

Organic Contaminants in Mountains

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The study of organic contaminants at high altitudes is motivated by the potential risk that they pose to humans living in, or depending on resources derived from, mountains, and to terrestrial and aquatic ecosystems in alpine areas. Mountains are also ideal settings to study contaminant transport and behavior along gradients of climate and surface cover. Information on organic contaminants in mountains is compiled from the literature and synthesized, with a focus on atmospheric transport and deposition, contaminant dynamics in alpine lakes and aquatic organisms, and concentration differences with altitude. Diurnal mountain winds, in connection with enhanced deposition at higher elevations caused by low temperatures and high precipitation rates, conspire to make mid-latitude mountains become convergence zones for selected persistent organic chemicals. In particular, the more volatile constituents of contaminant mixtures seem to become enriched, relative to the less volatile constituents at higher altitudes. For selected contaminants, concentration inversions (i.e., concentrations that increase with elevation) have been observed. A notable difference between cold trapping in high latitudes and high altitudes is the likely importance of precipitation. High rates of snow deposition in mid- and high-latitude mountains may lead to a large contaminant release during snowmelt. Regions above the tree line often have little capacity to retain the released contaminants, suggesting the potential for a highly dynamic contaminant fate situation during the snow-free season with significant revolatilization and runoff. The chemical and environmental factors that control the orographic cold trapping of organic contaminants should be examined further by measuring and comparatively interpreting concentration gradients along several mountain slopes with widely different characteristics. Future efforts should further focus on the bioaccumulation and potential effects of contaminants in the upper trophic levels of alpine food chains, on measuring more water-soluble, persistent organic contaminants, and on studying how climate change may affect contaminant dynamics in mountain settings.

Introduction and Motivation

The year 2002 was the United Nations International Year of Mountains, which culminated in the Global Mountain Summit and the Mountain Watch Report (1). This report, while identifying and evaluating many of the important

threats to mountain ecosystems, did not consider the issue of organic contaminants. Recently, there has been increased interest in identifying and quantifying organic contaminant levels in mountain regions. Semivolatile organic contaminants (SOCs), such as the polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), organophosphate pesticides (OPPs), and other organic substances have been measured at high altitudes in Europe and western North America, with a few studies being performed in the Himalayas, and a mountainous island in the subtropical Atlantic. In this comprehensive review, we seek to compile and synthesize all existing information on organic contaminants in mountains, paying special attention to atmospheric transport and deposition, alpine lakes, contaminants in mountain food chains, and concentration differences with altitude. We also identify knowledge gaps to suggest where further research should be directed.

Using the definition of mountains being areas of high altitude as well as areas of low altitude with large slopes, ~27% of the Earth's landmass is mountainous and 22% of the world's population lives within mountains (1). Although high mountain areas, because of their different locations across the globe, are very diverse, they also share many characteristics (2). Most have relatively low daytime air temperatures and relatively high precipitation amounts compared to lowlands, and most experience long-lasting snow cover. Wind speed and solar radiation flux are higher than in adjacent lowlands, whereas atmospheric pressure is lower. The interaction of elevation and topographical gradients results in a high degree of habitat diversity, or environmental patchiness. Many of these characteristics will influence the environmental behavior of organic chemicals, suggesting that contaminant fate is different in mountains than in neighboring lowlands. Of particular interest in this regard is the suggestion that the specific conditions of mountain areas may result in the amplification of certain contaminants, i.e., concentration levels that are elevated in high altitudes, relative to those in surrounding lowlands (3, 4).

There are primarily three reasons for studying organic contaminants in alpine ecosystems. The first relates to the potential impact on humans, either living within, or depending on resources derived from, high altitude regions. Mountain regions serve as a water supply, both directly and indirectly through provision of lowland surface water. Contamination of snow and lake water in high mountain regions thus has the potential to impact drinking and agricultural water supplies. Although the presence of organic contaminants in such water supplies is worrisome, it should be stressed that human exposure to sparingly soluble organic compounds through the intake of water is usually negligible, when compared to the exposure caused by the consumption of animal lipids. Many high mountains—in particular, the

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European Alps and the Himalayas—are important areas of dairy and meat production, and both sport fishermen and indigenous people consume fish from alpine areas.

The second incentive for studying organic contaminants in mountain regions is the potential impact on the alpine ecosystems themselves. High mountain areas often support unique plant and animal communities of great ecological value. Bioaccumulating compounds may pose a threat to the top predators in alpine ecosystems, in particular, birds of prey and carrion birds. Examples are eagles, ospreys, and vultures, but also lynx, bears, wolves, and cougars feed at the top of alpine food chains. Although efficient biomagnification of organic contaminants has been demonstrated in the Arctic lichen–caribou–wolf food chain (5), no such studies have been conducted in alpine food chains. Not only top predators may be at risk. Amphibian population declines have been linked to organic contaminants in the Californian Sierra Nevada (6). Higher lipid storage, longer lives, and slower growth of aquatic species in alpine lakes may exacerbate the bioaccumulation of hydrophobic contaminants. Organic contaminants may also affect high-altitude vegetation. In particular, nitrated phenols and haloacetic acids and their halogenated hydrocarbon precursors may act as phytotoxins and have been detected in needles from the European Alps (7–9).

Aside from evaluating the risks to humans and alpine environments, gaining an understanding of the fate and transport of organic compounds in mountains holds value in itself. Mountains are steep environmental gradients on a small scale. Studying how different chemicals behave and are distributed along altitudinal gradients can assist in understanding the influence of various environmental parameters (climate, vegetation, etc.) on chemical behavior. Contaminant transport and distribution processes in mountains may be indicative of mechanisms operating on a larger scale. Identifying concentration gradients along an altitudinal slope is logistically much simpler to accomplish than trying to identify differences between polar, temperate, and tropical systems. In addition, many alpine areas have not been directly impacted by human activities and thus are suitable to study effects of remote pollution sources.

Mountain Meteorology and Atmospheric Contaminant Transport

Intense industrial activity or agriculture in mountain areas is rare. Although major urban and industrial pollution sources exist at high altitudes, in particular at lower latitudes, these are often located on high-altitude plateaus and not in regions with alpine characteristics. Although smaller settlements, traffic arteries, tourism, and resource industries may often constitute local contaminant sources within mountains, most organic contaminants will reach alpine regions by atmospheric transport. In some cases areas of high population density, intense agriculture, and substantial industrial activity are in close vicinity to, or are surrounding, mountain regions. Examples are the European Alps, the southern slope of the Himalayas, and the Californian Sierra Nevada. Understanding the presence of organic contaminants in mountain regions thus requires an understanding of their atmospheric transport and deposition. It is not our objective to review the general principles and mechanisms of atmospheric transport and deposition of organic contaminants. Instead, building upon a brief review of mountain meteorology, we will focus on the specific circumstances that may impact on these processes within an alpine setting. Mountain climates differ from each other because of latitude, distance from the sea, and exposure to regional circulation; however, there are some common features.

Mountain Winds. There are two main types of wind associated with mountains (10). Terrain-forced flows are

produced when large-scale winds, stemming from the circulation around high- and low-pressure centers, are modified or channeled by underlying mountain ranges. This can cause airflow to be carried over or around a barrier, to be forced through gaps in a barrier, or to be blocked by it. Diurnal mountain winds, on the other hand, are produced by temperature contrasts that form within mountains or between mountains and the surrounding plains. Diurnal wind systems carry air into the mountains at low levels during the daytime and out of the mountains during the nighttime (Figure 1), and these systems may range over several hundred kilometers (11). Wind patterns, for example, influence the transport of organic contaminants in central California. Large-scale westerly winds bring polluted air from the coastal cities and the Central Valley to the Sierra Nevada. During the daytime, the sun heats the slopes of the foothills, causing the air mass over the slopes to rise and move up-slope from the valley. At night, the processes reverse and wind movement is down-slope.

Periodical wind systems in and out of mountain ranges may also operate on a seasonal scale. Two distinct tropical monsoon systems operate in the Indian Ocean region: the southwest or summer monsoon (June–September) and the northeast or winter monsoon (December–April). The seasonal change in wind direction is often combined with variable precipitation patterns, suggesting distinct seasonal differences in the direction and extent of atmospheric transport and deposition. The effect of the monsoon on air pollution in the Himalayas has been documented for aerosols and inorganic contaminants (12–15) and may also affect the transport of organochlorine pesticides (OCPs) from the Bay of Bengal to the Tibetan Plateau (16). Studies in the Arabian Sea indicated the potential of the monsoon to influence the transport of SOCs in this region (17).

Air–Surface Exchange along Elevation Gradients. Wind systems as described previously would simply advect contaminants back and forth between lowlands and higher elevations, unless degradation or deposition occurs. Whether contaminants are delivered to mountain ecosystems by such wind systems depend on the extent of air–surface exchange along the elevation gradient. Such exchange for SOCs is dependent on surface characteristics and climatic parameters, with deposition being favored by low temperatures, high precipitation rates, and high surface roughness, and evaporation being favored by high temperatures and low capacity of the surface for the contaminant. This suggests that many factors conspire to enhance deposition rates at high elevations. Forced rising motions cool the ascending air and enhance the formation of clouds and precipitation, whereas sinking motions warm the air and cause clouds to dissipate. Therefore, precipitation rates typically increase with elevation. During the daytime, when airflow is into the mountains at ground level, high temperatures should favor the evaporation of organic chemicals in lowlands. Chemicals are transported with the up-slope wind to higher elevations, where it is cooler, precipitation is more likely and deposition therefore is enhanced. During the nighttime, low temperatures prevent contaminant evaporation along the entire gradient. The down-slope wind system can thus be expected to carry less chemicals back into the valley than the daytime up-slope wind brought, resulting in many cases, in a steady transfer of chemical up-slope (Figure 1). Rapid degradation in the daytime atmosphere, by photolysis and reaction with photooxidants, may prevent such transfer.

Snow and Fog. Not only do mountains experience higher precipitation rates, the precipitation also occurs more frequently in forms that could enhance the deposition of selected organic pollutants. For example, snow, which often can be a more efficient scavenger than rain (18), accounts for 60%–70% of the annual precipitation in the Sierra Nevada

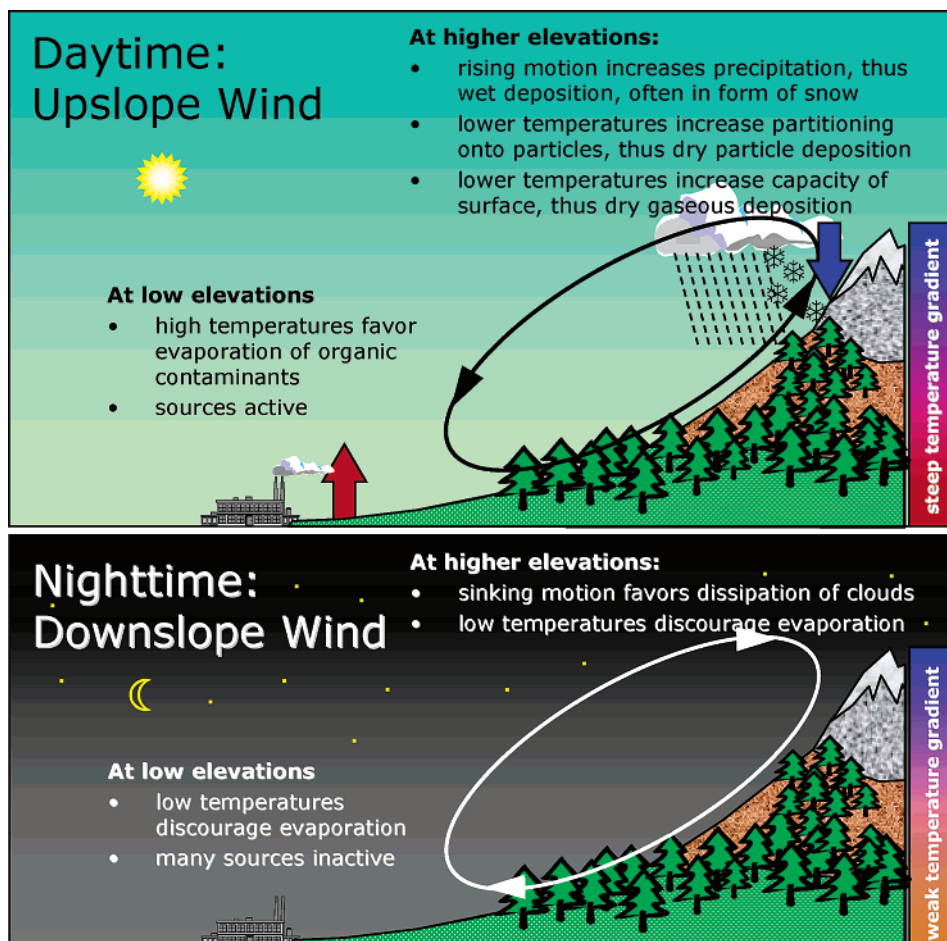


FIGURE 1. Illustration of the processes affecting organic contaminant distribution along a mountain slope. Daytime and nighttime situations differ in terms of the wind direction, the temperature gradient with elevation, and the activity of local sources.

and Rocky Mountains (19). Fogs are also common features of mountain regions. They are ground-based clouds and form when a decrease in temperature causes an increase in relative humidity beyond the dew point (10). Most relevant are up-slope fogs, whereby moist air is cooled by being lifted up slopes, and radiation fogs, which develop in valleys and mountain basins when outgoing long-wave radiation cools the near surface air during the nighttime and in the winter. The latter often have a distinct upper limit, defined by a temperature inversion caused by the same conditions (also see the next paragraph). Fog droplets are much smaller than rain droplets and have a much higher surface-to-volume ratio. This can lead to significant enrichment of surface-active organic chemicals in fogwater, when compared to the concentration expected from Henry's law (20–23), and suggests that fog may be an important mechanism for organic contaminant deposition in mountain regions. In particular, it could explain differences in contaminant concentrations within mountains, if the occurrence and frequency of fogs is localized (e.g., below temperature inversions, on slopes experiencing up-slope winds).

Temperature Inversions. Increases in temperature with elevation can also impact the behavior of organic contaminants. These temperature inversions are common in mountainous areas and affect atmospheric stability, vertical air exchange, and cloud formation. Surface-based inversions form at night and in the winter, when the ground loses heat by long-wave radiation. Inversion breakup occurs during the day when up-slope flows remove air from the valley (24). Inversions resist vertical motions so that contaminants emitted below it will not mix with the air above it. This

trapping effect can lead to the accumulation of contaminants during the inversion and generally limits their transport along elevation gradients. Also, the frequency and elevation of fogs and clouds will be strongly influenced by inversions and, thus, the wet deposition processes associated with them. In some cases, long-lasting inversions at higher altitudes are observed. For example, an elevated inversion at 1000 m is present year-round in the subtropical troposphere off the Atlantic coast of northwestern Africa (25), and the higher peaks of the Canary Islands reach beyond the height of this inversion. It separates air masses of lower altitude from the free troposphere and leads to the accumulation of clouds below the inversion. When interpreting concentration gradients in mountain areas, persistent or frequent temperature inversions must be taken into consideration, if the contaminant sources are located at low elevations.

Atmospheric Transport and Deposition in Mountain Regions

Several studies have addressed the question of the origin and delivery of organic contaminants in mountain regions. A large variety of environmental media, including air, rainwater, snow, glacier ice, foliage, and soil, have been sampled to address this issue. Measurements of organic contaminants in mountain air, summarized in Table 1, often sought to establish concentration gradients. OPPs and their oxon reaction products have been repeatedly quantified in air sampled at multiple elevations within the Californian Sierra Nevada (26–28). OPP air concentrations decreased rapidly with altitude, and many OPPs were undetectable at the highest sampling sites. Absolute levels of the oxons also

TABLE 1. Studies Reporting Organic Contaminant Concentrations in Mountain Air

chemical	location (elevation ^a)	date	ref
OPPs and Other Currently Used Pesticides			
chlorpyrifos, diazinon, parathion	3 elevations (114/200, 533, 1920 m) in and below Sequoia National Park, Sierra Nevada, CA	15 days (Nov. 1990–Feb. 1991)	26
chlorpyrifos, methidathion		8 days (May–Oct. 1994)	27
chlorpyrifos, diazinon, malathion, chlorothalonil, trifluralin, endosulfan		5 days (May–Sept. 1996)	28
SOCs			
PCBs in 2 HiVol samples	Lake Tahoe, Sierra Nevada, CA, USA (ca. 1900 m)	July 1995	29
OCPs in 7 HiVol samples	Bow Lake, Rocky Mtns., Canada (1940 m)	summer 1998	61
PAHs in 36 HiVol samples	Estany Redó, Pyrenees (2240 m)	1996–1997	34
	Gossenköllesee, Austrian Alps (2413 m)	1996–1997	
	Øvre Neådalsvatn, Norway (728 m)	1998	
PCBs and OCPs in 23 HiVol samples	Bow Lake (1974 m), Donald Station (770 m), Lower Kananaskis Lake (1667 m), Sundre, Rocky Mountains, Canada (1115 m)	June 1999–Aug. 2000	33
PCBs and OCPs in 20 HiVol samples	Pico de Teide, Canary Islands (47 and 2367 m)	June 1999–July 2000	25
α-HCH, HCB, DDT	Yahiko Mtn. (634 m), Japan	May–Nov. 2001	30
OCPs in 8 passive samples	Rocky Mountains, Canada (800–2300 m)	2000–2001 (annual average)	31, 32
PCBs and OCPs in 30 HiVol samples	Estany Redon, Central Pyrenees (2240 m)	Nov. 2000–Jan. 2003	106
	Skalnate Pleso, High Tatras (1778 m)		
OCPs in 6 HiVol samples	Dingri (4340 m), Rongbu Valley (5050 m), Tibetan Plateau	May–June 2002	16

^a Elevation given in meters above sea level.

decreased with elevation but increased in abundance relative to their parent compound. While most of the OPPs were undetected at high elevations, PCBs were present in air sampled near Lake Tahoe (29). Considering that PCBs have not been in use for many years, their presence at higher elevations reflects their greater persistence and ability to undergo long-range transport. DDT, α-HCH, and HCB have been detected in air sampled at higher elevations in Japan, with concentrations lower than those measured at two lowland sites (30). A greater proportion of the lighter SOC was observed at higher altitudes in both Japan and California (29, 30). Annual mean concentrations of α-HCH, γ-HCH, PeCB, HCB, and α-endosulfan, as determined with passive air samplers, were observed to increase significantly with altitude in the Canadian Rockies (31, 32), whereas another study in the same region concluded that summertime SOC air concentrations were not correlated with altitude (33). The more volatile tri- and tetrachlorinated congeners dominated the PCB profile, but no fractionation with altitude was observed (33). Very low concentrations of PCBs, HCB, HCHs, and DDTs have been detected at high elevations (2367 m) on Pico de Teide on Tenerife in the subtropical Atlantic (25). Air samples that were taken close to sea level showed much higher levels, which was explained by a persistent inversion that obstructs vertical air exchange. PAH air concentrations were similar across various European mountains and showed a seasonal variation that was dominated by greater emission during the winter (34).

Generally, it appears that air concentrations of organic contaminants tend to decrease with altitude, either as a result of efficient atmospheric degradation (e.g., OPPs in California), limited vertical air mass movement (e.g., SOC in Pico de Teide), or general dispersion and deposition of contaminants emitted at lower altitudes. However, several studies (29, 30) indicate a relatively higher atmospheric mobility of the more volatile SOC, and Shen et al. (31, 32) even observed a concentration increase of relatively volatile, yet persistent, SOC with altitude. Generally, gradient studies so far have relied on only a few sites at different elevations, presumably because most air sampling techniques require electricity and operating personnel, which often are not available at high elevations. Passive air sampling (31, 32) may prove useful in

obtaining information on SOC in mountain air and, in particular, may facilitate the derivation of more highly resolved elevation gradients.

Several attempts have been made to quantify the atmospheric deposition of organic contaminants in mountain areas by measuring concentrations in precipitation samples (Table 2). Following the patterns observed in air, currently used pesticides had a tendency to experience a decrease in deposition with altitude (26, 35). On the other hand, higher PAH concentrations were reported in snow along Bavarian mountain barriers, with lower concentrations on the lee side of the mountains, suggesting the influence of regional wind patterns (36, 37). The concentrations of several OCPs and the di- and trichlorobiphenyls in snow samples from the Canadian Rockies were positively correlated with elevation (38). The increase in PCB concentrations with altitude was greater for the lighter congeners than for the heavier, less volatile congeners. A similar increase in concentration with altitude has been found for HCHs and endosulfan in snow from the Sierra Nevada (39). PAHs in snow from the catchment area of five European mountain lakes were dominated by the low-molecular-weight compounds (40). In summary, evidence suggests that atmospheric SOC deposition is weighted toward the lighter, more volatile compounds at high altitudes. These substances may even occur at higher concentrations in high-altitude precipitation. Combined with precipitation rates that increase with elevation, higher snow concentrations could result in greatly elevated deposition rates at higher altitude (41).

Some of the snow falling at high altitudes will undergo firnification and eventually become incorporated into glaciers. This led to suggestions that glacier ice cores may be useful to provide an indication of the historical deposition of SOC to high-altitude areas. However, the extent to which the ice record really reflects atmospheric conditions in the past is subject to debate (42), and the relationship between current concentrations in the ice and past atmospheric concentrations is likely to be very complex. Upon melting, glaciers may also release contaminants deposited in the past (43). Confirming the trends observed in snow, ice samples from high altitudes (Table 2) tend to be dominated by the more volatile constituents of SOC mixtures. Seven relatively

TABLE 2. Studies Reporting Organic Contaminant Concentrations in Deposition Samples from Mountain Regions

chemical	location	elevation	ref
Dry Deposition			
OPPs, endosulfan	glass plates in Sequoia National Park, Sierra Nevada	200, 533, 1920 m	28
Wet Deposition			
SOCs	snowpack in Bavaria, Germany	up to 1000 m	36, 37
SOCs	rain at Field, Yoho National Park, Canada, and snowpack from various lakes in Canadian Rockies	1032–1940 m	87
OPPs	wet deposition in Sierra Nevada	114, 533, 1920 m	26
OPPs, SOCs	rain and snow from Sierra Nevada (Sequoia NP, Lake Tahoe)	533, 1920, 2200 m	35
OCPs, PCBs	snowpack from Rocky Mountains and Coast Range, Canada	770–3100 m	38, 43
PCBs	2 snowpack samples from Lake Tahoe basin, Sierra Nevada	1900 m	29
OCPs	snowpack samples from Sierra Nevada	ca. 2300, 3150 m	39
PCBs, OCPs, PAHs	snowpack samples from:		
	• Estany Redó, Pyrenees, Spain	2240 m	40
	• Jöri III, Swiss Alps	2519 m	
	• Gossenköllesee, Austrian Alps	2413 m	
	• Starolesnianske, Tatra, Poland	2000 m	
	• Øvre Neådalsvatn, Norway	728 m	
PCBs, OCPs	bulk atmospheric deposition at:		
	• Estany Redó, Pyrenees, Spain	2240 m	66
	• Gossenköllesee, Austrian Alps	2413 m	
	• Øvre Neådalsvatn, Norway	728 m	
SOCs	rain from Niigata, Japan	up to 634 m	30
SOCs	snowpack samples from parks across the western United States	up to 3567 m	107
aliphatics and phenols	snowpack samples from the Austrian Alps	3106 m	108
Glacier Ice			
PAHs, PCBs	Agassiz Ice Cap, Ellesmere Island, Canada	1600 m	44, 45
OCPs	Snow Dome, Columbia Icefield, Canadian Rocky Mountains	3300 m	109
biogenic and petroleum organics	Himalayas	7000 m	110
PCBs, OCPs	Lys Glacier, Italian Alps	4250 m	46, 111
OCPs	Mt. Mushitage, Tibetan Plateau	6450 m	16

TABLE 3. Studies Reporting Organic Contaminant Concentrations in Vegetation and Soil Samples from Mountain Regions

sample	location	elevation	chemicals	ref
pine needles	Sierra Nevada	114, 533, 1920 m	OPPs	27
foliage, soil	Alps	126–1800 m	PCBs	55
soil	Holy Cross Mountain, Poland	up to 612 m	PAHs and PCBs, OCPs, phenols	48
spruce needles, humus layer	Alps	840–1650 m	OCPs, PCBs, PAHs	51
spruce needles, humus layer	Alps	480–1600 m	OCPs, PCBs, PAHs, chloroform, TCA, nitrophenols	54
spruce needles, humus layer	Alps	480–1600 m	OCPs, PCBs	53
soil	Pico de Teide	10–3400 m	OCPs, PCBs	49
pine needles	Pico de Teide	0–2100 m	OCPs	50
conifer needles	Canadian Rockies	770–2200 m	OCPs, PCBs	33, 98
soils	Alps	1783–2240 m	PCBs, HCBs, DDTs	67
soil	Tibetan Plateau	not reported	HCHs, DDTs	80

small PAHs were identified in the Agassiz Ice Cap (44); however, those with five or more rings were not detected. PCBs in ice cores from the Agassiz Ice Cap and Lys Glacier were also dominated by lighter congeners (45, 46).

Measurements in Foliage and Soils. Because of the accumulation of SOCs in the waxy cuticula of needles and leaves, matrixes such as foliage, humus, and surface soils have been used repeatedly to discern trends in contaminant deposition with altitude (Table 3). Mirroring the gradients in air and precipitation, concentrations of OPPs and their oxons in pine needles decrease with increasing elevation in the Sierra Nevada (27). Altitudinal trends for SOCs are more complex. Concentrations of HCB in foliage sampled at high altitudes in the Tropics are surprisingly high when compared to very low levels measured at low altitudes, whereas DDT and HCHs do not show this behavior (47). Elevated concentrations at higher altitudes have also been found for several more volatile OCPs in conifer needles from the mountains of Western Canada, although concentrations were lower than in valleys closer to the Pacific (33). In a study of the regional spatial distribution of PCBs, PAHs, and OCPs in soil sampled around Kielce, Poland, the highest levels were observed at the highest sampling sites (48). Higher organic

carbon normalized concentrations of pentachlorobenzene, HCB, and several PCB congeners were also found in soil that was sampled at higher elevations (but not above the inversion layer) on Pico de Teide (49). However, another study conducted in Tenerife showed no significant altitudinal trend in the concentrations of HCB in pine needles (50). Geo-spatial orientation was a more important factor in the concentrations of DDT and metabolites than altitude, presumably because of the different historical use of DDT on the north and south sides of the island.

Weiss and colleagues extensively investigated the distribution of various SOCs in needles and humus layers of the Austrian and Slovenian Alps (8, 9, 51–54). A network of 25 stations across Austria indicated a generally higher pollutant load in the northern and eastern part of the Alps, which was attributed to closer proximity to the emission sources and a barrier effect that limited transport across the mountain range (52). Of particular interest are transects on forested slopes in Achenkirch, Tyrol (840–1650 m) (51, 52) and Koralpe, Carinthia (480–1600 m) (53, 54), with three and five sites at different elevations, respectively. In both cases, most of the quantified chemicals, which included PCBs, PCDD/Fs, and several OCPs, showed the highest concentrations in

the needles and humus from the highest sampling site. The change in the relative composition of PCDD/Fs and PAHs, and in the ratios DDT/DDE and α -HCH/ γ -HCH with altitude were often different between needles and humus, and also between the two transects (51, 54). Possible factors contributing to the shifts in SOC composition with elevation are differences in the origin of the contaminants (local at low altitudes versus long-range transport at higher altitudes) and possibly also in the deposition mechanism (higher particle bound fractions at colder, higher elevations). In contrast to the findings in Austria, PCB concentrations in soils decreased with elevation in northern Italy, whereas foliage from the same sites showed no apparent relationship with altitude (55). The relative proportion of heavier PCBs increased in foliage samples from high altitude sites in Italy relative to the composition at lower elevations (55). Both Weiss et al. and Notarianni et al. noted marked differences in the composition of SOC mixtures between foliage and surface soil, namely, a higher relative abundance of the heavier constituents in soil relative to foliage from the same location (51, 55). The more hydrophobic and less volatile PAHs are also found in greater proportion in soils compared to that in needles (51). This is likely due to the different relative importance of gas and particle-associated deposition for foliage and soils. Wet deposition—in particular, in the form of snow—will often be quite ineffective in delivering SOCs to needles (because of the throughfall and blowoff of snow) but effectively transfers contaminants to soil.

Nitrated phenols and haloacetic acids are phytotoxic organic chemicals, and their presence in the foliage of alpine vegetation has been measured to assess their potential contribution to forest decline. Several European studies (7, 9, 54) noted the presence of nitrophenols—in particular, 4- and 2-nitrophenol—in foliage from alpine sites. A relationship between the levels and proximity to road traffic suggests the importance of vehicular emissions (54). Two profiles with three stations at different elevations showed highest levels at the intermediate altitude (9, 54), which was linked to the occurrence of inversions. Nitrophenols are much more water-soluble than most of the other organic compounds considered here, which supports the possibility that radiation or up-slope fogs, forming preferentially at certain altitudes, could enhance their deposition to vegetation. The measured concentrations were similar to levels that are believed to cause damage to plants (7, 9, 54), and a site with serious forest damage had significantly higher nitrophenol levels than a healthier site (7). Another set of potentially phytotoxic compounds identified in foliage at high altitudes are haloacetic acids and a variety of volatile halogenated hydrocarbons, which are believed to be their precursors (e.g., chloroform, tetrachloromethane, trichloroethylene). Trichloroacetic acid (TCA), in particular, was repeatedly detected in spruce needles of remote Austrian forest sites, but no clear altitudinal gradients were apparent (8, 9, 56). Enrichment of haloacetic acids in fogwater, relative to rainwater (57), suggests that altitudes with a high incidence of fog may be affected more strongly. Although it has been noted that the TCA levels in soils of coniferous forests in mountainous regions of Central Europe are significantly elevated, there is evidence that this compound is naturally formed in soils (58).

Organic Pollutants in Alpine Lakes

Characteristics of Alpine Lakes. Traditionally, research on bioaccumulating contaminants has focused on aquatic food chains. This is also the case for alpine regions, where mountain lakes have attracted considerable attention, mostly as result of a series of European Union projects (AL:PE, MOLAR, EMERGE) on alpine lakes (59) and intensive studies aimed at understanding the dynamics of organic contami-

nants in Bow Lake (43, 60, 61) and other lakes (62) in the Canadian Rocky Mountains. High mountain lakes are situated above the treeline, are often seasonally ice-covered, and form the headwater catchments of water supplies. Aquatic ecosystems of alpine environments are normally oligotrophic and have a low content of suspended particulate matter (63, 64). Because low ambient temperatures restrict species diversity, pelagic food webs of these lakes tend to be rather simple (64, 65). Some studies on organic contaminants in alpine lakes have focused on investigating the pathways that deliver the contaminants to the lakes, whereas others quantified the occurrence of organic contaminants in the water column and sediments of alpine lakes. A large body of work seeks to explain the variability in the contaminant concentrations in biota from different lakes. Surprisingly few studies actually explored the transfer of pollutants in an alpine lake foodweb.

Pathways of Organic Contaminants into Alpine Lakes.

Contaminants may enter alpine lakes via atmospheric deposition or runoff. Alpine lakes may be especially sensitive to airborne contaminants, because their catchments often have only thin soils and sparse vegetation cover and, thus, are unable to act as filters to prevent contaminants from reaching surface waters (63). On the other hand, studies of European mountain lakes have seemed to assume that direct atmospheric deposition to the lake is the major input route. The input with runoff was not quantified, presumably because it was deemed negligible (66). Grimalt et al. (67) noted that sediments in Estany Redó and Lake Ladove had generally higher SOC concentrations than soils in the same alpine watershed but excluded the possibility of watershed-to-lake transfer, based on the low water solubility of the contaminants and radiometric measurements, which indicated no soil erosion. A study on the SOC dynamics in Arctic lake systems, which share many characteristics with high alpine lakes, revealed that the lakes received most of their SOC load with snowmelt that entered via tributary inflow (68). The mass balance of SOCs in Bow Lake also suggests that input with tributaries cannot always be neglected (43, 61). Measured and calculated fluxes of OCPs and PCBs to the lake were used to determine the relative contribution of precipitation, runoff, and gas absorption, concluding that glacially derived tributary inputs are dominant SOC sources to glacier-fed alpine lakes. In terms of contaminant loss, outflow was proven to be more important than volatilization in Canadian Rocky Mountain lakes (69).

Measurements in Lake Water. Even though it is often logistically difficult to collect the large volume water samples from alpine lakes that are required for reliable SOC quantification, water concentrations have been reported for several high altitude lakes (Table 4). Most commonly detected are PCBs, HCHs, and endosulfan. The latter compound, which is one of the few OCPs still in widespread use, had much higher levels in lakes in the Sierra Nevada (28) than in Europe (63), presumably because of intensive use in the nearby Central Valley. Levels of HCHs were similar in the investigated lakes (63, 70), and high when compared to other continental waters (63). Comparison of PCB concentrations is complicated by the different reporting conventions, but Lake Tahoe and Lake Marlette (29) had similar levels to those reported for the European lakes (71). PCB concentrations reported for a Himalayan lake are unreasonably high, and a detection limit of 0.1 ng/L suggests that the sampling volume (5L) was too small (70). Vilanova et al. (63, 71, 72) studied, in detail, the occurrence of PAHs, PCBs, and OCPs in the water column of three European lakes. HCH levels in these lakes were among the highest reported for continental waters, whereas the concentrations of DDT and HCB were comparatively low (63). PAH concentrations in the three lakes were an order of magnitude lower than those in waters from natural environ-

TABLE 4. Studies Reporting on Organic Contaminant Concentrations in the Water and Sediments of Alpine Lakes

lake (elevation, ^a mountain region, country)	compound(s)	year	ref
Water			
Cabin Lake (1219 m), Emerald Lake (1300 m), Annette Lake (1019 m), Maligne Lake (1671 m), Bow Lake (1940 m), Rocky Mountains, Canada	toxaphene	1994	88
Inferior Lake (5067 m), Sagarmatha National Park, Nepal	PCBs, OCPs	1994	70
Lake Tahoe (1899 m), Marlette Lake (2384 m), Sierra Nevada, CA/NV, USA	PCBs	1995	29
eight surface waters (118–3322 m), Sierra Nevada, CA, USA	OPPs, endosulfan	1997	28
Estany Redó (2240 m), Pyrenees, Spain, Gossenköllesee (2417 m), Alps, Austria, Øvre Neådalsvatn (728 m), Norway	PCBs, PAHs, OCPs	1996–1998	63, 71, 72
Bow Lake (1940 m), Rocky Mountains, Canada	OCPs, PCBs	1998	43, 61
lakes in parks across the western United States	SOCs	2003	107
Sediments			
Sagamore Lake (586 m), Woods Lake (615 m), Adirondack Mountains, NY, USA	PAHs	1978	112
Lake Husted (3380 m), Lake Louise (3365 m), Lake Haiyaha (3109 m), the Loch (3109 m), Rocky Mountain National Park, CO, USA	PAHs, PCBs	1981	73
Dösener See (2270 m), Central Alps, Austria	PAHs	1987	74
Superior Lake (5213 m) and Inferior Lake (5067 m), Sagarmatha National Park, Nepal	PCBs, OCPs	1992	70
Grosser Arbersee (935 m), Bavarian Forest, Germany	PCBs, PCDD/Fs	1993	79
Moab Lake (1204 m), Cabin Lake (1219), Emerald Lake (1300 m), Annette Lake (1019 m), Beauvert Lake (1021 m), Bow Lake (1940 m), Rocky Mountains, Canada	toxaphene	1994	88
Escura (1680 m), Serra Estrela, Portugal, Cimera (2140 m), Sierra Gredos, La Caldera (3050 m), Sierra Nevada, Estany Redó (2240 m), Pyrenees, Spain, Noir (2750 m), Alps, France, Schwarzsee ob Sölden (2799 m) and Gossenköllesee (2417 m), Alps, Austria, Dlugi Staw (1783 m) and Starolesnianske Pleso (2000 m), Tatra, Poland/Slovakia, Øvre Neådalsvatn (728 m), Norway	PAHs, PCBs, HCB, DDE	1993–1995	75, 81, 82
Estany Redó (2240 m), Pyrenees, Spain, Dlugi Staw (1783 m), Starolesnianske Pleso (2000 m), Lake Ladove (2057 m), Tatra, Poland/Slovakia	PCBs, DDTs, HCB	not given	67
Lake Planina (1430 m), Alps, Slovenia	PAHs	2000	76
Chungara Lake, Andes, Chile	PCBs	1999	78
wetland and lake sediments, Tibetan Plateau	HCHs, DDTs	not given	80

^a Elevation given in meters above sea level.

ments near urban centers or polluted areas but higher than those in more remote lakes (72).

Measurements in Lake Sediments. Concentrations of PAHs and halogenated SOCs in sediments have been reported for numerous mountain lakes (see Table 4). Many of these studies inferred temporal atmospheric deposition trends from the concentration profiles with depth in sediment cores. Higher PAH concentrations toward the surface of the sediments are explained by increases in the atmospheric deposition of combustion-generated PAHs (73–77). Sometimes, the most recent sediments indicate a decrease in pyrolytic PAH deposition, and many lakes show peak concentrations in layers corresponding to the 1950s and 1960s (75, 76). Low concentrations of alkylated PAHs suggest a predominance of combustion sources. Retene and perylene often had depth profiles different from other PAHs, which is attributed to diagenetic generation from natural precursors (75–77). PCBs were observed to be elevated in surface sediments compared to samples taken at greater depths in several of the investigated mountain lakes (67, 73, 78), whereas, in some European lakes (67, 79), the uppermost sediment layers had lower concentrations, suggesting a reduced atmospheric input in more recent times. Decreasing concentrations at the surface were also noted for PCDD/Fs in the Grosser Arbersee (79). In the Tibetan Plateau, OCP levels were higher in the upper layers of the core (80). The analysis of high-altitude lake sediments also served to infer spatial trends in the atmospheric deposition flux of SOCs to several European mountain areas (81, 82). The geographical pattern of the PAH flux paralleled the atmospheric deposition flux of sulfate, indicating atmospheric PAH transport by combustion particles (81). The sediment inventories for

several PCB congeners were positively correlated with altitude and inversely correlated with temperature across 19 European high mountain lakes (82), although no such relationship was observed for DDE or HCB.

Measurements in Organisms from Alpine Waters. In what possibly constitutes the first study on organic contaminants in mountains, Cory et al. (83) reported the widespread occurrence of DDE in frogs in the Californian Sierra Nevada in 1970. DDE was present at elevations as high as 4500 m, and DDE contamination was higher on the western slope of the mountains than across their crest on the east face. Since then, PCBs, OCPs, and OPPs have been quantified in fish and amphibians from European and North American mountains (Table 5). Even PAHs, which are readily metabolized, have been monitored in alpine fish through the analysis of fish bile (84). Before discussing the result of these studies, it is worthwhile to explore whether the common characteristics of alpine lake ecosystems previously discussed favor the bioaccumulation of SOCs. Studying the relationship between lake productivity and contaminant bioaccumulation in predatory fish from numerous lakes across southern Scandinavia, large differences in PCB and DDE levels could be attributed to variation in lake trophic status and content of humic substances (85). Lower contaminant levels in fish from lakes of higher productivity were explained by a higher growth rate of fish and a higher turnover and sedimentation of particles (phytoplankton) to which the pollutants sorb. Growth rates in alpine aquatic organisms are slow not only because of the oligotrophic status of the lakes, but also because low water temperatures may slow metabolism and excretion of contaminants in ectothermic organisms. Indeed, Blais et al. (62) attributed higher concentrations of more

TABLE 5. Studies Reporting on Organic Contaminant Concentrations in Aquatic Organisms from Alpine Lakes

species	chemicals	variables that correlated with some concentrations	variables that did not correlate with any concentrations	ref
lake trout	PCBs, OCPs	fish weight, lake drainage basin area, fish lipid content	trophic structure, lake latitude	87
rainbow trout, brook trout, lake trout, longnose suckers, mountain whitefish	toxaphene	fish age, dissolved P, OC in lake sediment, lake temperature, lake altitude	fish weight, fish lipid content	88
amphipods	HCB, HCHs, PCBs, DDTs, other OCPs	fish lipid content, fish growth rate, lake altitude, lake temperature	lake DOC, lake total phosphorus lake chl a concentration	62
salmonids (arctic charr, brown trout, rainbow trout, brook trout)	HCB, HCHs, PCBs, DDTs	lake altitude, lake temperature	fish length, fish weight, fish age, fish lipid content, lake DOC, lake total P, mean annual rainfall, lead deposition, sulfur deposition	82
salmonids (arctic charr, brown trout, rainbow trout, brook trout)	HCB, HCHs, PCBs, DDTs	fish age and sex, lake altitude, lake temperature, lipid content, conditioning factor	fish length, fish weight	90
European minnow	p,p'-DDE, PCBs	lake latitude	lake altitude, all water parameters (including pH, conductivity, alkalinity, calcium, phosphorus, nitrate)	89
Pacific tree frog tadpole	PCBs, toxaphene	east-west drainage aspect, lake altitude, lake latitude, central valley-to-site distance		91

volatile SOCs in amphipods sampled at higher elevations in the Canadian Rocky Mountains to slower growth rates and longer lifetimes. On the other hand, food chains in alpine lakes tend to be short, which should limit the extent of potential biomagnification. Stable isotope data demonstrated that the food chain in Bow Lake consists only of three trophic levels (60), suggesting that, unlike in Arctic areas (86), high contaminant concentrations in mountain fish cannot be explained by a long food chain.

Several of the studies on contaminants in biota from alpine lakes sampled the same or similar species from several high-altitude lakes and then tried to explain differences in the measured contaminant concentrations by statistical correlation with a variety of ecosystem parameters characterizing the lake, its catchment, chemistry, and biological community (62, 82, 87-91). These analyses are meant to shed light on the factors controlling the bioaccumulation of SOCs in alpine freshwater food webs. However, the issue is quite complex, because differences in the measured organism concentrations can be the result of differences in (i) the atmospheric deposition to the lakes' catchment, (ii) the watershed retention of the contaminants, (iii) the in-lake processing of the contaminants, (iv) the trophic status of the lake, and (v) the age of the organisms. The possibility for a direct comparison between studies is further impeded by differences in the ecosystem variables that were examined.

The content of DDE and PCB in trout from 14 lakes in the Canadian Rocky Mountains was directly related to the drainage basin area and weight of the fish but not to fish age or trophic class (87). DDE levels were also related to the lipid content. The correlation with drainage basin size was interpreted as suggesting that the atmosphere is an important contaminant source. DDE and PCB concentrations in minnows from 17 remote European mountain lakes were independent of altitude, catchment area, and all measured water parameters, including pH, alkalinity, and dissolved phosphorus (89). However, DDE concentrations were linked to agricultural source regions and prevailing trajectories. Grimalt et al. (82) performed congener-specific regression analyses between the SOC content in the muscle tissue of salmonids from 19 European high mountain lakes and a variety of lake characteristics. The concentrations of DDE and the heavier PCB congeners increased with elevation and decreased with temperature, meaning that higher concentrations were observed in fish from colder, higher lakes. Correlations with temperature and altitude were not significant for α - and γ -HCH, HCB, and the lighter PCBs. Neither were correlations observed with lipid content, lake productivity, or mean annual rainfall. This study concluded that the less volatile compounds are accumulating in fish at high altitudes. Vives et al. (90) extended the data set with fish samples from additional European lakes and came to similar conclusions; however, they also noted that fish age explains some of the variance in the concentrations of the less volatile SOCs. In contrast to these studies, work on an amphipod in several western Canadian mountain lakes showed that only the more volatile SOCs increased in concentration with altitude (62). The concentrations were independent of lake productivity. For these more volatile chemicals, significant correlations were also observed with water temperature, lipid content, and growth rate. In fact, multiple regression analysis revealed that the growth rate removed the significance of elevation, temperature, and lipid content for all compounds, suggesting that it was the factor with the most influence on the contamination levels. Regression analysis on toxaphene residues in fish from 13 western Canadian lakes showed concentrations to be positively correlated with lake elevation and fish age and negatively correlated with mid-summer temperature, dissolved phosphorus level, and percentage of organic carbon in lake sediment; the last three variables are

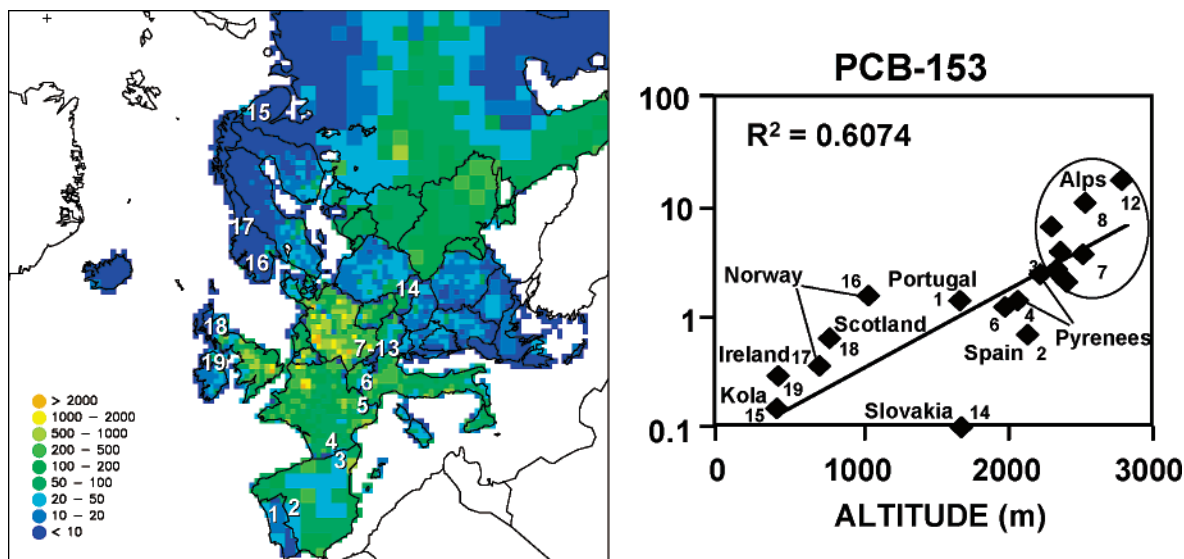


FIGURE 2. Location of alpine lakes studied by Grimalt et al. (82) superimposed onto a map of the cumulative usage of PCBs in Europe (92). The regression of the concentrations of PCB-153 and other heavier PCB congeners in fish and sediments with lake altitude reported by Grimalt et al. (82) are likely a result of the closer proximity of high-altitude lakes in the Alps to PCB sources in central Europe. Low concentrations in the low-altitude alpine lakes in northern Europe are a result of greater distance from PCB sources.

indicators of lake productivity (88). Relationships with fish weight and lipid content were not significant, which means that oligotrophic lakes at high elevations had the highest levels of toxaphene, whereas eutrophic lakes at low elevations had lower levels.

One should note that the above correlations can be fortuitous and are not necessarily an indication of a causal relationship. In particular, the correlations on fish and sediments from European lakes (82, 90) must be treated with caution, because they involve data from several mountain regions, which differ in terms of altitude and proximity to SOC source areas. A closer inspection of the investigated lakes shows that virtually all of the higher lakes (>2400 m) are located in the European Alps, whereas the lower-lying lakes (>1100 m) are in northern Europe (Norway, Scotland, Ireland, Kola peninsula) (see Figure 2). The lakes from the Pyrenees, the Iberian Peninsula, and eastern Europe cover the intermediate elevation range (1600–2300 m). The SOC, especially the PCB, source strength is much higher in densely populated and highly industrialized central Europe, and much lower in remote northern Europe (92) (see Figure 2). In other words, the altitude of the mountain lakes is not independent from the proximity to the sources of SOCs. Could the higher concentration of the heavier PCBs in the high-altitude lakes be due to their location in central Europe, and the lower concentrations in the low-lying lakes in northern Europe be due to their relative remoteness? The more volatile SOCs have a more uniform atmospheric distribution across Europe, and air concentrations are less influenced by the proximity to source regions (93). This would explain why these chemicals show less of a relationship with “altitude”.

Surprisingly, studies that report on organic contaminants in mammals and birds from mountain regions are very rare, although there are a few studies on birds from the mountains of Western Canada. Elliott et al. (94) measured the content of SOCs in fish-eating osprey, and Morrissey et al. (95, 96) studied the uptake of PCBs and OCPs in American Dipper from the Chilliwack River watershed, located in the Coast Range, west of the heavily populated and agriculturally cultivated lower mainland of British Columbia. Concentrations in eggs were positively related to drainage basin area but not elevation. Also, levels were higher in eggs that were collected during a year with unusually high snow deposition.

Concentration Differences along Altitudinal Gradients

A common theme throughout the research of organic contaminants in mountains is the investigation of concentration differences along altitudinal gradients. Especially intriguing are incidences when contaminant concentrations increase with altitude, because they may indicate that mountain regions act as regional convergence zones, leading to the localized enrichment of, and thus potentially to elevated exposure of sensitive ecosystems to, contaminants at high altitudes (3). One approach to derive such altitudinal gradients, exemplified by the European studies on fish from alpine lakes discussed previously (82), measures contaminant concentrations at a selection of sites from various regions at several altitudes and then seeks statistical relationships with elevation. For example, Calamari et al. (47) studied contaminants in plants across 26 areas worldwide and thus was able to compare concentrations measured at four high-altitude sites with the other measurements. However, this approach neglects that the various sites will most likely differ in terms of proximity to contaminant sources. In this context, it is noteworthy that Weiss et al. (51) observed altitudinal patterns when investigating transects along one mountain slope but failed to observe altitude-related differences in SOC concentrations when using the entire foliage and soil data set, which included samples from across the Austrian Alps. This is likely due to the different proximity to sources of these sampling sites, even though they are all within the same mountain range. In other words, studies that are intended to reveal contamination gradients with altitude must be conducted using sites that have comparable distances from the sources.

Of most use, therefore, are studies that sample along a mountain slope. This way, the effect of altitude can be assessed more directly. Increasing SOC concentrations with altitude have been noted in a range of different sampling media in the Canadian Rocky Mountains and along two mountain slopes in the Alps. Substances for which this has been observed include HCHs (31, 38, 62, 97), β -endosulfan, heptachlor, and heptachlor epoxide (32, 38, 97), DDT-related compounds (32, 51, 62, 97), HCB (32, 62, 97), dieldrin (62), and toxaphene (91). PCB concentrations have also been shown to increase with altitude (38, 91), as well as PCDD/Fs

(51). To provide an idea of the magnitude of the increase, Blais et al. (38) reported a 10- to 100-fold increase of snow concentrations between altitudes of 770 m and 3100 m. Smaller, but still significant, increases of up to 1 order of magnitude were observed in air and foliage across elevation ranges of 770–2200 m (97), 840–1650 m (51), 800–2300 m (31, 32), and 1024–2320 m (62). The Canadian studies (31, 32, 62, 97) had a tendency to show increases with elevation for only the more volatile SOCs; however, the picture is not entirely clear. Weiss et al. (51) observed increases for fairly involatile SOCs, such as the PCDD/Fs along transects in Austria. Similarly, hepta- and octachlorinated PCNs predominated in frog tadpoles at high-altitude sites in the Sierra Nevada (91).

The field data on SOC contamination patterns with altitude can be bewildering in their complexity. Some studies show clear concentration gradients or compositional shifts, whereas others do not. Sometimes such gradients and shifts are seen in some compartments, but not in others. Sometimes, a certain altitudinal pattern is observed for one group of compounds, but not another. Here, we try to discuss the various factors that are likely controlling the altitudinal gradients in SOC concentrations. The location of the mountain sampling sites relative to the sources, is obviously an important consideration, because atmospheric concentrations and deposition rates are generally higher in the proximity of sources. If the mountains are remote from contaminant sources, source location should not impact significantly on altitudinal gradients in concentrations. If, however, the contaminant sources are within or close to the mountains, it may be the dominant factor that controls the concentration gradients. This is clearly shown by the studies on OPPs in the Sierra Nevada. Concentrations of SOCs in surface media (soil, foliage, sediments, water, etc.) are established as the result of a dynamic process of deposition and volatilization, with higher concentrations occurring when deposition rates are high relative to volatilization rates. For more volatile SOCs, such as HCB and HCHs, even the atmospheric concentrations are strongly influenced by reversible air–surface exchange. Many chemical and environmental factors influence the rates of deposition and volatilization. Many of these environmental factors change with altitude, and often not in a simple fashion. To make matters even more complicated, these environmental factors undergo strong seasonal variations in most mountain environments. Thus, the lack of simple and recurring contamination patterns with altitude is not surprising. To shed some light on this issue, we may nevertheless attempt to discuss how the rates of SOC deposition and volatilization may typically change with altitude. We restrict this discussion to gradients of relatively persistent and not highly water-soluble chemicals in temperate mountains, because only for these do we have some field evidence.

With respect to wet deposition, we already stated that the amount of precipitation generally increases with elevation, and precipitation at higher elevations is generally occurring at lower temperatures and is thus more likely to be in the form of snow. The extent of wet deposition of SOCs is relatively independent of the underlying surface, but transfer of SOCs will be more efficient to soil than to foliage, because of throughfall and the lack of foliage in the winter. The wet deposition route is most important for water-soluble SOCs (gas scavenging, e.g., HCHs) and SOCs with low volatility (particle scavenging, e.g., heavier PCBs, PAHs), and the relative importance and efficiency of wet deposition of SOCs increases as the temperature decreases (18). Overall, this suggests that wet deposition of most SOCs should generally increase with elevation (38); however, this is more likely reflected in snow and soil gradients, and less so in foliage.

Dry deposition of gases has a tendency to be higher at low temperatures. The association of SOCs with particles in the atmosphere is also higher at low temperatures, and particles tend to deposit faster than gases. Dry deposition also increases with wind speed. Because it is generally colder and windier at higher elevations, this would enhance dry deposition of both gases and particles at higher altitudes. On the other hand, the dry deposition of SOCs is strongly influenced by the surface type. For example, the deposition of gaseous and particle-bound SOCs to forested surfaces is higher than that to open areas (98) and thus may be higher below the treeline than above it. The issue is made more complex as the surface type and its roughness changes with the season, depending on snowcover and the status of the vegetation. We must conclude that the altitudinal dependence of the dry deposition of SOCs is not easily generalized and dependent on local conditions, with respect to forest and snowcover.

The volatilization rate of an SOC is related to the capacity of the surface to retain the chemical, with volatilization being higher when the capacity is low. This capacity is higher at low temperatures. It is also higher for an organic-rich soil and dense vegetation cover than for bare rock and sparse alpine vegetation. Volatilization is greatly diminished from a snow-covered surface. Theoretical investigations suggest that the capacity of the snowcover itself is related to snow surface area (especially for volatile chemicals) and particle content (for less volatile chemicals) (99). Wind speed enhances volatilization to the same extent that it does dry vapor deposition. The capacity of the surface is also much greater for involatile chemicals than for more volatile SOCs. We may conclude that, whereas lower temperatures at high altitudes reduce volatilization, the capacity of the surface compartments above the treeline is likely to be fairly small, especially during the summer and for relatively volatile SOCs. Therefore, these may volatilize quite readily from both aging snow and from alpine ecosystems during summer. Extensive summertime revolatilization of large contaminant loads deposited with snow during the winter served as an explanation for increasing air concentrations of HCHs with elevation in the Canadian Rocky Mountains (31).

Based on these considerations, the following general picture of SOC dynamics along a temperate mountain slope can be drawn (see Figure 3). SOCs that are subject to precipitation scavenging (i.e., both particle-sorbed and water-soluble chemicals) may experience elevated deposition at higher elevations, especially during the wintertime. During snowmelt, some of the more volatile SOCs may partially revolatilize, whereas most SOCs will be delivered to the surface media. During the summer, revolatilization (especially of the more volatile SOCs) at high elevations (above the treeline) may be quite high, whereas in forested areas, it may be low. Less volatile SOCs have the potential to accumulate at all elevations. Several important findings transpire: (i) snow has a crucial role in the behavior of SOCs in temperate mountains by modifying the air–surface exchange; (ii) SOCs will behave quite differently based on volatility, and more volatile SOCs may undergo intense air–surface exchange above the treeline; (iii) SOC gradients in soil can be expected to be different above and below the treeline; and (iv) altitudinal gradients can be expected to be quite different in different environmental media. The aforementioned considerations assume that the exchange of air along the altitudinal transect is not constrained. If a mountain slope experiences regular and strong inversions, this is likely to disrupt the generalized transport patterns. Considering the complexity of the processes that control the altitudinal gradients in SOC concentrations, model simulations may have a useful role in assessing the relative importance of the various contributing factors. This would also allow an

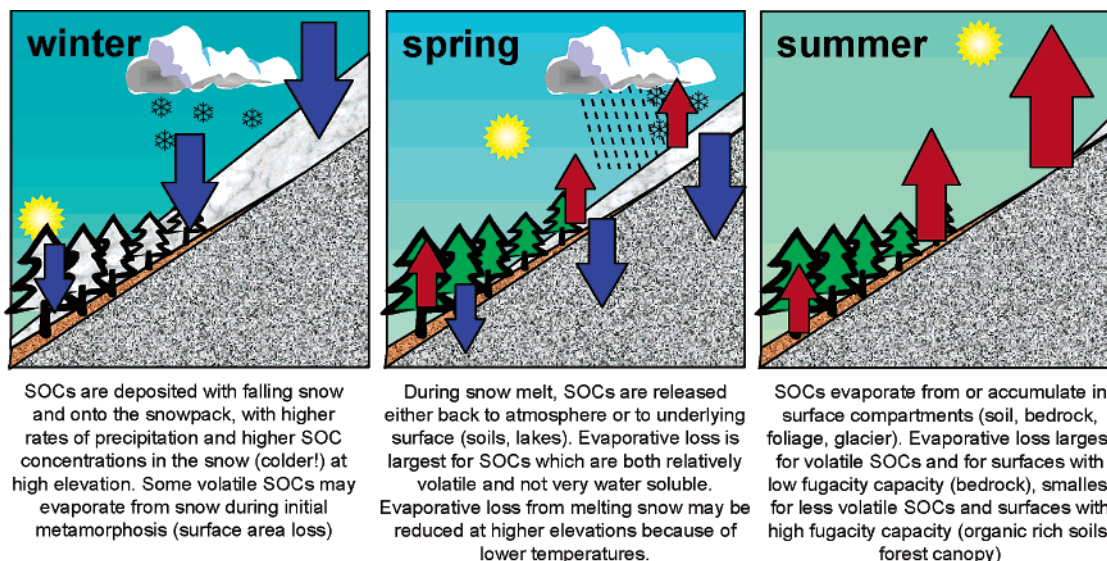


FIGURE 3. Illustration of the processes governing air–surface exchange of semivolatile organic contaminants (SOCs) along a temperate mountain slope in different seasons. The balance of deposition (blue arrows) and revolatilization (red arrows) will determine the change of SOC concentrations along an elevation gradient. SOC of different volatility will differ in terms of their atmosphere–surface exchange.

examination of the influence of chemical properties, as accumulation patterns will be different for water-soluble chemicals.

Research Needs

There are indications that mountains act as regional convergence zones for selected organic chemicals. The studies with OPPs suggest that only relatively persistent chemicals are subject to this convergence. Mimicking the divergent transport behavior of different SOC on a global scale (“global fractionation”) (100, 101), the more volatile constituents of SOC mixtures seem to become enriched relative to the less volatile constituents at higher altitudes (“altitudinal fractionation”). For selected contaminants, it is even possible that concentration inversions (i.e., higher concentration at higher altitudes) are established. This also has parallels on a global scale. In fact, it is very often the same substances (HCB, HCHs, lighter PCBs) that show this behavior along latitudinal and altitudinal transects. We suggest that the same factors that have been implicated in the global fractionation process are operating along elevation gradients. In particular, strong temperature gradients with elevation, in combination with the temperature-driven exchange of the more volatile SOC between the surface and the atmosphere, can result in the net transfer of chemicals from lowlands to higher altitudes. The effect may be even more enhanced on a regional scale relative to the global scale because of (i) the much smaller distances that the chemicals must travel to experience large temperature differences and (ii) the specific diurnal wind systems that are characteristic of mountain regions, which blow up-slope when it is warm and down-slope during the night. Other factors that enhance the concentrations in polar areas, such as low degradation potential due to limited biological activity, are likely also operating at high altitudes. In comparison to the polar regions, mountain regions are often much closer to emission sources, which suggests that chemicals can reach these regions even if they have a relatively low long-range transport potential.

A notable difference between the cold trapping in high latitudes and high altitudes is the likely importance of precipitation. Whereas the polar areas are almost deserts with often very low rates of precipitation, high altitudes generally experience high rates of precipitation, and elevation gradients are often also gradients in the rate of precipitation. This may actually constitute the dominant orographic

trapping mechanism for some SOC. Precipitation rates and heavy metal deposition rates were strongly correlated along several gradients in the Austrian Alps, leading to concentrations in mosses that increase with elevation (41). Persistent particle-bound SOC can be expected to show a similar convergence behavior. In mid- and high-latitude mountains, snow is likely to have a dominant role in the fate of organic chemicals. Not only may falling snow be a very important deposition route, but a long seasonal snow cover strongly affects the air–surface exchange. Of particular importance for the delivery of organic contaminants to alpine ecosystems will be the fate during snowmelt.

In trying to understand the contaminant convergence mechanisms in mountains, the large diversity of mountain regions must be kept in mind. For example, it is important to recognize the differences and similarities between tropical and temperate mountains. Mountains in both climate zones feature temperature and precipitation gradients up-slope. However, in many tropical mountains, snowcover and seasonal temperature fluctuation are obviously of little concern, whereas there is often a marked seasonality in the precipitation rate. Also, transitions in surface cover with altitude are often very different, with lush and dense forests on tropical mountains at elevations that are barren in temperate mountains. Beyond differences between mountain areas of different climatic zones, it is likely that every mountain area is possibly quite unique with respect to contaminant fate. Examples of potentially important regional peculiarities are the influence of the monsoon in the Himalayas, and of a strong elevated inversion at Pico de Teide.

Although the research conducted over the past decade has greatly improved our understanding of organic contaminants in mountains, there are several areas where additional efforts are required to fill important knowledge gaps. The most pressing among these are as follows.

There have been very few studies on concentrations, bioaccumulation, and potential effects of SOC at higher trophic levels in alpine aquatic food chains, in particular, birds of prey and predatory mammals (94). Even less work has been conducted on natural terrestrial food chains in alpine regions and their top predators. Surprisingly, there are also no studies on the bioaccumulation of organic contaminants in alpine agricultural food chains. Given that the concern surrounding SOC is based on their potential effects on animals at the top of the food chain, including

humans, this is a knowledge gap that should be addressed (102). A focus should be placed on mountain regions where humans rely on traditional high-altitude agriculture (especially meat and dairy production) for subsistence.

Research projects should be designed with the intent to identify the chemical and environmental factors (such as temperature and precipitation gradients, surface cover and biological characteristics, source location) that control organic contaminant convergence in mountain regions, and specifically to determine what is responsible for the concentration gradients that increase with altitude. Important questions to be addressed include the following: What mountain regions will be particularly effective convergence zones, and what contaminants will be particularly prone to orographic cold trapping? Measurements of gradients along mountain slopes with only a weak influence of local pollution sources are likely to have a significant role in this regard, because the possibility of identifying the controlling factors from measurements at different altitudes within the same major mountain range (51), or even in several mountain regions (82), is limited. It should be particularly useful to compare accumulation patterns along gradients on the leeward and windward slope of a mountain range, or gradients in mountains with widely different characteristics and environmental gradients. In particular, more work must be directed at organic contaminants in mountains at low latitudes and the Southern hemisphere. Major mountain regions such as the Andes and the Himalayas have received little attention, and, so far, no work on contaminants in tropical mountains has been reported. However, indications are that organic contaminant fate and bioaccumulation in mountains may be very different under different climatic, geographic, and ecological circumstances. We expect simulation models to have a useful role in disentangling the interactions of environmental gradients, in regard to controlling organic chemical fate in different mountains.

Most studies in mountains, so far, have focused on traditional organic contaminants, such as the PCBs and OCPs. Future research should be conscious of the potentially large number of emerging organic contaminants that may find their way into alpine ecosystems. Of particular interest should be substances that are both persistent and water-soluble, and can be emitted to, or formed in, the atmosphere. Such substances could potentially experience elevated deposition rates in mountains because of high precipitation rates and high wash-out efficiency. The Western Airborne Contaminants Assessment Project (WACAP) (103) is currently investigating OCPs, PCBs, and currently used chemicals (i.e., pesticides, flame retardants) in mountainous parks across the western United States and thus is promising to address these data gaps.

For logistical reasons, most research on contaminants in temperate mountain regions has been conducted in the summertime. However, mountains at mid and high latitudes experience dramatic seasonal changes, in particular, as a result of long-lasting and thick seasonal snow covers. What happens during snowmelt in temperate mountains could be dominating the overall contaminant behavior at high elevations (99), and year-round investigations, with a focus on deriving better temporal resolution during the springtime, should be a priority.

Another research topic that may merit attention is the likely impact of climate change on organic contaminant fate in mountains. Many of the models and scenarios used to forecast global climate change predict that mountain regions will be strongly affected by rising temperatures and changing precipitation patterns (1). The sensitivity of the phase transition of water at the melting point to even small temperature variations, and the major impact that snow and ice can have on the environmental behavior of organic

contaminants (99), led MacDonald et al. (104) to suggest that climate change's impact on organic contaminant fate may be amplified in cold regions. High-altitude ecosystems—in particular, alpine lakes (105)—are believed to be particularly sensitive, and even slight environmental changes may substantially affect ecosystem functions. Thus, mountain regions should constitute the ideal field laboratory to study the interaction of climate change and organic contaminant fate.

Acknowledgments

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