the Western Airborne Contaminant Assessment Program (WACAP) in 2002. The purpose of this study was to determine the presence and ecosystem impacts of trans-Pacific airborne toxics over an area encompassing the range of high-elevations and northern latitudes in National Parks of the western United States. During the WACAP study, scientists sampled several matrices in eight NPs from the Arctic to central California, and east to the Rocky Mountains. Among Pacific Northwest parks, Olympic and Mount Rainier NPs were included in the WACAP project.

c. Specific objectives addressed

The purpose of this project was to complement the WACAP effort, adding more intense snow sampling in National Parks of the Pacific Northwest, the area of the U.S. that is the most frequent recipient of Asian air masses. This project sampled and analyzed snow and bulk precipitation over 3 years at sites in OLYM, MORA, and NOCA. Through this analysis, we determined the seasonal flux, and spatial and temporal variability in semivolatile organic compounds attributable to snow deposition, and examined the role of geospatial factors such as location and elevation in determining contaminant fluxes.

d. Principal project managers

Donald Campbell of the USGS in Denver Colorado oversaw the project and managed snow sample collection. Dr. Staci Simonich of Oregon State University in Corvallis managed sample processing and analysis. Dr. Dan Jaffe of University of Washington in Bothell managed atmospheric modeling and linkages to atmospheric data. Dr. Dixon Landers of the US-EPA in Corvallis Oregon coordinated with WACAP and other projects. Marilyn Erway of Dynamac Corporation in Corvallis managed data including

quality assurance, and coordinated logistics with park units. More information about the investigators can be found in the WACAP Research Plan (Landers et al. 2003).

Methods

a. Approach and Methods

Each park provided unique opportunities to meet the objectives of the study. In MORA, an elevational gradient spanning over 1500 m on the south side of Mt. Rainier from Paradise to Camp Muir was emphasized, with additional samples spatially distributed on the northwest and northeast sides of the mountain. In North Cascades NP, spatial distribution on east-west and north-south axes was emphasized. In Olympic NP, bulk precipitation sampling at a single site was emphasized in order to evaluate temporal variability of contaminant deposition.

In water years 2003-2007, a total of 42 snowpack and 11 bulk precipitation samples (including field replicates) were collected and analyzed from the 3 parks as part of the WACAP and PNW Snow projects. Samples were collected for method development during FY03 and FY04; samples were collected as part of WACAP in FY03-FY05; and samples were collected for this project in FY05-FY07. Snowpack samples were collected in March in MORA and OLYM, and in April in NOCA. Bulk precipitation samples were collected from December through March in OLYM. Seasonal snowpack samples were also collected in OLYM in March of 2004, 2006, and 2007 (in other years, seasonal snowpack sampling was not feasible because warm temperatures or rain-on-snow events caused snow ablation during the snow accumulation season).

Site selection and samples collected

Specific site selection criteria for snowpack and bulk precipitation collectors were similar, and are described in more detail in Landers et al., 2003. Sampling locations had to be free from avalanche activity and reasonably accessible to sampling crews. Samples were collected in clearings or open areas that were uniform and free of human effects or other disturbances such as excessive tree litter or animal activity. Sites were located at least 200 m away from potential local sources of contamination such as roadways, snowmobile trails, or aircraft landing zones. Where feasible, location of sampling sites near other data collection sites (eg. Surface water sampling sites, glacier monitoring sites, or weather data collection platforms) enhanced the value of the deposition data. Sample collection for this project was done collaboratively with other projects including WACAP, the Pacific Northwest Critical Loads project, NPS glacier studies, and others.

Specific sampling sites changed from year to year based on snowpack conditions, and logistical and safety considerations. General site selection issues for each park are discussed below. For snowpack sample collection, a team of NPS personnel from each of the PNW parks--were assembled and trained by USGS. The contributions of Rebecca Lofgren, Bill Baccus, Mike Larrabee, and Stefan Lofgren were key to the success of the sampling effort and the interpretation of the data in this study. NPS staff collected all samples in all three parks, with additional help as needed from USGS, and others. Utilizing a locally-

based team facilitated sampling during most favorable weather conditions, which was necessary for helicopter access and to ensure safety of personnel. Bulk precipitation samples were collected by NPS personnel and volunteers based near the sample collection site at Hurricane Ridge in Olympic NP.

Olympic NP:

Mid-winter rain-on-snow and melt events confound snowpack sampling results at most sites that can be accessed on the ground, and remote access by helicopter is discouraged because of environmental issues. Therefore bulk precipitation sampling at a site near the Hurricane Ridge visitor center was used, and determination of temporal rather than spatial variability was emphasized (Figure 1). Contaminant deposition was measured by analyzing individual samples collected over intervals of 11 to 67 days. Snowpack samples were also collected at Hurricane Ridge in 2004, 2006, and 2007; and at Hoh Lake in 2004 and 2006.

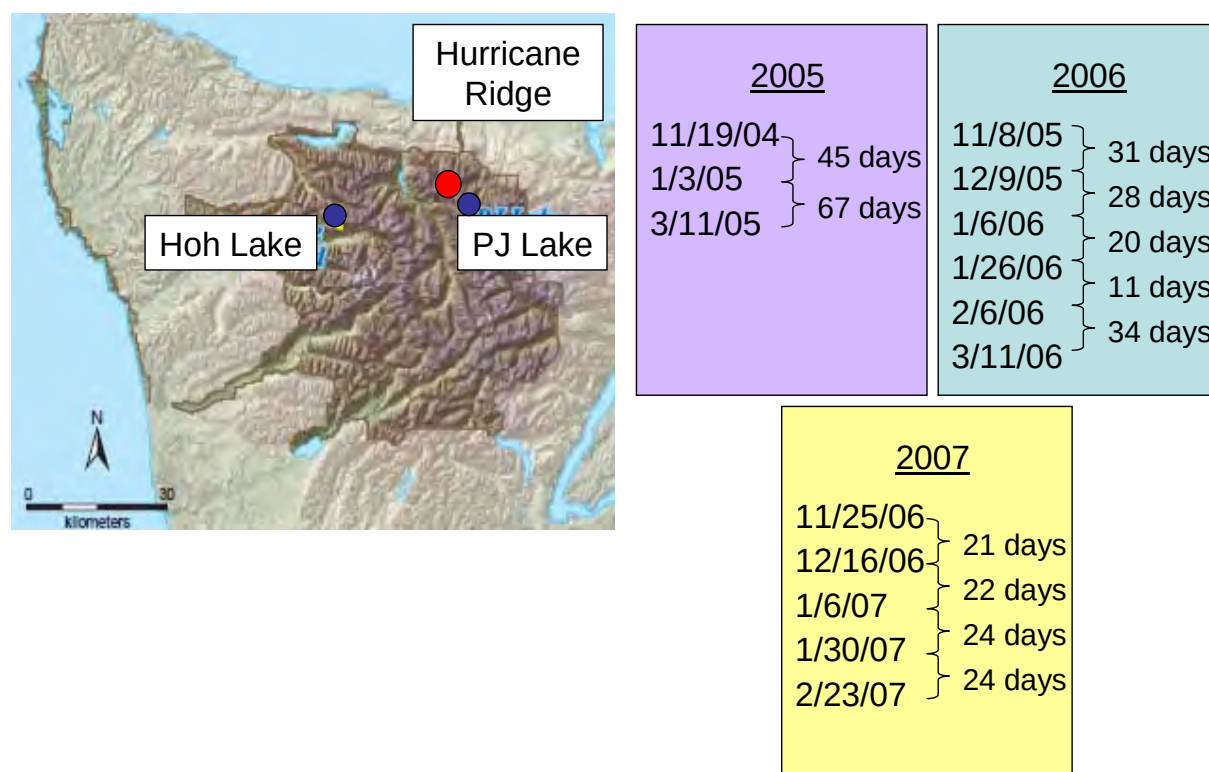


Figure 1: Location of snowpack sampling sites and primary WACAP sampling sites, (Hoh Lake and PJ Lake). Bulk precipitation samples were collected at Hurricane Ridge in 2005, 2006, and 2007 on the dates shown in the colored boxes. The number of days between sampling dates is also given.

North Cascades NP:

Because of the remote nature of this park, access was by helicopter. Snow sampling sites were co-located with long-term sites used for glacier monitoring by NPS, and the sampling was done by the glacier monitoring field crews during their annual glacier surveys (Figure 2).

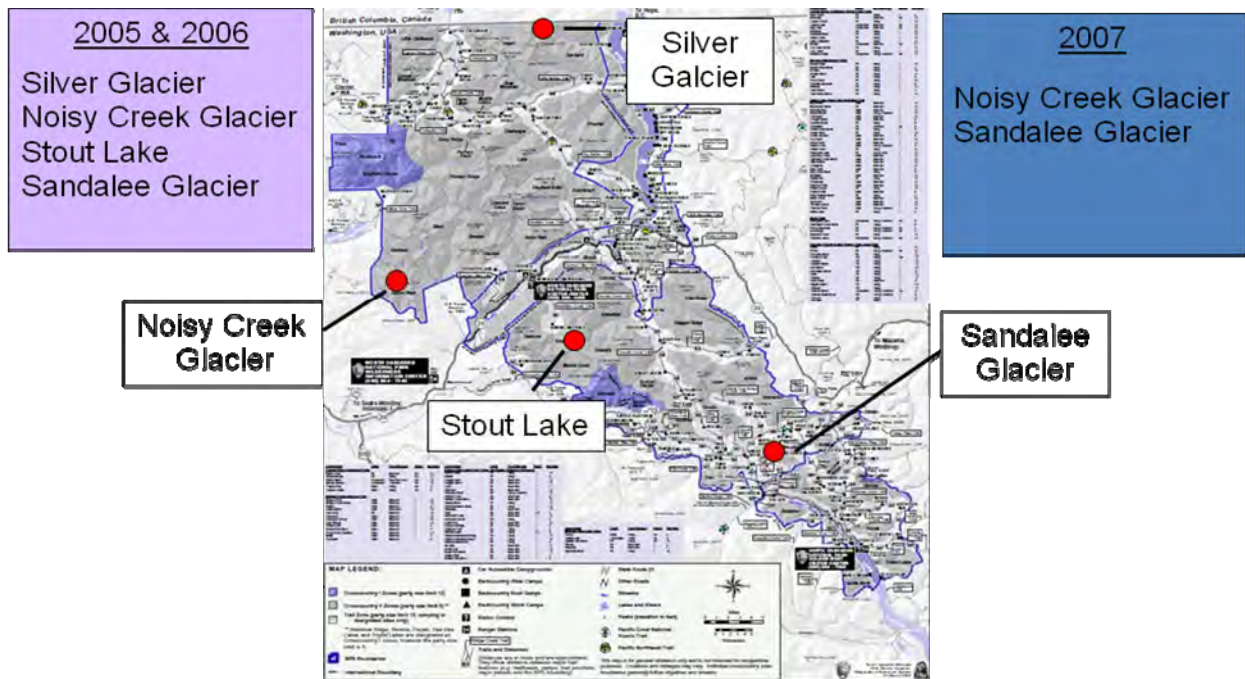


Figure 2: Sampling sites at North Cascades NP and the years in which the sites were sampled.

Mount Rainier NP:

The sites for Mount Rainier focused on an elevational transect spanning 1500m from Paradise to Camp Muir (Figure 3). Spatial variability was also assessed by sampling on the northeast and northwest sides of the mountain. The Paradise and Mowich Lake sites were near the lakes that were monitored for the PNW Critical Loads project, and bracket the WACAP project lake sampling sites.

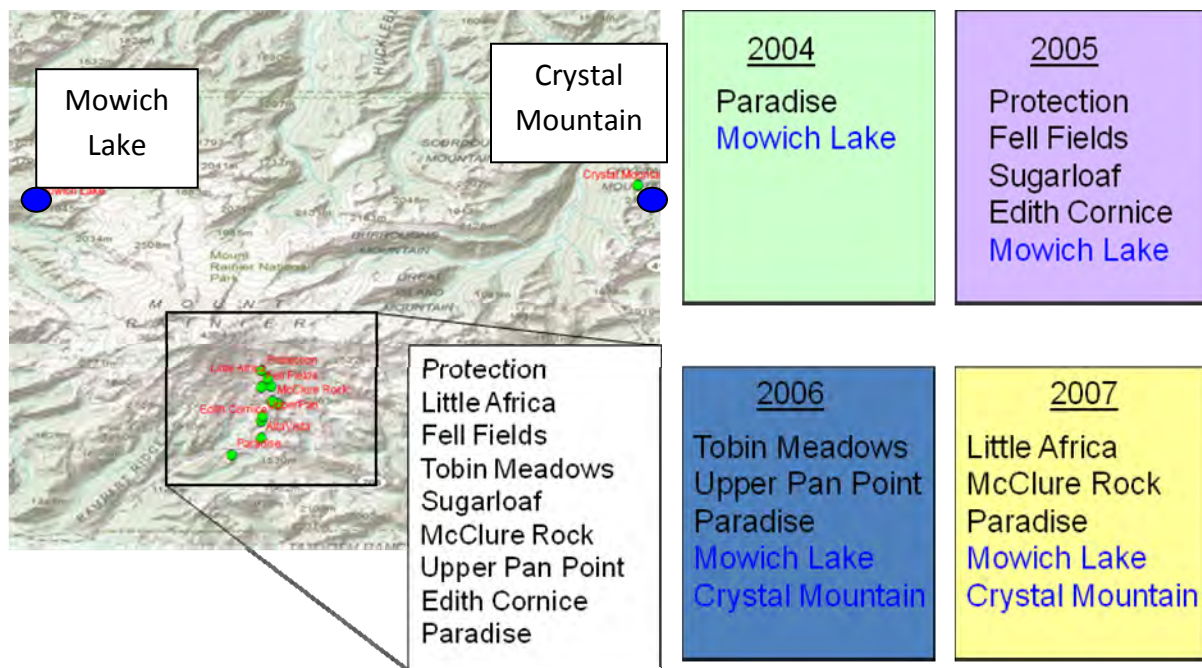


Figure 3: Snowpack sampling sites at Mount Rainier NP, and sites sampled in each year. The primary WACAP sampling sites, Golden Lake and PJ Lake, are located near the western boundary of MORA, northwest and southwest of Mount Rainier, respectively.

Sample collection methods

Collecting a single sample of the entire season's snowpack is a cost-effective means of sampling snow deposition at higher elevation sites where the snowpack temperature remains below freezing all winter. However, at sites below approximately 1500m elevation, mid-winter rain-on-snow and melt events can cause ablation of the snowpack and disproportionate loss of contaminants, rendering the seasonal snowpack a poor medium for sampling seasonal deposition. In order to ensure accurate measurement of contaminant deposition, both seasonal snowpack sampling and bulk precipitation collection were used in Olympic NP, where access to high-elevation sites is difficult.

Seasonal snowpack sampling was conducted in March and April, near the time of annual maximum snow accumulation but before the onset of spring melt. Snowpits were excavated to the ground and a vertical column of snow was cut from the pit face. For each sample, ~50 kg of snow were collected in six solvent-rinsed 60 × 60-cm polytetrafluoroethylene (PTFE) bags using solvent-rinsed Lexan shovels. Filled PTFE bags were wrapped in low-density black polyethylene bags to minimize light exposure and high-density polypropylene bags for protection. Samples were shipped to the laboratory on dry ice and stored at -20 °C until analysis. Field replicates were collected from the same snowpit for approximately 10% of the samples. Field blanks were also collected for approximately 10% of the samples by pouring VOC/pesticide-grade water (EMD Chemicals, Gibbstown, NJ) over sampling shovels and collecting it in the PTFE bags. Thereafter, field blanks were treated identically to snowpack samples.

Bulk precipitation collectors measured total deposition equivalent to snowpack sampling, but without the risk of losing the sample during rain-on-snow or midwinter melt episodes. The disadvantages over snowpack sampling are logistical: equipment installation is required, and the site must be visited regularly during the accumulation period. Bulk precipitation collection has been used successfully in other studies of inorganic and organic contaminant deposition (Carrera et al., 2002). For this project, a large open container 0.2 to 0.5 m in diameter was secured with the opening approximately 1 m above the ground or snow surface. This was done by installing 3-4 pieces of rebar in the ground and adding additional sections of rebar as the snowpack accumulates. The container was lined with a Teflon bag (similar to snowpack sample vessels but larger) that collected rain, snow, and dry deposition for a period of 1-4 weeks, or until the bag was full. At that time the bag was sealed and shipped frozen to the laboratory at Oregon State University. The collection container was elevated to 1m above the new snow surface when necessary, and a clean bag was installed.

Analytical methods

The target analyte list was composed of 47 pesticide and pesticide degradation products and included amide, organochlorine, organochlorine sulfide, phosphorothioate, and thiocarbamates pesticides as well as triazines herbicides. A detailed description of the analytical method and the target analyte list is provided elsewhere (Landers et al., 2003). In brief, snow samples were removed from the freezer at the time of analysis and allowed to melt in the dark, without heat, for ~36 hrs in sealed PTFE bags. Once melted, a methanol solution containing 16 isotopically-labeled pesticides, for use as recovery

surrogates, was spiked into the sample. Target analytes were extracted from melted snow using solid-phase extraction disks that were prepared in-house by combining hydrophobic and hydrophilic 1-g divinylbenzene *Speedisks*TM from Mallinckrodt Baker (Phillipsburg, NJ). Efforts were not made to separately analyze dissolved-phase and sorbed-phase pesticides because their phase distribution in the snowpack is not maintained when the snow is melted. Thus, concentrations reported herein are for total pesticide concentrations in snow, i.e. the sum of pesticides in the dissolved and sorbed phases. Gel permeation and silica gel adsorption chromatography were performed to remove matrix interferants. The final extract was spiked with an ethyl acetate solution containing four isotopically-labeled internal standards. Analyte separation, detection, and identification were performed on Agilent (Palo Alto, CA) 6890N gas chromatographs equipped with Agilent DB-5MS 30 m × 0.25 mm × 0.25 µm columns and 5943N mass selective detectors. Approximately one-half of the target analytes were quantified using electron impact ionization while the other half were quantified using electron capture negative ionization.

Target analyte loss was corrected for surrogate recovery by using target analyte-to-surrogate response ratios in calibration curves. The mean recovery over the entire analytical method for all surrogates in all samples was 55%, which is similar to that reported previously for this method (18). A laboratory blank was included with every set of two snowpack samples. Laboratory blanks were generated by spiking designated extraction disks with surrogate solution and then following all elution and clean-up steps. Snowpack concentrations reported herein were calculated by subtracting the mass of target analyte in a laboratory blank from that in corresponding snowpack samples. On average, analyte mass in laboratory blanks was 3% of that in corresponding snowpack samples for the concentrations reported in SI Table 2. Concentrations are not reported for cases in which the mass in the laboratory blank exceeded 33% of that in the sample; those cases are designated in SI Table 2. On average, concentrations in field blanks were 15% of those in corresponding snowpack samples. In correlation analyses, non-detects were replaced by concentrations representing one-half of estimated method detection limits. Estimated method detection limits for specific pesticides are provided elsewhere; values ranged from 0.20 to 120 pg/L for the 47 target pesticides.

Financial accounting:

Budgets shown for FY04-07 were for expenditures from this projects funding only and do not include funding from the WACAP project.

Cost sharing:

This effort was integrated with the ARD WACAP study. By integrating the snow sampling of this proposal with the larger WACAP study, much of the sample handling and interpretation costs was shared. Furthermore, the cost of developing and testing appropriate sampling methods was borne by the WACAP study during the 2002 pilot phase. The total cost sharing for this project was over 80%. Sources of this cost sharing were:

- (1) The WACAP study for methods development (\$50,000), analytical equipment (\$105,000), field sample design (\$15,000), QA/QC procedures (\$9,000), data analysis (\$4,500), database design and establishment (\$9,000) and final reporting (\$4,500).
- (2) EPA staff time for permitting and coordination with WACAP contributed by Dr. Dixon Landers and Marilyn Erway (\$20,000).
- (3) NPS staff time for permits, coordination, and field collection (\$7,000 at MORA, \$3,000 at NOCA) contributed by ONPS and Vital Signs Monitoring.
- (4) Matching funds provided by USGS (\$15,078).

Budget

NPS PNW SNOW PROJECT

Institution	Category/Activities	Year 1('04)	Year 2('05)	Year 3('06)	Year 4('07)	Total
OSU	Simonich					
	SOC Sample Analysis and Interpretation					
	24 x \$750	\$1,500	\$20,700	\$21,150	\$24,000	\$67,350
	Chiral Analysis and Interp		\$4,200	\$11,000	\$15,000	\$30,200
USGS	Planning, field collection, data mgmt, Interp	\$5,500	\$30,100	\$31,350	\$43,915	\$110,865
NPS	Field collection- personnel and supplies	\$1,500	\$6,000	\$6,500	\$7,000	\$21,000
NPS	Field collection- aircraft	\$1,500	\$10,000	\$11,000	\$12,000	\$34,500
UW-B	*Jaffee					
	Modeling Back Trajectories		\$5,000	\$8,000	\$11,000	\$24,000
Working budget, NPS funding		\$10,000	\$76,000	\$89,000	\$112,915	\$287,915
USGS Matching Funds		\$748	\$4,094	\$4,264	\$5,972	\$15,078
Total funds avail. For USGS		\$6,248	\$34,194	\$35,614	\$49,887	\$125,943

Results and Discussion

Differences in snowpack contaminant fluxes between the PNW parks, and comparison to other WACAP parks.

Snowpack contaminant fluxes in this study were similar to those reported in the WACAP study (Figure 4). In general, the PNW parks had relatively low contaminant loading relative to parks in California, Colorado, and Montana; but somewhat higher loadings than those in Alaska, which was related to proximity of local and regional sources of contaminants from agriculture, industry, and urban areas. (Hageman et al., 2006; Hageman et al., 2010; Landers 2010).

Variance of flux values in this study supported the analysis of variance for the larger WACAP study (Hageman et al., 2010). Variability was relatively low for site replicates (replicate samples collected at the same location and time) indicating that differences between samples collected from different locations and time periods are representative of real spatial and temporal differences in snowpack contaminant fluxes. The exception was Olympic National Park, where mid-winter rain-on-snow and melt events may have confounded results.

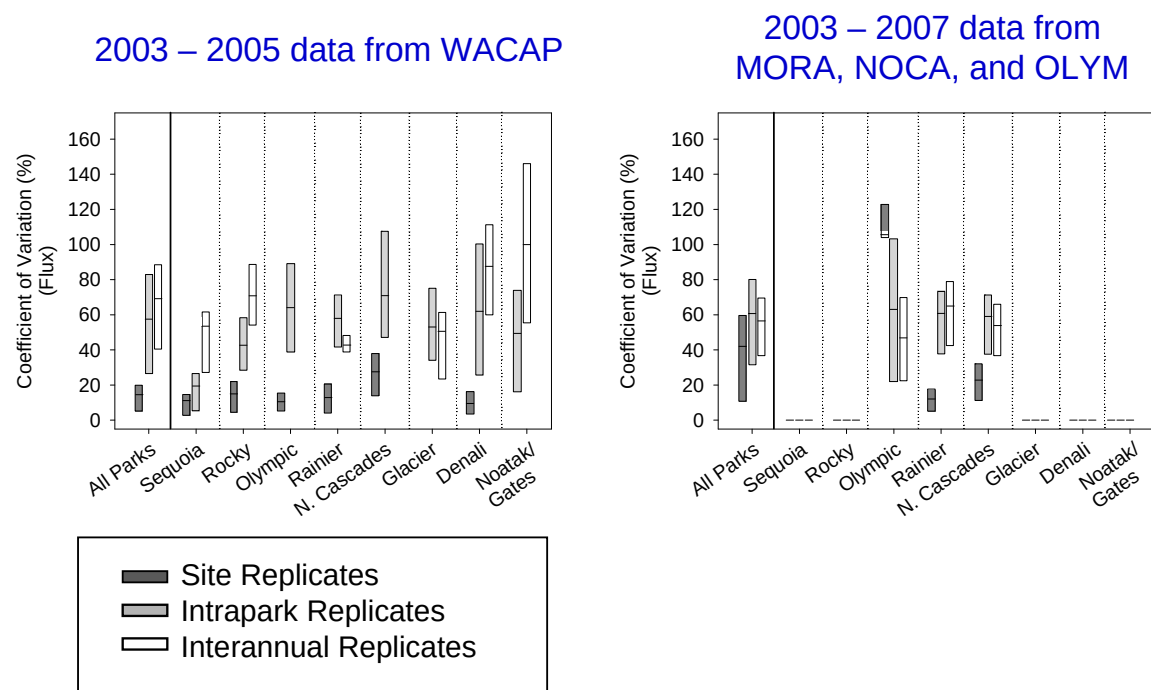


Figure 4: Comparison of variance in snowpack contaminant fluxes from the WACAP study (on the left) to results for PNW parks in this study (on the right).

Spatial gradients of snowpack contaminants in Mount Rainier and North Cascades NPs

Deposition of pollutants at sites on opposite sides (West-East and North-South) of North Cascades and Mount Rainier NPs were compared. Agricultural activity is most intense on the eastern sides of both parks, whereas urban and industrial activity is more intense to the west. However, no significant differences in fluxes of agricultural or urban contaminants were observed between sites on opposite

sides (West-East or North-South) of the parks. The absence of a statistically significant relationship may have been due to the natural variability in contaminant fluxes, and the relatively small number of samples available for the spatial analysis (generally 1-2 samples per year from each site, and just a few sites for each comparison).

Elevational gradient of snowpack contaminants in Mount Rainier NP

Contaminant fluxes were generally greatest at the lower elevation (1500-2000 m) snowpack sampling sites in Mount Rainier NP (Figure 5-7). Precipitation amounts are large at these sites, and meteorological conditions may focus transport and deposition of contaminants in this elevation range. An unusually sparse coverage of snow and shallow snowpack led to smaller fluxes in 2005 and likely confounded the relation with elevation that year.

In 2006 and 2007, the highest snowpack fluxes of current-use pesticides were measured at elevations less than 2000 m (Figure 5). A significant negative correlation with elevation was found in 2007 for dacthal, but not for the other current-use pesticides.

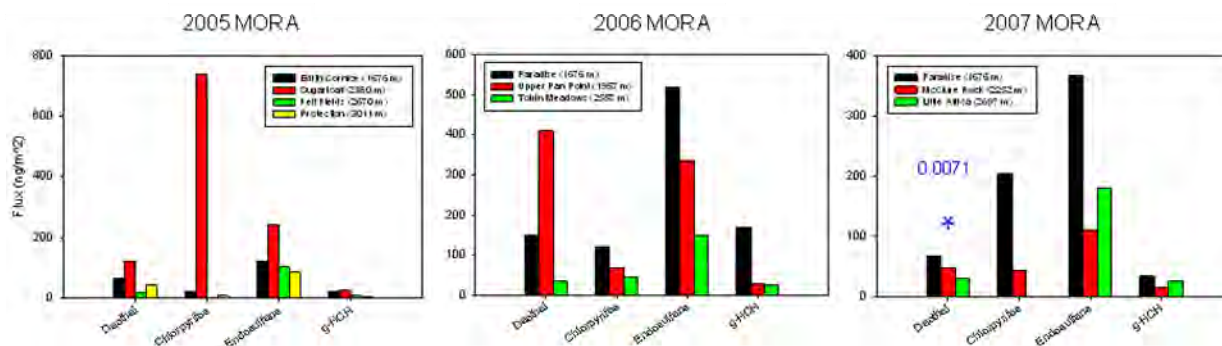


Figure 5: Fluxes of current-use pesticides along an elevational gradient on the south side of Mount Rainier. The asterisk indicates a statistically significant correlation for the concentration of the pesticide with elevation. The p-value is listed above the asterisk.

As with the current-use pesticides, there was a general pattern of historic-use pesticide fluxes decreasing with elevation, especially in 2005 and 2006 (Figure 6). The relation was statistically significant for a-HCH in 2005, and chlordanes in 2006. There was no relation between historic-use pesticide fluxes and elevation in 2007.

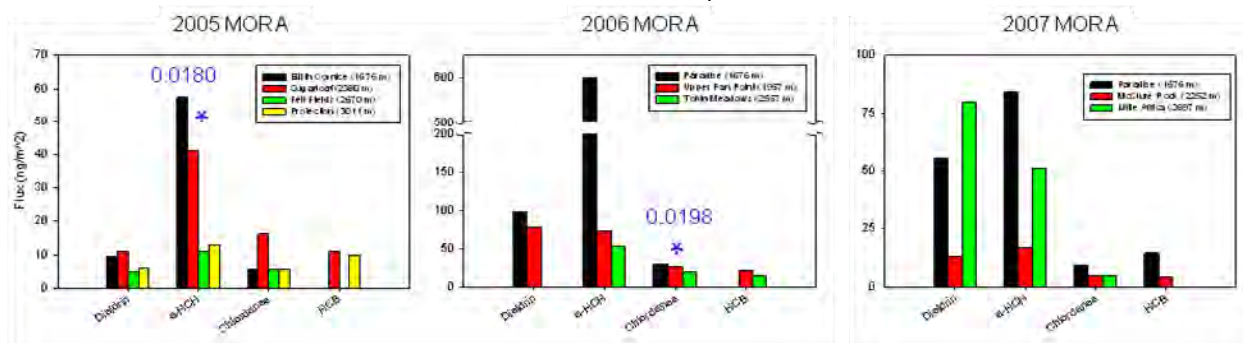


Figure 6: Fluxes of historic-use pesticides along an elevational gradient on the south side of Mount Rainier. The asterisk indicates a statistically significant correlation for the concentration of the pesticide with elevation. The p-value is listed above the asterisk.

In 2006 and 2007, polycyclic aromatic hydrocarbons (PAHs) showed a trend of decreasing snowpack fluxes at higher elevations (Figure 7). Significant negative correlations for Chrysene/Triphenylene and Benzo[k]fluoranthene are noted on the figure.

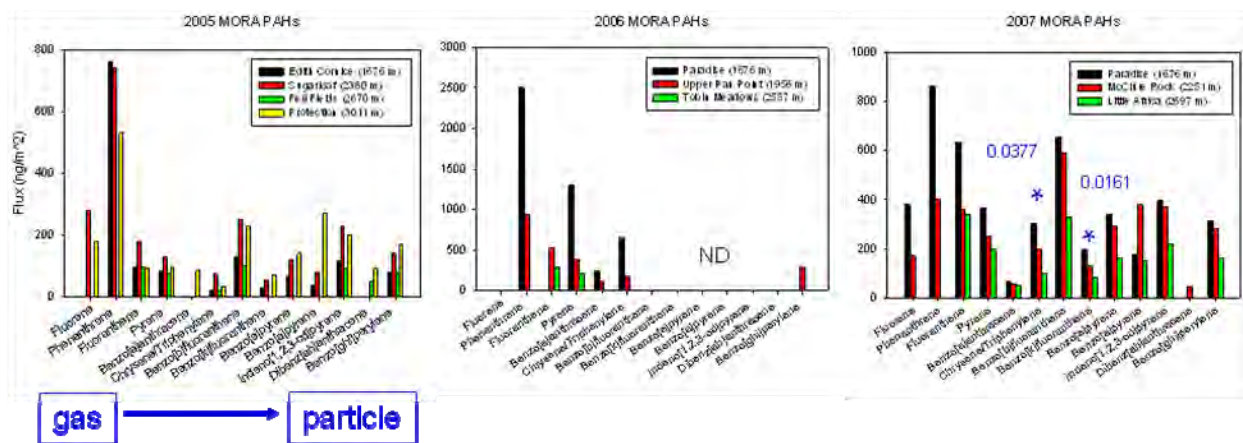


Figure 7: Fluxes of polycyclic aromatic hydrocarbons (PAHs) along an elevational gradient on the south side of Mount Rainier. PAHs are ordered from those typically found in the gas-phase on the left-side of the graph to those in the particle-phase on the right-side. The asterisk indicates a statistically significant correlation for the concentration of the pesticide with elevation. The p-value is listed above the asterisk.

Seasonal variability of contaminants in bulk precipitation in Olympic NP

Concentrations of current-use pesticides, particularly dacthal, chlorpyrifos, and endosulfans, generally increased in late winter and spring in WY 2005 and in WY 2006 (Figure 8). This might be linked to the time of year when CUPs are applied in local or regional agricultural areas, or it might be caused by warmer temperatures that mobilize semi-volatile compounds out of soils and vegetation at lower

elevations, making them available for re-deposition at higher elevation sites. In WY 2007, there were no samples collected in late winter or spring, so the apparent trend of decreasing concentrations in endosulfans and g-HCH represents the period from late autumn through mid-winter.

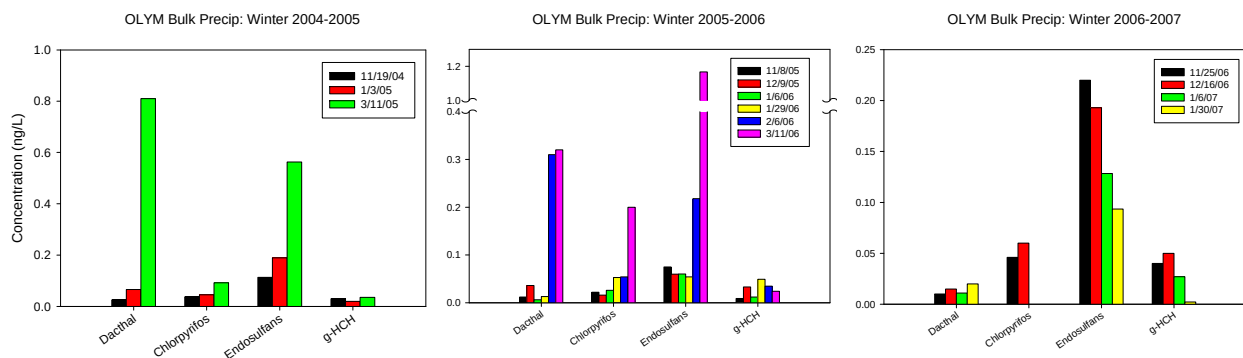


Figure 8: Concentrations of current-use pesticides in bulk precipitation at Olympic NP.

There were no clear seasonal patterns in concentrations of historic-use pesticides in Olympic NP (Figure 9).

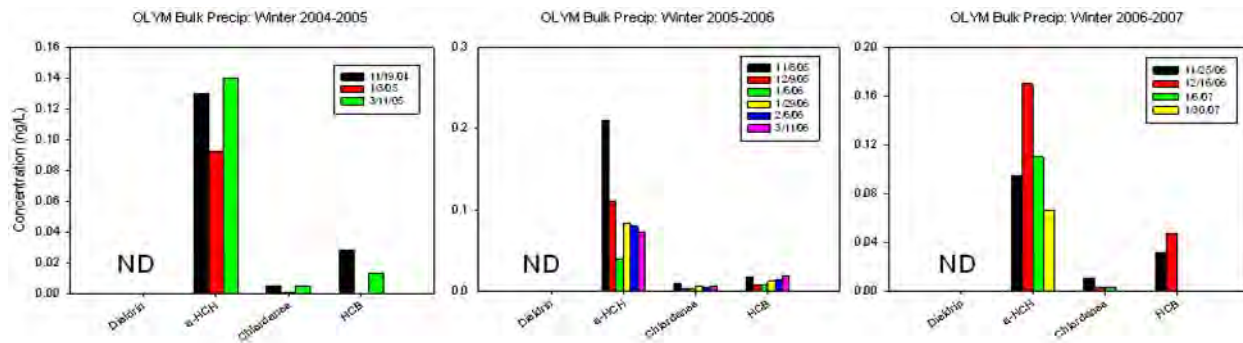


Figure 9: Concentrations of historic-use pesticides in bulk precipitation in Olympic NP. Pesticides that were not detected are labeled as “ND” on the graph.

There were no clear seasonal patterns of PAH concentration in bulk precipitation (Figure 10), because low PAH concentrations and small sample volume limited the number of values reported. However there was some indication that particle-phase PAHs decreased in the spring, based on a single year of data (WY2007).

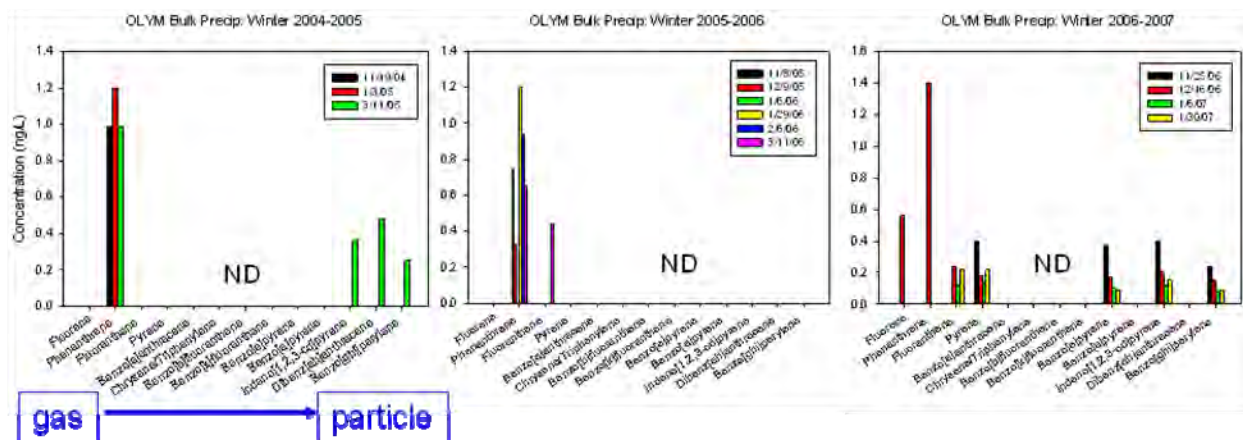


Figure 10: Concentrations of PAHs in bulk precipitation at Olympic NP. PAHs are ordered from those typically found in the gas-phase on the left-side of the graph to those in the particle-phase on the right-side. PAHs not detected are labeled as “ND” on the graph.

Conclusions

Seasonal snowpack sampling was a cost-effective means of measuring concentrations of contaminants in bulk deposition during the cold months in the PNW parks, however at lower-elevation sites, especially those in Olympic NP, unusually warm conditions and rain-on-snow events rendered the snowpack somewhat unreliable as a medium for preserving the seasonal deposition. Earlier snowmelt and a shift in winter precipitation type from snow to rain has been a documented effect of climate change throughout North America. In this study, we demonstrated that monthly bulk precipitation collection can serve as a substitute for the single seasonal snowpack sampling.

Elevational gradients in the fluxes of many contaminants were found on the south side of Mount Rainier. Highest fluxes of both current-use pesticides (CUPS) and historic-use pesticides (HUPS) were found at lower elevations of the transect (1500-2000m), with fluxes decreasing at higher elevations. Local meteorological conditions appear to favor both higher precipitation amounts, and higher concentrations of contaminants in deposition at these elevations.

Samples of bulk deposition collected approximately in Olympic NP indicated that concentrations of pesticides were higher during late winter and spring relative to late fall and early winter months. The higher concentrations may be the result of local or regional application of pesticides during the spring, or may be related to revolatilization of previously deposited pesticides during the warmer months.

Previously published analyses of the WACAP snowpack deposition data indicated that the intensity of regional agricultural activity affected the variability of contaminant deposition between the WACAP Parks (Hageman et al, 2006; Hageman et al., 2007). In this study, within North Cascades and Mount

Rainier NPs, no strong differences in fluxes were observed between sites on opposite sides (West-East or North-South) of either of the parks. Thus there was no evidence for strong local sources of contaminants that would tend to impact nearby areas of the Park more than sites located farther away. However, this does not rule out the possibility that local sources exist but their impact was not detectable given the other factors that affect deposition to the snowpack. In addition, snowpack sampling does not capture deposition that occurs during the warmer months, when many agricultural chemicals are applied, so year-round sampling of deposition might be needed to gain a better understanding of local agricultural sources.

Acknowledgments

We thank Dixon Landers for his leadership of the Western Airborne Contaminants Assessment Project and for writing the proposal for this project. This work would not have been possible without field, laboratory, and administrative support from Rebecca Lofgren, Stefan Lofgren, Glenn Kessler, Barbara Samora, Bill Baccus, Mike Larrabee, Jon Riedel, George Ingersoll, Alisa Mast, Marilyn Erway, and a host of others at Mt. Rainier, Olympic, and North Cascades NPs. All of them also contributed to the understanding of pollutant deposition in the Pacific Northwest Parks.

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