

Accumulation of Current-Use Pesticides in Neotropical Montane Forests

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In Central America, chemical-intensive tropical agriculture takes place in close proximity to highly valued and biologically diverse ecosystems, yet the potential for atmospheric transport of pesticides from plantations to national parks and other reserves is poorly characterized. The specific meteorological conditions of mountain ranges can lead to contaminant convergence at high altitudes, raising particular concern for montane forest ecosystems downwind from pesticide use areas. Here we show, based on a wide-ranging air and soil sampling campaign across Costa Rica, that soils in some neotropical montane forests indeed display much higher concentrations of currently used pesticides than soils elsewhere in the country. Specifically, elevated concentrations of the fungicide chlorothalonil, the herbicide dacthal, and the insecticide metabolite endosulfan sulfate on volcanoes Barva and Poas, lying directly downwind of the extensive banana plantations of the Caribbean lowland, indicate the occurrence of atmospheric transport and wet deposition of pesticides at high altitudes. Calculations with a contaminant fate model, designed for mountain regions and parametrized to the Costa Rican environment, show that chemicals with a log K_{AW} between -3 and -5 have a greater potential for accumulation at high altitudes. This enrichment behavior is quantified by the Mountain Contamination Potential and is sensitive to contaminant degradability. The modeling work supports the hypothesis suggested by the field results that it is enhanced precipitation scavenging at high elevations (caused by lower temperatures and governed by K_{AW}) that causes pesticides to accumulate in tropical montane areas. By providing for the first time evidence of significant transfer of currently used pesticides to Central American montane cloud forests, this study highlights the need to evaluate the risk that tropical agricultural practices place on the region's ecological reserves.

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Introduction

Central America has a significant agricultural industry, with coffee, sugarcane, rice, pineapple, and banana as major crops (1). Tropical agriculture is often very chemical-intensive. The use of plant protection products per hectare is higher in Costa Rican agriculture than in most industrialized countries (2). Central America is also a region of exceptionally high biodiversity, with ecosystems that are highly valued both ecologically and economically (3). The Central American Cordillera in particular is home to forests which are among the most diverse ecosystems in the world (4) and also among the most vulnerable (5). Plantations with high usage of pesticides are often in immediate vicinity and/or upwind of protected areas, such as National Parks and Biological Reserves (6).

Temperature gradients can cause contaminant convergence at high latitudes (7) and high altitudes (8). In particular, lower temperatures and efficient snow scavenging, in combination with effective air mass transport, have been implicated in focusing semivolatile organic contaminants in temperate mountains (9). Transport of current-use pesticides into high elevation ecosystems has been documented in North America (10, 11). Trade winds blowing easterly from the Caribbean Sea for most of the year cause rising air motion, orographic precipitation, and persistent cloud cover at higher elevations of the Central American Cordillera. High rain rates, steep temperature gradients, and soils rich in organic matter should also favor the accumulation in tropical mountains of organic chemicals that are readily scavenged by rain and fog. We know of no previous studies that have investigated this issue, even though tropical montane forests have experienced high amphibian extinction rates (12), and pesticides have been implicated in the decline of amphibian populations elsewhere (13). In order to quantify and interpret the spatial distribution of pesticides in Costa Rica's abiotic environment, air and soil were sampled at numerous sites across the country and analyzed for the content of chlorinated organic pesticides in current use. While some of the target chemicals have been measured at select locations in Costa Rica before (6, 14, 15), this is the first time that a truly national picture of pesticide distribution in a Central American country emerges.

Experimental Section

Sampling. Twenty-three sampling sites were located throughout Costa Rica (Table S1 and Figure S1 in the Supporting Information). With the aim to investigate potential factors that may influence pesticide distribution, the sampling sites varied widely in terms of temperature, precipitation, vegetation cover, soil properties, altitude, exposure to prevailing winds, and proximity to pesticide usage. Figure S2 shows the agricultural and urban areas of Costa Rica. Sampling occurred mostly in protected areas where no pesticides had been used in the past.

Annually averaged concentrations in the atmosphere were determined with passive air samplers. These samplers consist of a stainless steel mesh cylinder that is filled with XAD-2 resin and suspended in a steel can with an open bottom (18). Contaminant uptake occurs by diffusion, whereby previous experiments established independence of the sampling rate over a wide range of wind speeds (18). Measurements of sorption onto XAD-2 resin and calibration experiments confirmed that most semivolatile organic compounds do not reach equilibrium between the atmospheric gas-phase and the resin (18). XAD-2 was cleaned by Soxhlet extraction, and

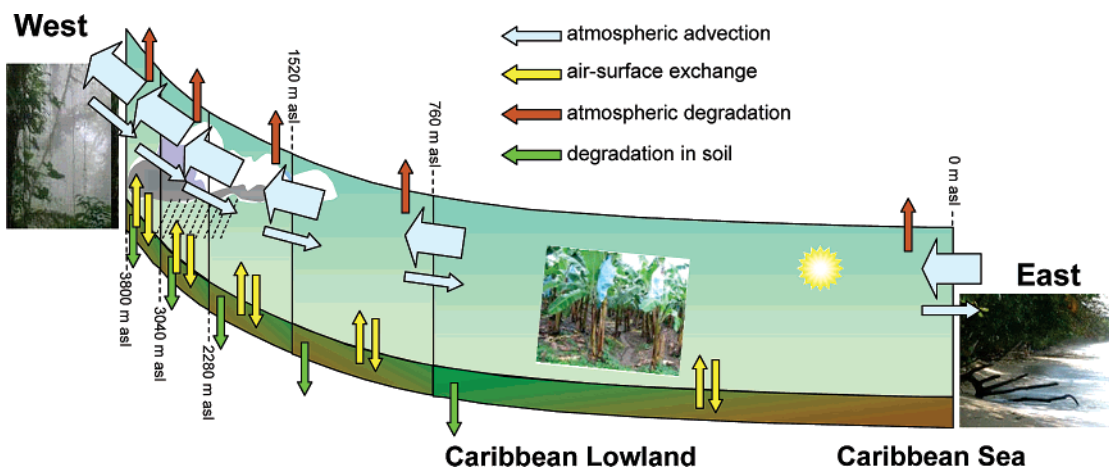


FIGURE 1. Schematic representation of the mountain-POP model in a tropical area. Arrows indicate various contaminant fate processes.

mesh cylinders were filled in a dedicated clean room with high efficiency particulate air (HEPA) and carbon-filtered air supply at Environment Canada. The cylinders were stored and transported in airtight aluminum tubes. Duplicate samplers were installed in February 2004 at approximately 1.5 m above the ground, retrieved a year later, and stored frozen until analysis. Field blanks were transported to the sites and taped close to the sampler for the entire year of deployment.

In the vicinity of 20 air samplers, 10 individual soil samples were taken in February 2004 in a grid, penetrating with an auger to a depth of approximately 25 cm. These samples were mixed with a clean steel shovel in a steel bucket. Two subsamples were wrapped in pre-cleaned aluminum foil, sealed in plastic bags, and stored frozen until analysis.

Chemical Analysis. The XAD resin was extracted and analyzed for endosulfan isomers I and II and degradation product endosulfan sulfate (referred together as Σ Endo) as has been described previously (18). The conditions used for gas chromatography (GC)-electron capture detection and the quality assurance steps, including treatment of procedure, resin, and field blanks, have also been detailed before (18). The XAD extracts were separately analyzed for chlorothalonil and dacthal on an Agilent 6890 GC equipped with an Agilent 5973 mass selective detector (MS) and a DB-5MS column (J&W Scientific; 60 m \times 0.25 mm i.d., 0.10 μ m film thickness). Analysis was completed by negative chemical ionization in selected ion monitoring mode. The temperature program was 90 $^{\circ}$ C for 1 min, 20 $^{\circ}$ C min $^{-1}$ to 160 $^{\circ}$ C, 2 $^{\circ}$ C min $^{-1}$ to 255 $^{\circ}$ C, and 20 $^{\circ}$ C min $^{-1}$ to 270 $^{\circ}$ C and held for 5 min. Ions 266 and 264 were monitored for chlorothalonil and ions 332 and 330 for dacthal. Detection limits and the coefficient of variation between duplicates are given in Table S2.

Aliquots (10–15 g) of wet soil, which had not been exposed to laboratory air, were mixed with sodium sulfate and ground to a granular consistency. The samples were Soxhlet extracted with dichloromethane for 20 h, concentrated, transferred to iso-octane by rotary evaporation and nitrogen blow-down, and purified on 3 g alumina columns. The columns were eluted with 45 mL of 1:1 dichloromethane/petroleum ether. The sample was reduced in volume to 1 mL, and 13 C-PCB-32 was added as an internal standard. A blank sample was completed with every five samples by following the same extraction steps for sodium sulfate alone. The soil extracts were analyzed by GC-MS with conditions as described above for chlorothalonil/dacthal. Ions monitored were 404, 406 for endosulfan-I and -II and 386, 388 for endosulfan sulfate. Concentrations in air and soil are blank corrected and reported as the average of two samples for air and two to four samples for soil. Field and lab blanks were low, and detection limits are given in Table S2.

Soil Characterization. Soil water content was determined by drying an aliquot of soil at 75–80 $^{\circ}$ C until constant weight was achieved. Moisture contents varied widely from 1 to 78%, with a mean of 32%. Soil concentrations are reported per gram dry weight. Dry soil was ground to a fine powder, and the total carbon content was determined by an elemental analyzer (2400 Series II CHNS). A titrimetric method was used to determine the inorganic carbon content (19). After subtracting inorganic carbon from total carbon, the organic carbon content ranged from 0.4 to 19% (Table S1).

Airshed Calculation. Five-day back trajectories were calculated for 7 stations across Costa Rica at 10, 100, and 200 m above ground level at 6 h intervals for each day the passive air samplers were deployed using the Canadian Meteorological Centre Trajectory Model. This information was used to produce back trajectory probability maps, referred to as “airsheds”, which identify where the air parcels are most frequently originating. The probability density is calculated as the number of trajectories per grid divided by the total number of trajectories (4380). All seven stations displayed the same pattern of primarily easterly winds (Figure S1).

Fate Modeling. Results from the field samples revealed elevated soil concentrations in mountains downwind from agricultural regions in the Caribbean lowland. To study the transport and accumulation of pesticides along a tropical elevation gradient quantitatively, a dynamic, ten-compartment, fugacity-based fate and transport model (level IV) was developed and used in an evaluative fashion. The model consists of five pairs of air and soil compartments that correspond to different altitudinal zones, each spanning 760 m in elevation, on the northeastern slope of the Costa Rican Cordillera Central (Figure 1). Clean air from the Caribbean Sea enters the model domain from the east in the lowest air compartment and is advected gradually to higher elevations, to eventually leave the model region at the crest of the mountain range. Some air mass movement in the opposite direction leads to atmospheric mixing. Bidirectional exchange between the air and soil compartments of each elevational zone occurs by diffusive gas exchange, precipitation scavenging, and dry particle deposition and is parameterized as in the CoZMo-POP model (20). Chemical can be degraded by reaction with OH radicals in the atmospheric compartments and by unspecified pseudo-first-order reactions in soil. No other loss processes from soil are considered.

The model was parameterized for the environmental conditions of Caribbean Costa Rica using information from refs 16 and 21 (Tables S3 and S4 and Figure S3). In particular, actual dimensions were used as well as monthly temperature and precipitation values. The size of the altitudinal zones decreases from the lowlands to the mountain crest, resulting

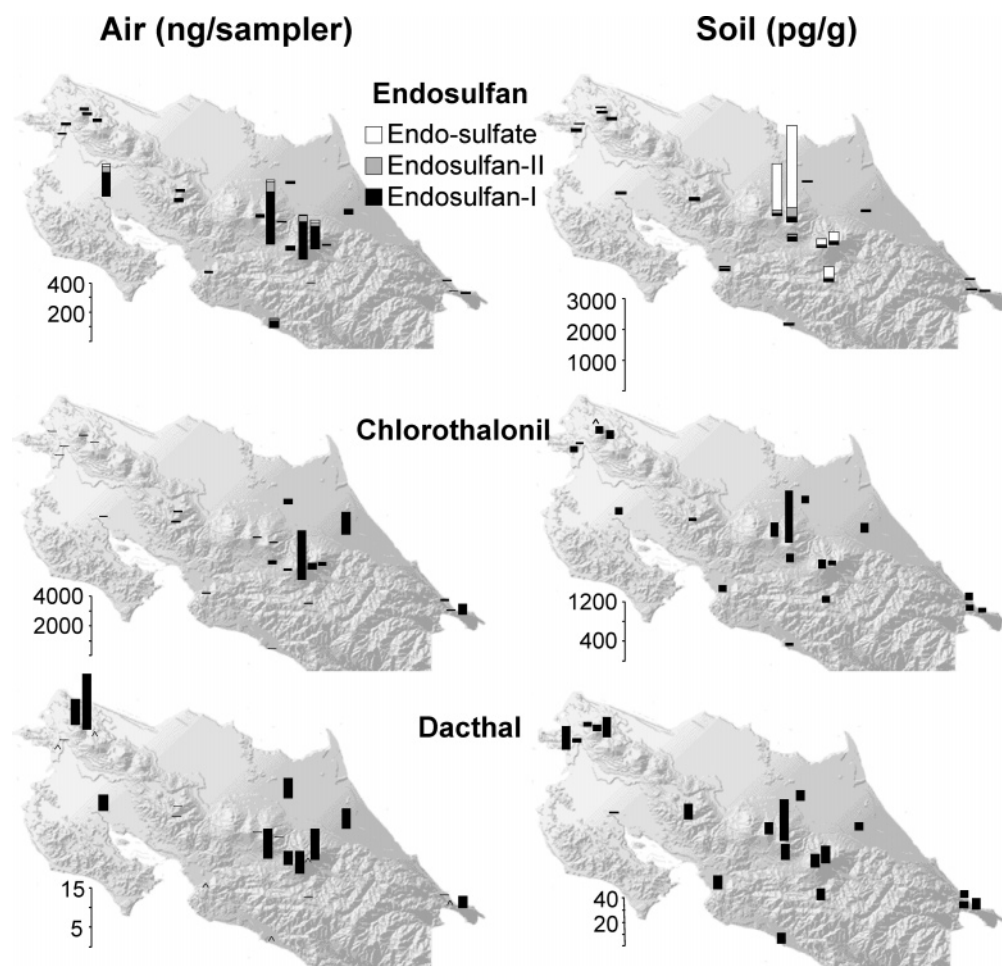


FIGURE 2. Air (ng/sampler) and soil (pg/g) concentrations of endosulfan-related compounds, chlorothalonil and dacthal, across Costa Rica (map reprinted with permission from ref 17).

in greatly decreased atmospheric residence times at higher altitude (ranging from 38 h to 1 h). The mean annual temperature gradient ranges from 26 °C in the lowland to 9 °C in the highest zone. While there is a clear decrease in temperature with rising elevation, the precipitation pattern is more complex (Figure S3). Precipitation is seasonally variable and is highest in the second highest zone, with a mean annual precipitation of 264 mm. Tropical OH concentrations (22) are given in Table S3. Soil in the model was assumed to have a constant mass fraction of organic carbon in solids of 2%.

To quantify pesticide accumulation at higher elevations in downwind mountains, a mountain contamination potential (MCP) was defined as

$$\text{MCP in \%} = M_{4+5}/M_{\text{tot}} \cdot 100\% \quad (1)$$

where M_{4+5} and M_{tot} represent the mass of chemical in air and soil in the two topmost elevational zones and in the total model domain, respectively. The MCP thus quantifies the fraction of the total amount in the model domain that is present in the region above 2280 m. Reflective of the location of agricultural activity in the region, emission is assumed to occur continuously for 1 year into the air compartment of the lowest zone. The MCP was calculated for perfectly persistent hypothetical chemicals with dimensionless air–water partition coefficients K_{AW} ranging from 10^{-2} to 10^{-6} and octanol–air partition coefficients K_{OA} ranging from 10^6 to 10^{12} . Enthalpies of air–water (ΔH_{AW}) and octanol–air phase transfer for these substances were 60 and $-80 \text{ kJ}\cdot\text{mol}^{-1}$, respectively. A sensitivity analysis assessed how different

environmental and physical-chemical properties affect contaminant accumulation at high altitudes. Specifically, ΔH_{AW} was varied, the effect of increased air mixing was explored, and degradability in soil and atmosphere was introduced.

Results and Discussion

The measured air and soil concentrations for endosulfan-I, -II, endosulfan-sulfate, chlorothalonil, and dacthal are listed in Table S5 in the Supporting Information. Air concentrations are reported in ng/sampler as well as pg/m^3 , generated by dividing the sampler concentration by the product of the deployment period (365 days) and the sampling rate ($\text{m}^3\cdot\text{d}^{-1}\cdot\text{sampler}^{-1}$) (18). Structures of the analytes are given in Figure S4. The spatial distribution maps in Figure 2 show that endosulfan and chlorothalonil were ubiquitous across the country, with very high concentrations found at several sites. Dacthal was only present at low levels in soil and was not detected in air at six of the sites.

Use and Distribution of Pesticides Across Costa Rica. Endosulfan is increasingly being used in Costa Rica as an insecticide on pineapple, rice, ornamental plants, vegetable crops, and fruit (23, 24). Approximately 40 tons were imported annually between 2000 and 2004 (23). Endosulfan concentrations in air and soil were up to 6 times greater than pp'-DDE levels, the banned organochlorine pesticide that had the highest concentrations among those quantified in the same samples (25). In a network of 40 passive air sampling stations, we have previously found endosulfan to be abundant and ubiquitous across North America, but the maximum sampler concentration reported here (430 ng endosulfan I+II per sampler) corresponds to an annually averaged air concen-

tration in excess of 1 ng/m³ and greatly surpasses the maximum endosulfan levels found in any of the North American samples (150 ng per sampler) (26). Across Costa Rica, the spatial distribution of endosulfan was distinctly different in air and soil. Highest air levels were found in Costa Rica's densely populated central valley (sites #18, #20, and #21) and in Palo Verde National Park (#9) (Figure 2), which is close to rice growing areas. The five highest soil concentrations of ΣEndo (>250 pg/g) occurred at the five sites with elevations above 2500 m (Prusia #21, Irazu #22, Tapanti #23, Barva #1, and Poas #12). In particular, samples taken at high elevations on volcanoes Barva (>3000 pg/g) and Poas (>1600 pg/g) had concentrations greatly elevated over the national average. The same five soil samples also stand out by having a greater abundance of the degradation product endosulfan sulfate than the parent endosulfan. The rate of endosulfan biodegradation has been shown to increase with endosulfan concentration (27).

Chlorothalonil is widely and increasingly applied in Costa Rica as a fungicide in banana (28), coffee, vegetable (potatoes, tomatoes, onions), and fruit (melons, pineapples) production (23, 24). Annual mean importation of chlorothalonil between 2000 and 2004 exceeded 700 tons, making it one of the most heavily used pesticides in the country (28). Very high levels of chlorothalonil (>1000 ng/PAS) were found in air samples, particularly at EARTH #14 and Cot #20, which are located in banana and vegetable growing areas, respectively. Chlorothalonil has been shown to volatilize readily from application areas, even under temperate climate conditions (29). Its ability to undergo atmospheric transport has previously been discussed (30). Again, the spatial distribution of chlorothalonil is different in air and soil. Like endosulfan, it is the two soils from volcanoes Barva and Poas that had the highest levels of chlorothalonil (Figure 2), with concentrations above 1000 and 275 pg/g, respectively. The next highest soil concentration (~200 pg/g) was measured at EARTH (#14).

In agreement with insignificant importation and usage (23), dacthal (otherwise known as chlorthal-dimethyl or DCPA) levels in Costa Rican air were low, on average 3 ng per sampler. Soil concentrations were also low, about an order of magnitude less than those of chlorothalonil and endosulfan. Dacthal was fairly evenly distributed in air and soil, although yet again the Barva site had by far the highest soil concentration.

Contaminant Amplification Along a Tropical Elevation Gradient. The following explains the widespread occurrence of chlorothalonil and endosulfan in Costa Rican air and soil, including remote sites at high altitudes: (i) plantations located in the Caribbean lowland (Figure S2) use pesticides on a large scale, (ii) back trajectory analysis shows predominantly easterly winds (Figure S1), and (iii) rain is abundant year-round from the lowlands to the highlands. The picture that emerges is that of pesticides traveling along with the trade winds from the plantations in the east to the volcanoes in Central Costa Rica. Forced atmospheric uplift leads to the formation of orographic clouds and precipitation, thus delivering the pesticides from the atmosphere to the soil. This picture however does not yet fully explain why soil concentrations at high altitude sites downwind of the plantations are higher than close to the plantations. An intriguing issue is therefore the nature of the mechanism by which the pesticides become enriched upslope.

The air-water partition coefficients K_{AW} of endosulfan, dacthal, and chlorothalonil at 25 °C are $10^{-3.5}$, 10^{-4} (30), and 10^{-5} (31), respectively. This is the K_{AW} range where precipitation scavenging is starting to become important, and where a drop in temperature (causing a drop in K_{AW}) is thus expected to greatly increase the efficiency of rain scavenging (32). We thus hypothesize that pesticides that are only slightly

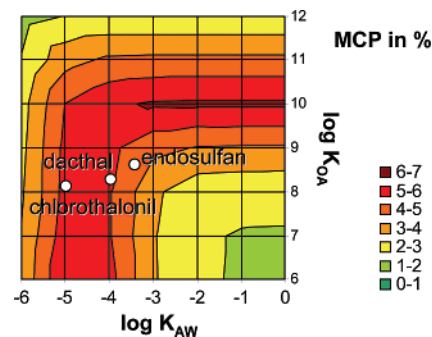


FIGURE 3. Mountain contamination potential MCP in % of a hypothetical, perfectly persistent organic chemical emitted continuously for 1 year to the atmosphere of the lowest air compartment of the model in Figure 1 as a function of its partitioning properties K_{AW} and K_{OA} .

scavenged at the temperature prevalent in the Caribbean lowland could be subject to much more efficient scavenging at the lower temperatures occurring at higher altitudes. The temperature dependence of rain scavenging could thus explain why higher soil concentrations occur at greater distance from application areas and, in particular, in high altitude cloud forests. Furthermore, all three of the investigated pesticides are amphiphilic, which raises the possibility of strong enrichment in cloud and fog droplets as a result of adsorption to the water surface (33).

Furthermore, once deposited to vegetation and soils, the pesticides are more likely to be retained at high altitudes. A lower temperature, a high organic matter content (Table S1), and a dense vegetation cover will retard revolatilization, and frequent cloud cover limits direct solar radiation and therefore photolytic degradation. Microbial degradation can occur under the moist conditions of montane forests, as is clear from the high abundance of endosulfan sulfate in high altitude soils (Table S5). However, in the case of endosulfan and chlorothalonil, the major degradation products (endosulfan sulfate and 4-hydroxy-2,5,6-trichloroisophthalonitrile) are more persistent and as, if not more than, toxic as the parent compound (34). Dacthal readily hydrolyzes in soil (35), which may contribute to its lower soil concentrations.

Model-Based Assessment of Mountain Contamination Potential. The measured spatial distribution of pesticide residues in air and soil suggests that high concentrations in high altitude soils are the result of atmospheric transport from the sites of pesticide application, efficient deposition by orographic rain and fog, and strong retention in cool soils that are rich in organic matter and covered by dense vegetation. Evaluative model calculations were aimed at testing this hypothesis and providing further insight into the mechanism of contaminant amplification. The calculated Mountain Contamination Potential, MCP, for perfectly persistent, hypothetical chemicals is plotted as a function of K_{AW} and K_{OA} at 25 °C in Figure 3. The chemical space map shows that the MCP is elevated for chemicals with K_{AW} values between 10^{-3} and 10^{-5} and for those with a K_{OA} between $10^{8.5}$ and 10^{11} . Largest enrichment at higher elevations is predicted for chemicals with a K_{AW} of around $10^{-4.2}$ and/or a K_{OA} of approximately 10^{10} . In both cases, these ranges of partition coefficients refer to substances that experience a temperature-driven change in atmospheric distribution behavior that results in inefficient precipitation scavenging in the tropical lowlands and efficient scavenging at higher elevations (32). In the case of K_{AW} it is a change between atmospheric gas phase and rain droplets, and in the case of K_{OA} it is a change between the atmospheric gas and particle phase.

Chemicals with a K_{AW} at 25 °C greater than 10^{-3} experience low precipitation scavenging at all temperatures encountered

in Costa Rica and will therefore not be deposited efficiently with rain even at higher altitudes, limiting the MCP value that can be attained. Chemicals with a K_{AW} at 25 °C less than 10^{-5} are very strongly scavenged by precipitation even at high temperatures and, since it rains in the lowlands, will be deposited there without the chance to accumulate at higher elevations. Finally, the chemicals with a K_{AW} at 25 °C in the 10^{-3} – 10^{-5} range are scavenged less efficiently at the temperatures prevalent in the lowland than at the colder temperatures at high elevations, allowing for enrichment in mountain soils. Accordingly, the MCP shows a maximum for such substances.

In an analogous mechanism, relatively volatile substances with a K_{OA} at 25 °C less than 10^8 remain in the gas phase even at the low temperatures of tropical mountains and thus are only scavenged efficiently by rain if they also have a K_{AW} less than 10^{-3} . Substances with a K_{OA} above 10^{11} tend to be particle-bound even at the temperature of the Caribbean lowland and therefore are rained out close to application areas. Substances with a K_{OA} at 25 °C between $10^{8.5}$ and 10^{11} shift their distribution from the gas to the particle phase during the temperature drop encountered by an air mass that is being uplifted orographically on the central Cordillera and thus will experience increasing rain scavenging efficiency with increasing elevation. Locating the partitioning properties of dacthal, chlorothalonil, and endosulfan at 25 °C on the chemical space of Figure 3 reveals that the air–water partition coefficient of all three pesticides falls into the K_{AW} range of elevated MCP. For example, the K_{AW} of endosulfan is predicted to decrease from $10^{-3.5}$ to $10^{-4.1}$ between the 26 °C prevalent in the lowlands and the 9 °C at the top of the mountain. This suggests that an increase in the precipitation scavenging of their vapors is responsible for the high soil concentrations measured on volcanoes Barva and Poas. The three pesticides are too volatile for the K_{OA} -controlled mechanism to play a major role in their accumulation at higher tropical altitudes.

Because of the dominance of the K_{AW} -controlled process for currently used pesticides, sensitivity analyses were conducted for hypothetical chemicals with a fixed K_{OA} (10^8) but a variable K_{AW} (Figure S5). The MCP increases with a higher enthalpy of air–water phase transfer ΔH_{AW} (Figure S5A). The ΔH_{AW} determines how strongly a K_{AW} depends on temperature and a higher ΔH_{AW} results in a greater difference between the scavenging efficiency at low and high elevations and thus a higher MCP. Increasing downslope air flow, i.e., enhancing the extent of atmospheric mixing along the elevation gradient, lowers the MCP (Figure S5A). Enhanced atmospheric mixing means less unidirectional upslope air flow and therefore less chemical staying at high elevations. To evaluate the impact of pesticide degradability, the hypothetical chemicals were assumed to degrade with reaction half-lives in the range applicable to chlorothalonil and endosulfan. Chlorothalonil has an estimated soil half-life on the order of 3 years (36), while that of other currently used pesticides ranges from a few months to more than a decade (30). Introducing a soil half-life of 3 years has little impact on the MCP in the K_{AW} range of interest, while a soil half-life of 3 months results in an elevated MCP (Figure S5B). A shorter soil half-life will reduce the total amount of chemical in the environment, but it can result in a higher relative enrichment (37). Since degradation rates are temperature dependent and thus higher in the lowlands, the more significant the process of soil degradation the greater the enrichment of contaminant at high elevations. Muir et al. (30) estimated gas-phase reaction rates of a range of pesticides with OH radicals k_{OH} between $4.4 \cdot 10^{-13}$ and $1.3 \cdot 10^{-10}$ $\text{cm}^3 \text{molec}^{-1} \text{s}^{-1}$. Endosulfan and chlorothalonil have estimated k_{OH} values of $8.2 \cdot 10^{-12}$ (30) and $6.2 \cdot 10^{-15}$ $\text{cm}^3 \text{molec}^{-1} \text{s}^{-1}$, respectively (36), suggesting a very high persistence of the latter in air. A k_{OH} of 10^{-12} causes a slight reduction in the

MCP (Figure S5B). Some of the chemical will degrade at lower elevations and never make it upslope. Overall, the calculations suggest that while reactivity in air decreases the MCP, reactivity in soil increases it. Adopting the measured soil organic carbon contents across the zones instead of a uniform content of 2% made no difference to the calculated MCP.

Implications of Pesticide Enrichment in High Altitude Tropical Forests. Clearly, chlorothalonil and endosulfan sulfate have the potential to accumulate in soils at high altitudes in the humid tropics. It is also established that chlorothalonil, endosulfan, and endosulfan sulfate are highly toxic to fish and other aquatic organisms (34, 38). Not only the pesticides quantified in air and soil in this study, but also many other pesticides in current use have a K_{AW} between 10^{-3} and 10^{-5} and a K_{OA} around 10^8 – $10^{9.5}$ (30) and thus might be subject to the identified contaminant amplification mechanism operating along tropical elevation gradients. Among the other pesticides used in Costa Rica that, based on their partitioning properties, fall into the range of high MCP are the organophosphate insecticides diazinon and chlorpyrifos (K_{AW} of $10^{-5.1}$ and $10^{-3.6}$, ref 30). A recent toxicity assessment in Costa Rica showed that when considering both usage and toxicity, diazinon and chlorothalonil are among the top five pesticides which cause 75% of the total aquatic ecotoxicity in the country (39).

There is growing evidence that neotropical amphibian populations, and those inhabiting montane areas in particular, are declining (12, 40, 41). In one Costa Rican study, unusual female-biased amphibian sex ratios pointed toward estrogen-disrupting contaminants as a possible source of influence (41). Precipitous declines in amphibian populations in the mountains of California have been linked to pesticide deposition (13, 42). Davidson (42) noted a correlation between organophosphate pesticide use and declines in amphibian populations. Even though pathogens in combination with global warming have recently been implicated in the decline of amphibian populations in the neotropics (43), this does not exclude a potential role of contaminants in this disturbing phenomenon. In particular, our study provides an alternative explanation for the puzzling observation that tropical amphibian population declines are more prevalent at higher altitudes, which are often further removed from human encroachment.

Both field and modeling work have demonstrated the potential of tropical mountains to accumulate pesticides currently used in neighboring lowlands. Montane cloud forests downwind of major application areas appear particularly vulnerable to inflowing atmospheric contamination. It is clearly imperative to quantify the transport and deposition of pesticides to these forests and assess their impact on the local ecosystems.

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Supporting Information Available

Additional tables and figures. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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