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The Western Airborne Contaminant Assessment Project (WACAP): An Interdisciplinary Evaluation of the Impacts of Airborne Contaminants in Western U.S. National Parks

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How much are contaminants impacting wildlife in Western U.S. National Parks?

EPA

The National Park Service Organic Act of 1916 (*1*) required protection of the national parks for perpetuity by tasking the National Park Service (NPS) to maintain these lands "...unimpaired for the enjoyment of future generations." Near the close of the last century, the NPS became aware of a new body of research describing a potential ecosystem threat that could not be ignored. 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, and the risks to the national parks of the western U.S. were unknown. Airborne contaminants present a broad range of potential risks to these ecosystems, largely due to bioaccumulation and or biomagnification of toxicants in biota, particularly vertebrates, that can result in loss of fecundity, unfit offspring, maladaptive behavior, and even death. As an outgrowth of these concerns, the Western Airborne Contaminants Assessment Project (WACAP) was initiated in 2002 to determine the risk from airborne contaminants to ecosystems and food webs in national parks of the U.S. The specific objectives that guided design and implementation of WACAP were the following:

1. Determine if contaminants were present in western national parks.

2. If contaminants were present, determine in what way and where they were accumulating (geographically and by elevation).

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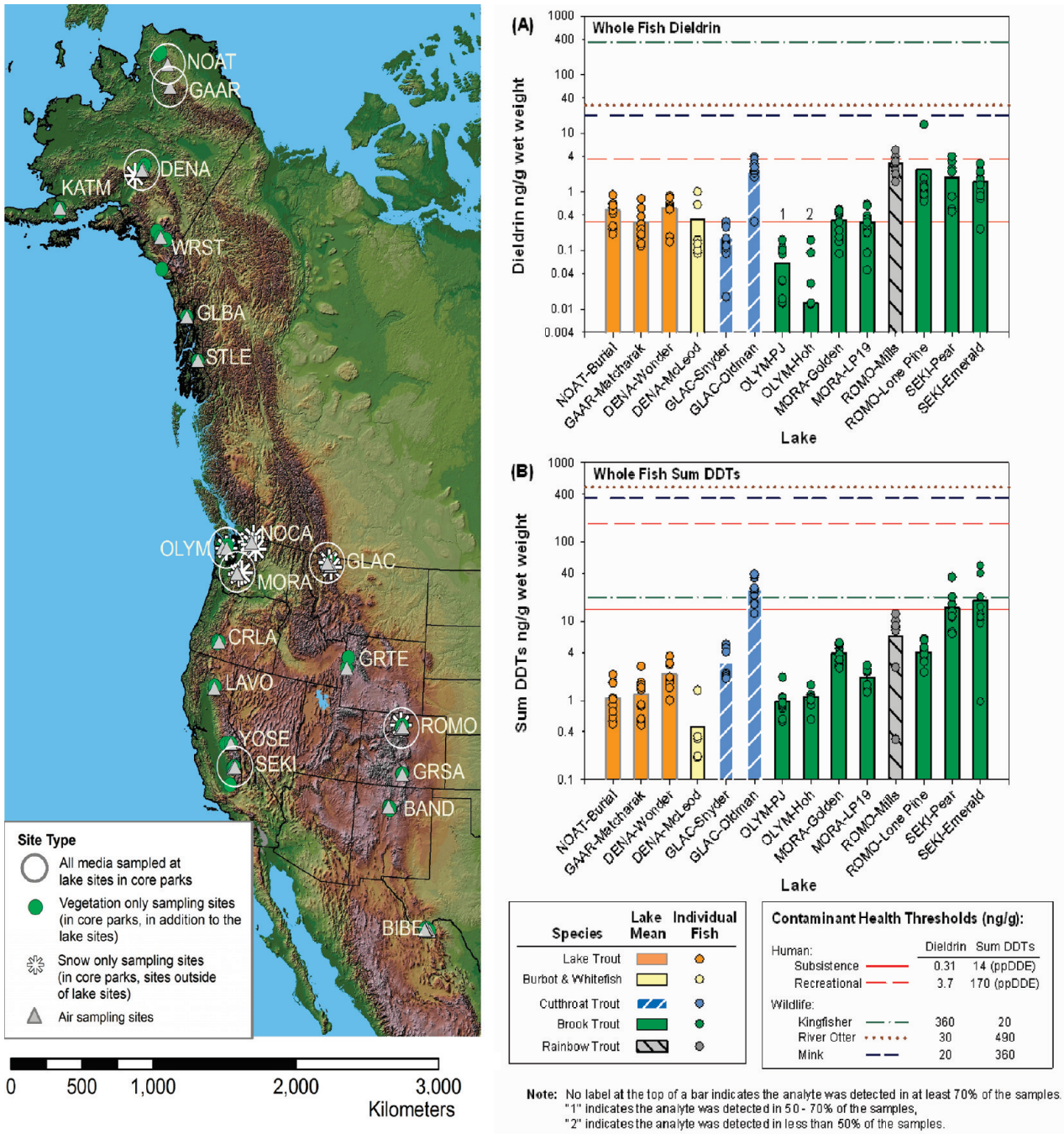

FIGURE 1. Relief map of western North America showing the WACAP core study sites in the western National Parks of the U.S. Graphs show individual and mean whole fish tissue concentrations for (A) dieldren and (B) sum DDTs for each lake sampled. Contaminant health thresholds for humans and wildlife appear as horizontal lines on the graphs. Similar information is available for other semivolatile organic compounds and metals in the WACAP report and database. See SI-E for more details.

3. If contaminants were present, determine which ones posed an ecological threat.

4. Determine which indicators appeared to be the most useful for assessing contamination.

5. If contaminants were present, determine the source of the air masses most likely to have transported contaminants to the national park sites.

From 2002 through 2007, an integrated, multidisciplinary team of WACAP researchers analyzed the concentrations and biological effects of airborne contaminants in air, snow, water, sediments, lichens, conifer needles, and fish in watersheds in each of eight parks in the western U.S., including Alaska (Figure 1 and section A of the Supporting Information [SI-A]). The U.S. Environmental Protection Agency (EPA), U.S. Geological Survey (USGS), U.S. Forest Service (USFS), Oregon State University, and the University of Washington worked in partnership with the NPS in an effort to achieve these objectives.

We evaluated contaminant concentrations in samples from multiple characteristic ecosystem components to evaluate patterns and variability in temporal integration of airborne contaminants (e.g., snow represented seasonal accumulations while lake sediments represented decennial time scales). We also used these samples to help determine the origin of airborne contaminants and to infer whether sources were local, regional, or global. The rationale for selecting these specific sample matrices is provided in SI-B. In addition to the eight core parks, WACAP identified 12 secondary parks (or monuments, preserves, or national forests) for expanded spatial and environ-

mental assessment (Figure 1 and SI-C). In the secondary parks, three ecosystem components: air, lichens, and conifer needles, were sampled.

Semivolatile organic compounds (SOCs), including some persistent organic pollutants (POPs), and metals were a key focus of this study due to their likelihood to undergo atmospheric transport to remote ecosystems. Specific contaminants served as markers for different probable sources. The SOCs fell into four general classes: current-use pesticides (CUPs), North American historic-use pesticides (HUPs), industrial/urban use compounds (IUCs), and combustion byproduct. Of the metals analyzed, Hg was of particular concern due to its detrimental neurological effects on humans, fish, and other organisms.

The final report (*2*) interprets and summarizes the information developed by WACAP and relates it to the current scientific literature with over 300 cited references. It strives to enhance scientific understanding of the global fate, transport, and associated ecological impacts of airborne contaminants on sensitive ecosystems in western parks and help the NPS decide what actions are needed to further understand, mitigate, and communicate the potential effects of these contaminants. A comprehensive WACAP database, published in 2010, is available on the Web site containing the report (*2*) to make these data widely and permanently available. The objective of this article is to provide a brief guide to the most significant and interesting results derived from the WACAP and to relate some policy-relevant actions resulting from this work.

Contaminant Distribution and Source Attribution

We measured over 100 different SOCs spanning a wide range of volatility, water solubility, and hydrophobicity, as well as persistence in the environment (SI Table 1). The physicochemical properties of the SOCs were used to help interpret the atmospheric transport, deposition, and accumulation of these compounds to and by WACAP ecosystems. Due to the remote locations of the WACAP sites, atmospheric transport modeling was integral to understanding air source regions important to our sites. We modeled atmospheric transport via back-trajectory cluster analysis on three different time scales for each of the WACAP core parks (*2*).

Spatial patterns of the contaminants found in greatest general abundance within various environmental compartments are as follows. Total pesticide concentrations in snow were highest in the Rocky Mountain National Parks (Glacier [GLAC] and Rocky Mountain [ROMO]) and Sierra Nevada (Sequoia and Kings Canyon National Park [SEKI] (*3*)). This general pattern was often repeated for environmental compartments besides snow. The highest pesticide concentrations in vegetation were also measured in California (SEKI and Yosemite National Park) and Rocky Mountain parks (GLAC and Great Sand Dunes National Park and Preserve). At parks with the highest pesticide concentrations in vegetation, the total SOC concentration was dominated by the CUPs endosulfan and dacthal. In lichen, the concentrations of many pesticides and polychlorinated biphenyls (PCBs) increased with elevation at most parks, suggesting that these compounds are undergoing cold fractionation or orthographic cold trapping (*4*). Concentrations of polycyclic aromatic hydrocarbons (PAHs) generally decreased with increasing elevation at most parks, suggesting that these less volatile incomplete combustion byproducts do not undergo cold fractionation and are sometimes found in higher concentrations at lower elevation due to wildfires and localized human activities. Nitrogen concentrations in lichens from SEKI, GLAC, Bandelier National Monument, and Big Bend National Park exceeded 90% quantiles for historic data from western public lands, indicating enhanced nitrogen deposition in these parks. Lichen data indicated enhanced sulfur deposition at SEKI and GLAC.

Fish whole-body lake mean and individual fish dieldrin and sum DDTs (dichlorodiphenyltrichloroethane (DDT), dichlorodiphenyldichloroethane (DDD), and dichlorodiphenyldichloroethylene (DDE)) concentrations are shown in Figure 1, along with contaminant health thresholds for humans and piscivorous wildlife. This type of information is available for all primary WACAP sites (*2*). Concentrations of dieldrin in fish, notably at ROMO, SEKI, and GLAC, were significantly elevated compared to those in fish from similar ecosystems in Canada (*5*). WACAP DDT concentrations in fish from SEKI, GLAC, and ROMO were higher than those reported for many fish collections analyzed from elsewhere in the world, including some fish from sites in Africa where DDT is used for mosquito control. Comparing contaminant concentrations in WACAP fish to those from similar European alpine environments, the flame retardant compound polybrominated diphenylether (PBDE) was ~3 times higher while HUPs were 2−9 times lower (*5*). All WACAP fish had lower PCB, hexachlorocyclohexane (HCH), and hexachlorocyclobenzene (HCB) concentrations than literature values from some recent surveys of atmospheric contaminants conducted elsewhere (references in (*2*)).

In general, Hg concentrations in trout from the WACAP parks in this study were lower than those reported for trout in lakes in the Midwest and Northeast U.S. However, Hg concentrations were higher in WACAP fish than in some species of fish from northern lakes in Canada and from mountain and subarctic ecosystems in Europe (see Table 5-1 in (*2*)). It is important to note that many contaminants we sought to measure were either not present or present in relatively low concentrations. This information is useful in identifying the contaminants of most current concern and certainly to make future comparisons of concentrations for the same sample types.

At WACAP parks in the conterminous 48 states, strong correlations were found between CUP concentrations in snow and vegetation (*2, 3*) and percent cropland within 150 km of the park. We attributed pesticide deposition in the Alaska parks to long-range atmospheric transport because there are no significant pesticide sources in close proximity (<150 km) to the sample sites. When present, concentrations of HUPs in parks in the lower 48 states were significantly higher than those found in Alaska, suggesting that regional or local revolatilization of the generally persistent HUP compounds in the lower 48 is still occurring after domestic usage bans.

In GLAC, PAH concentrations in snow, sediment, and vegetation in the Snyder Lake watershed were higher than those in the Oldman Lake watershed and 10−100 times higher than those in all other WACAP parks. Several lines of evidence suggest that the Al smelter in Columbia Falls, Montana, is the most likely source of elevated PAHs. The EPA confirmed that the smelter emitted PAHs at a rate of ~14 t/y from 1999 to 2005 and WACAP showed that comparisons of pre- and post-smelter PAH ratios in sediments suggest this smelter is the primary current source for these PAHs (*2*). At ROMO, higher SOC and Hg deposition in snow and sediment cores was found in the Mills Lake watershed (on the east side of the Continental Divide), than in Lone Pine Lake (on the west side), suggesting that the Continental Divide serves as a topographic barrier for transport of SOCs and Hg from agricultural and populated areas on the east side of ROMO to the west side (*6*). SOC concentrations in air as measured by passive air sampling devices (PASDs), conifer needles, and fish showed no clear differences between concentrations on the east side vs the west side of the Continental Divide.

Sediment cores provide a historical record of contaminant deposition over the past ~150 y. Deposition of anthropogenic Hg, indicated by temporal patterns in sediment cores, increased in nearly all parks during the 20th century. In SEKI, since about 1975, and in ROMO, since about 1990, Hg

deposition fluxes to all lakes have declined. However, in all the other lakes, the Hg sediment flux appears to be generally increasing or showing no change. This finding reflects a complex array of factors consistent with decreasing regional contaminant sources, increasing global contributions, and influences of individual watersheds on sediment Hg loading (*2*). Pb, Cd, and spheroidal carbonaceous particles (SCP) in sediment indicate deposition due to high-temperature combustion of fossil fuels (*7*). SCP increased in lake sediments in the conterminous 48 states during the 20th century, peaking around the 1960s. However, in recent decades, sediment Pb, Cd, and SCP have declined substantially, reflecting source reductions related to the Clean Air Act and regulation of Pb in gasoline. In Alaska lakes, SCP in sediment were nondetectable, indicating minimal regional sources of coal combustion. In these sites, Pb and Cd showed no evidence of a 20th century increase. Since there is minimal evidence for regional high-temperature combustion sources, the increase in Hg flux over time in the Alaska lake sediments is evidence of contributions from global anthropogenic sources (see SI-D for more on SCP).

Biological and Ecological Effects

WACAP assessed the impacts of contaminants on biota and ecosystems in a variety of ways, with the following key results. We observed biomagnification in all parks. Concentrations of SOCs were 5−7 orders of magnitude higher in fish than in snow, water, and air. Concentrations of SOCs were 2−3 orders of magnitude higher in fish tissue and vegetation compared to sediments. Vegetation tended to accumulate more PAHs, CUPs, and HCHs, whereas fish accumulated more PCBs, chlordanes, DDTs, and dieldrin. We expected SOCs in vegetation and air (PASD monitors) to have similar results; however this was not the case, likely because each medium absorbs different types of SOCs with varying affinities.

We assessed both fish-condition biomarkers and concentrations of contaminants in fish tissue (family, *Salmonidae*). All fish appeared normal during field necropsies. Cellular examination revealed that macrophage aggregates (MAs; an immune system response) generally increased with Hg concentrations and age in brook, rainbow, and cutthroat trout (*8, 9*). Vitellogenin (Vtg) concentrations in male fish are widely used as a biomarker for environmental estrogen exposure. Compounds such as dieldrin, DDT, PBDEs, PCBs, PAHs, endosulfan, and methoxychlor (among others) are known or suspected endocrine disrupting compounds (EDCs). Although sample sizes in this study were very small, significant correlations between contaminants (sum DDTs, sum PCBs, cis-nonachlor and endosulfan sulfate) and Vtg concentrations in male fish were found in the two lakes sampled in ROMO in 2003 (*2, 10*). Two male fish from GLAC and one male fish from MORA also had elevated Vtg concentrations. The fish from Oldman Lake (GLAC) with high Vtg concentrations also contained the highest concentrations of DDE of any of the fish sampled. These results support Vtg as a useful biomarker.

Intersex, the presence of both male and female reproductive structures in the same animal, is another commonly used biomarker of exposure to estrogen-mimicking chemicals. Six of 31 sites sampled during WACAP contained intersex fish. Intersex trout were found at Spirit, Dream, Haynach, Haiyaha, and Lone Pine Lakes, ROMO and at Oldman Lake, GLAC. Of the 24 sites we examined in pre-1935 museum collections (1800s−1934), we found intersex fish at only one site, Twin Lakes, CO (*10*). See SI-F for more information on intersex fish.

Hg concentrations increased with fish age in all fish species up to approximately 15 y of age. Hg concentrations in WACAP fish were highest in arctic Burial Lake (Noatak National Preserve), where mean Hg concentration exceeded the EPA contaminant health threshold for human consumption (*9*). The other arctic lake in this study, Matcharak (Gates of the Arctic National Park and Preserve), also contained fish with Hg concentrations exceeding human contaminant health thresholds. Because Hg concentrations in snow, sediment, and vegetation were low in these two parks, it is likely that factors such as Hg methylation rates, watershed biogeochemical characteristics, and food web efficiency in combination with fish age were the key contributors to the high observed Hg concentrations in fish (*2*).

Contaminant concentrations in 136 fish from 14 lakes in the 8 core WACAP parks were compared to the EPA's *Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories* (*11*). We increased the recreational fishing contaminant health threshold values for fish consumption by 32% to account for the estimated amount of chemical lost from fresh whole fish during filleting and cooking (*11*). See SI-G for additional information regarding the EPA guidance. Over half (77 of 136) of the individual fish from 11 of the 14 lakes analyzed had concentrations of dieldrin and/or p,p,-DDE that exceeded contaminant health thresholds for subsistence fishing (Figure 1).

The lake average fish dieldrin and/or p,p-DDE concentrations exceeded subsistence fishing thresholds in nine of the lakes and recreational fishing thresholds in none of the lakes, although five individual fish exceeded dieldrin thresholds for recreational fish consumption in Mills and Lone Pine lakes (ROMO), Pear Lake (SEKI), and Oldman Lake (GLAC). No other fish SOC concentrations exceeded human contaminant health thresholds. Of the other SOCs detected in >50% of fish (chlorpyrofos, dacthal, endosulfans, methoxychlor, mirex, HCB, a-HCH, g-HCH, chlordanes, heptachlor epoxide, and PBDEs), all were 1−7 orders of magnitude below human health thresholds.

We assessed impacts of contaminants on aquatic food webs by comparing fish contaminant concentrations to published thresholds protective of mink, river otter, and belted kingfishers (*12*). At numerous sites, mean Hg concentrations in WACAP fish exceeded thresholds—implying the possibility of detrimental health effects to these wildlife (*2*). Wildlife health thresholds for PCBs, banned from production and use in the U.S. in 1977, were not exceeded.

DDT production in the U.S. ceased in 1972. Some fish at the two sites in SEKI and the mean concentration of fish in Oldman Lake (GLAC), had concentrations of the sum of DDTs above the threshold for negative health effects for kingfishers (Figure 1B). A broad-spectrum pesticide widely used to control underground termites, chlordane was banned in the U.S. in 1988 as an EDC and a suspected carcinogen. Chlordane concentrations in WACAP fish were below wildlife thresholds, except for one fish in Oldman Lake (GLAC) that exceeded the threshold for kingfishers. The acutely toxic pesticide dieldrin was banned from agricultural use in the U.S. in 1974 and for most other uses in 1987. The highest dieldrin concentrations in WACAP fish were found in ROMO. Dieldrin was produced in nearby Denver, CO, from 1952 to 1982. Mean concentrations of dieldrin in fish at all WACAP sites were below the contaminant health thresholds for wildlife (Figure 1A).

WACAP Impacts

WACAP has received considerable recognition from the media with over 200 local, national, and international media outlets. At present count, 10 peer-reviewed papers have been published. In response to the WACAP results, the NPS convened multiagency workshops with WACAP PIs and local and regional policy makers to discuss implications of the WACAP findings and the actions that should follow. Efforts were made by several parks to issue communications targeted at users and employees of the National Parks and how these findings might affect their experiences and working environ-

ment, respectively. Warnings have been issued in several parks in regard to human fish consumption.

Source attribution regarding atmospheric contaminants was a motivating factor for initiating WACAP. The key issue was the relative importance of possible trans-pacific sources vs other local and regional sources. WACAP determined that deposition of CUPs and HUPs is most strongly influenced by proximity to agricultural and industrial areas rather than trans-Pacific sources. Alaska, having few regional sources and minimal agriculture, is mostly affected by transport from other source regions from outside the area.

One of the biggest concerns regarding the WACAP findings was how representative of other similar systems near WACAP sampling site were these results? There is no way of knowing this to certainty without additional data, but experience and other specific studies of less geographic scope suggest that the WACAP results are not anomalous. Several subsequent studies were implemented in specific response to the WACAP findings, particularly those associated with intersex fish and the endocrine disruption−contaminant linkage. These efforts are primarily intended to increase the sample size for the number of intersex fish and their contaminant burdens to render more definitive statistical analyses. WACAP results regarding the impacts of current use pesticides were provided by the NPS to the Stockholm Convention Treaty in the form of a briefing statement for EPA, NPS Office of International Affairs, and the U.S. State Department. WACAP results also were submitted to a Federal Register Notice requesting comment on a then proposed, now approved, endosulfan ban.

WACAP identified atmospherically deposited contaminants in multiple ecological samples representing different temporal integration periods from all WACAP sites. In some cases, the airborne contaminant concentrations in fish were found to exceed established human and wildlife thresholds. Sites were selected to provide "clean" and unambiguous signals of atmospherically deposited contaminants using ecological indicators but no claim is made that results from these sites are representative of the population of all lakes. Based on the interpretations of multiple lines of evidence provided by the WACAP data, it is clear that a variety of anthropogenic airborne contaminants are deposited in and, in some cases, impacting remote ecosystems in the western U.S.

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Supporting Information Available

More in-depth discussions of the results stemming from this study. This material is available free of charge via the Internet at http://pubs.acs.org.

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