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Review: mountain lakes as freshwater resources at risk from chemical pollution

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Abstract

Background Chemical pollution forms a severe threat for human and environmental health. While the risks for European lowland water bodies are well known, there is little knowledge on remote aquatic ecosystems and particularly mountain lakes, despite their importance for the provision of freshwater. Here, we critically review the current knowledge on the exposure and risk by chemical pollution for mountain lakes and present a tiered approach on how to advance effectively our understanding in the future.

Results Generally, pollutant monitoring data are currently incomplete, with many regions and substances having been only poorly investigated. More reliable data exist only for persistent organic pollutants (POPs). However, there is increasing evidence that even remote mountain lakes are exposed to a wide range of organic pollutants. Among them potent pesticides currently used in agricultural and biocidal applications, such as diazinon and permethrin. The exposure of mountain lakes to pollutants follows a complex pattern. Pollutants are introduced into mountain lakes via the atmospheric deposition and run-off from the watershed, but also local sources, like tourism and pastoralism. Our risk assessment and recent biomonitoring studies suggest that there are widespread chronic toxic risks on crustacean in mountain ranges. If mountain ranges are exposed to tourism and pastoralism, even acute toxic effects on crustacean are possible. Thereby, the vulnerability of mountain lakes to toxic effects has to be expected to be particularly high due to the harsh environmental conditions at high altitudes, the organism's traits, the insular position of mountain lakes and a lower species richness with increasing altitudes. Furthermore, there is little knowledge on the biological processes leading to the degradation of chemical pollutants under the environmental and ecological conditions of mountain ecosystems.

Conclusion While the exposure and sensitivity of mountain aquatic ecosystems is currently poorly investigated, the existing data suggest that it is very likely that also water bodies as remote as mountain lakes do suffer from pollution-induced toxicity. To verify this suggestion and expand the existing knowledge, it is necessary that future studies combine a more holistic pollution monitoring with exposure modelling and links to biological effects. Only then will it be possible to obtain a more reliable understanding of the impact of chemical pollution on aquatic mountain ecosystems and to protect these fragile ecosystems.

Keywords Organic micropollutants, Organic chemicals, Mountain lakes, Risk assessment, Zooplankton, Biodiversity

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Introduction

Human activities are degrading ecosystems to an extent that they become dysfunctional and lose their capacity to provide ecosystem services [1–3]. One of the most important ecosystem services is the provision of drinking water. Freshwater provision is also at the basis of numerous other ecosystem services, such as agricultural production, tourism, and the production of construction material. Mountains are the water towers of the world and provide freshwater to more than half of the world's population [4]. Protecting the health of mountain catchments is thus essential to safeguard the provision of high-quality freshwater and human well-being. A central characteristic of many mountain catchments are lakes. Mountain lakes are present in most mountain ranges, where they act as freshwater storage basins and are generally very biodiversity rich in comparison to terrestrial habitats [5]. They are, however, highly sensitive to anthropogenic impacts like chemical and nutrient pollution, climate change and habitat destruction, which in synergy put them at risk to become dysfunctional and to degrade their ecological state [5–7].

At low altitudes, the pollutants emitted by agricultural, urban, and industrial activities are known to degrade the ecological state of the aquatic environment [8, 9]. With increasing altitude, these sources are considered to become less, which should result in a limited exposure towards chemical pollution and hence no degradation of mountain water bodies by organic chemicals. However, at the example of the Arctic and Antarctic, it has been shown that atmospheric long-range transport and marine currents [10], as well as migrating species [11, 12], cause a global pollutant spread. As a result, organic chemicals, and persistent organic pollutants (POPs) in particular, have accumulated in arctic regions and impact wildlife and human health [13, 14]. In mountains, local economic activities, such as tourism [15], recreational angling [16], the exploitation of natural resources [17] and pastoralism [18, 19] have the potential to introduce or release pollutants into mountain lakes. Since the distance to source regions in many mountains is much shorter than in the arctic, also less persistent chemical pollutants (e.g., agricultural pesticides) enter mountain environments by atmospheric transport [20]. Furthermore, mountain lakes appear to be particularly vulnerable to chemical pollution, due to very harsh living conditions [6, 21] and the short and unbranched food webs that are characterized by specialized and sensitive top predators, such as amphibians, birds of prey, and introduced fish [21]. As a result, organic chemicals do not only lead to a widespread degradation of aquatic ecosystems at low altitudes [8, 9], but also in the mountain environment to a yet unknown extent.

Chemical pollution drives important changes in mountain lakes and disturbs their sensitive ecological equilibrium. With increasing impact and degradation, these ecosystems will become more and more dysfunctional, reducing their ability to provide the ecosystem service of clean drinking water. To mitigate this risk, it is essential to strengthen our knowledge on the exposure to and impacts by complex chemical mixtures on mountain lakes. Therefore, it is important to go beyond data-rich regions, like the lowland water bodies of Europe and North America, and collect more data on the impact of pollution on remote water bodies, like mountain ecosystems [22, 23].

Here, we synthesized the current knowledge on the exposure to and risk by chemical pollution in mountain lakes. This has been done along different lines of evidence including: (i) a toxic unit (TU) based assessment of mixture toxicity risks under the use of existing exposure monitoring data [24, 25]; (ii) the discussion of plausible pathways of exposure via atmospheric deposition and from local sources; (iii) the collection of evidence on toxic impacts in mountain lakes; and (iv) the discussion of habitat characteristics possibly enhancing the vulnerability of mountain lake ecosystems.

Mixture toxicity risks based on monitoring data

To assess the toxic risk for aquatic organisms in mountain lakes by chemical pollution, a dataset with water monitoring data was compiled, including 17 studies providing water concentration data (Table 1; Fig. 3). Additionally, one study could be found that offered surface sediment pollution data for 14 lakes in the Sierra Nevada Mountains (California, USA) and allowed for the calculation of equilibrium water concentrations, as also the total sediment organic carbon content had been determined (Additional file 1) [26]. However, water monitoring data on the contamination of mountain lakes with organic chemicals were scarce and scattered.

The obtained dataset includes data on a total of 36 different lakes of 11 different mountain ranges and countries, extending from the year 1994 to 2018. Most studies were performed in the Sierra Nevada Mountains, the Rocky Mountains, and the Pyrenees. Investigated were very small (0.1 ha) to very big lakes like Nam Co Lake in Tibet (201500 ha). Thereby, the presence of a total of 174 compounds was reported, including 16 legacy pesticides and their degradation products, 51 current-use pesticides (15 herbicides, 24 fungicides and 12 insecticides), 21 PAHs, 15 PCBs, 9 brominated diphenyl ethers (BDEs) that are used as flame retardants, 4 pharmaceuticals, 5 compounds commonly used in sunscreens and 53 compounds of various uses. In the most holistic monitoring study, passive sampling was combined with an extensive

Table 1 Overview of studies documenting water concentration data for mountain lakes from different parts of the world

Mountain range	Country	Publication	Sampling method[a]	Substances[b]	Lake	Sampling year	Altitude (m)	Size (ha)[c]
Sierra Nevada	US	Datta et al. [32]	180L SW from vessel in SST	PCBs	Tahoe	1995	1897	49620
					Marlette	1995	2500	55
		Noir et al. [33]	4L SW in glass	OCs	Crescent meadows	1997	2042	NA
					Tablelands	1997	3231	NA
					Sixty lakes basin	1997	3322	0.9
					Moro creek	1998	823	NA
		Bradford et al. [34]	100L direct SPE	OCs, CUPs	Frog	2003	3091	1
					E. Marjori	2003	3550	4
					Gorge of despair	2003	3042	2
					Wright	2003	3645	17
		Bradford et al. [26]	Hand corer at 1 m depth (sediment)	OCs, CUPs, PCBs, PAHs	60-lake	2005	3225	0.2
					9-lakes	2005	3189	6
					Bench	2005	3299	3
					Beville	2005	2786	0.1
					Blue Cyn	2005	3241	0.2
					Forgotten	2005	3262	0.7
					Gorge	2005	3188	0.4
					Laurel	2005	3177	0.5
					Observat	2005	3207	10
					Ouzel	2005	3338	0.2
					Palisades	2005	3260	0.1
					Tableland	2005	3243	0.1
					White chief	2005	3120	1
					Wright	2005	3375	0.8
Rocky M	Canada	Donald et al. [35]	40L, 5 m depth, centre of lake	Chlorobornane	Emerald	1994	1300	116
					Cabin	1994	1219	32
					Maligne	1994	1671	2066
		Blais et al. [36]	72L in SST	OCs	Annette	1994	1019	29
					Bow	1998	1940	280
		Wilkinson et al. [37]	70L SW in alu tank, sampled from shore	OCs	Bow	1998 1999 2000	1975	320[c]
					Kananaskis	2000	1667	780[c]
					Dixon Dam	2000	946	43030[§]
					Kinbasket	1999	770	1760[§]
Pyrenees	Spain	Vilanova et al. [38]	100L from several depths, direct SPE	OCs, PCBs	Redon	1996 1997 1998	2240	24
		Fernandez et al. [39]	100L several depths	OCs	Redon	2000 2001	2240	24
		Santolaria et al. [40]	5L SW, middle of lake	BDE, PAHs, OCs, PCBs	Sabocos	2011 2012 2014	1905	9.3
		Machate et al. [19]	Passive sampling	OCs, CUPs, PCBs, PAHs, BDEs, Personal care products, etc.	Ansabere	2018	1850	0.2

Table 1 (continued)

Mountain range	Country	Publication	Sampling method[a]	Substances[b]	Lake	Sampling year	Altitude (m)	Size (ha)[c]
					Acherito	2018	1880	6.0
					Puit	2018	1880	0.2
					Ayes	2018	1714	2.0
					Belonguere	2018	1907	0.1
					Coueyla Gran	2018	2159	0.5
					Madamete Bas	2018	2307	0.1
					Gourg de Rabas	2018	2400	1.0
Alps	Austria	Vilanova et al. [38]	100L from several depths, direct SPE	OCs, PCBs	Gossenkölle	1996	2417	2
	Austria				Gossenkölle	1997	2417	2
	France	Nellier et al. [41]	60L SW & DW	PCBs	Muzelle	2012 2013	2115	10
	France				Plan Vianney	2012 2013	2250	5
Tatra M	Slovakia	Fernandez et al. [39]	100L several depths	OCs	Ladove	2000	2057	2
Caledonia M	Norway	Vilanova et al. [38]	100L from several depths, direct SPE	OCs, PCBs	Øvre Neådalsvatn	1998	728	50
Southern Alps	New Zealand	Wu et al. [42]	800L subsurface water direct SPE	OCs	Brewster lake	2014 2015	1700	2
Nyenchen Tanglha M	Tibet	Ren et al. [43]	200L SW, direct SPE	OCs, PAHs, PCBs	Nam Co lake	2014	4718	201,500
		Ren et al. [44]	200L SW, direct SPE	OCs, PAHs, PCBs	Nam Co lake	2013	4718	201,500
Himalaya	Nepal	Galassi et al. [45]	5L	OCs, PCBs	Lake Inferior	1994	5067	2
		Guzzella et al. [46]	2L SW in glass container	OCs, PAHs, PCBs	Lobuche 152	2007	4893	NA
					Lobuche 14	2007	4893	NA
					Lobuche 12/13	2007	4968	57[c]
					Lake Pyramid 10	2007	5053	2[c]
					Lake Pyramid 9	2007	5215	0.6[c]
					Kalapattar 7	2007	5293	0.2[c]
Volcanoe Poas	Costa Rica	Shunthirasing-ham et al. [47]	10-20L SW in SST	OCs	Laguna Botos	2009	2580	10[c]
Volcanoe Barva					Laguna Barva	2009	2840	1[c]

SW surface water; *DW* deep water; *SST* stainless steel tank; *SPE* solid phase extraction: *OCs* organochlorine pesticides; *CUP* current-use pesticide; *PAH* polyaromatic hydrocarbon; *PCB* polychlorinated biphenyl; *NA* Not Assessed lakes could not be identified in google image

[a] Shortforms

[b] Shortforms

[c] Size of lake was estimated via google maps

target screening of 479 substances, including chemicals potentially introduced via local sources and atmospheric deposition (Table 1, Machate et al. [19]). Most other studies focused only on POPs. The best investigated substances, which have been investigated in more than 6 different studies and mountain ranges, are hexachlorobenzene (HCB), alpha-endosulfan, beta-endosulfan and endosulfan sulfate, as well as alpha and gamma-hexachlorocyclohexane (HCH). Only 16 substances have been investigated in more than two mountain ranges. This includes six PCBs, nine legacy pesticides and their metabolites, as well as one current-use pesticide (chlorpyrifos).

The available water pollution concentration data were used to estimate the existing toxic risk for algae and crustacea, two organisms of central importance in mountain lake food webs [6]. Toxic risks were assessed by calculating the individual TU for each compound at each site and finally these individual TUs per site were added together to obtain a cumulative TU, representing the mixture toxic risk at a given site at a certain time point based on the compounds analysed (Eq. 1) [19, 24, 25]:

$$\sum TU = \sum \frac{c_{i,water}}{EC_{xi}}, \tag{1}$$

where $c_{i,water}$ is the water concentration of the analyte i and EC_x is its effect concentration (EC) at level x. ECs were calculated as the 0.05 quantiles (Q 0.05) of the effect levels that could be obtained from the US EPA ECOTOX Knowledgebase (release version 09 December 2019) for exposure times of up to 120 h and including data on no observable effect levels (NOEL), the lowest effect levels (LOEL), and EC10 up to EC90 for algae and crustacea [27, 28]. If no measured entry was available, the EC50-value was estimated by using the baseline ECOSAR 1.0 type model [29, 30] in ChemProp v6.7.1 [31]. For this the baseline ECOSAR-type QSAR was used, whereat the QSAR was group-specific [28]. All ECs used during the assessment and their origin can be found in AF sheet 4. To assess the likelihood of adverse effects and display the risk that has been caused by the individual compounds, TUs and ΣTUs were compared with thresholds for acute (0.1 TUs for both organisms) and chronic (0.02 TUs for algae; 0.001 TUs for crustacea) toxic effects [8].

In a comprehensive investigation of a broad range of chemicals in Pyrenean lakes [19] mixture risks of 1×10^{-3} to 0.01 ∑TUs were reported for algae. Thus, maximum mixture toxic risk approaches about 50% of the threshold assumed for chronic toxicity (0.02 TUs). Considering uncertainty, due to non-analysed mixture components, temporal deviations and uncertainties related to sampling and analysis, a factor of 2 is a tiny buffer even if the findings do not support a chronic risk based on the analytical results. For crustacea, TUs ranged from 1×10^{-3} to 0.25 ∑TUs, thus exceeding the thresholds for acute (0.1 TUs) and chronic toxic risks (0.001 TUs) in 25% and 100% of their samples. In this study, the drivers of toxicity towards crustacea were the insecticides diazinon, and permethrin. For the remaining studies, which only assessed relatively small sets of pollutants, the mixture toxic risk ranged from 8.9×10^{-8} to 0.08 ∑TUs for algae and 3.4×10^{-7} to 9.5 ∑TUs for crustacea with a median of 6.7×10^{-6} ∑TUs and 1.4×10^{-4} ∑TUs, respectively (Fig. 1; AF sheet 4). The thresholds for acute and chronic toxic risks to crustacea were breached in 3 and 29% of the samples. Based on their median TUs, the main drivers of toxicity towards crustacea are the insecticides diazinon, malathion and chlorpyrifos with 3.4 TUs (detections $n=4$), 0.16 TUs ($n=1$) and 0.03 TUs ($n=34$), respectively. Also, the flame-retardant BDE 209 reveals a relatively high median of 0.03 TUs ($n=4$). The calculated TUs for algae never exceeded the thresholds for acute toxic risks, and only in 2% of the samples the thresholds for chronic toxic risks were exceeded. The highest median toxicity was caused by the flame-retardant BDE-209 with 0.03 TUs ($n=4$). In individual cases, also the two herbicides triallate and trifluralin contributed slightly higher amounts of TUs.

In the following paragraphs, the here obtained risk information will be compared towards the results of the currently most comprehensive risk assessment on the risk for mountain lakes by organic chemicals [19] and discussed in the context of the currently available knowledge on organic chemicals in mountain lakes.

Atmospheric deposition causes chronic toxic risks

The concept of organic chemicals being effectively transferred from metropolitan, industrial and agricultural areas into the mountain environment via atmospheric long- and medium-range transport is well established [20, 21]. The wet and gaseous deposition of more volatile compounds, as well as the deposition of particles carrying chemicals of little volatility (e.g., current-use pesticides), can lead to a direct introduction of organic chemicals from the atmosphere into mountain lakes (Fig. 2) [48]. As a result, mountain aquatic ecosystems closer to urbanized regions are exposed to a wide variety of POPs and PAHs, current-use agricultural pesticides, musks, and various other chemicals [19, 40, 49]. It has been described that, due to the colder temperatures at higher altitude, precipitation acts as an effective pump for water-soluble compounds from the atmosphere into mountain lakes and their catchment [50]. The water–air partitioning coefficient (K_{wa}) is a measure of how easily a compound can disperse between air and water and, therefore, influences the tendency of a compound to undergo wet deposition. Chemicals with a K_{wa} between 3.5 and 4 have the highest potential to be scavenged by precipitation and thus the highest mountain contamination potential [51, 52]. These chemicals are sufficiently volatile to reach high altitudes, as they are not efficiently scavenged by precipitation at moderate temperatures at lower altitudes, while at the same time not being too volatile to not being scavenged at colder temperatures at high altitudes. In addition to the direct deposition of organic chemicals from the atmosphere into the lakes, chemicals can also be deposited onto the surface of the catchment and consecutively be transported downslope into the next mountain lake [20]. This especially becomes effective when temperatures are cold enough that precipitation occurs in the form of snow. Snow possesses a scavenging efficiency that is higher for most compounds and especially for larger, non-polar organic compounds, than the one of rain [51, 53]. Furthermore, it allows for the accumulation of larger amounts of precipitation in form of snow and ice in mountain catchments, along with the

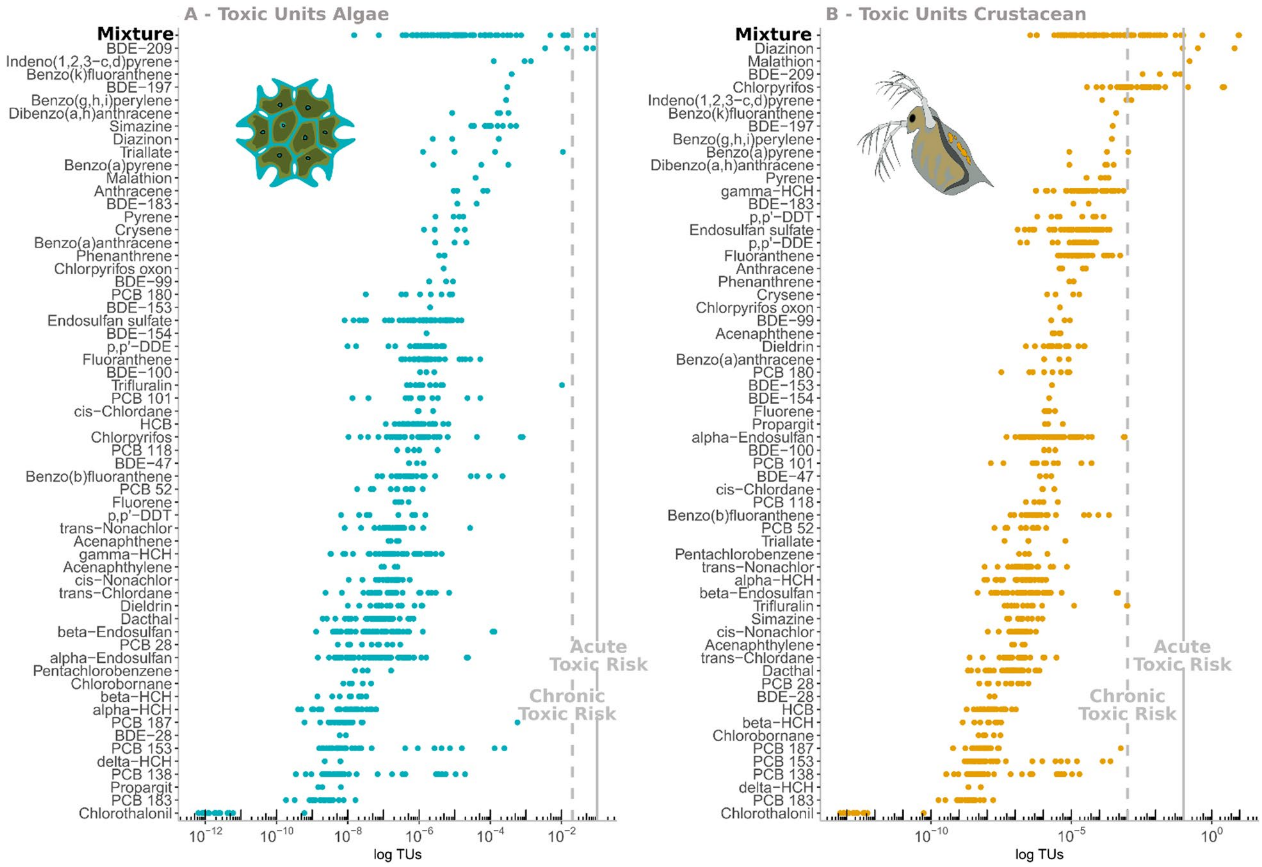

Fig. 1 Toxic risk by organic chemicals for crustaceans and algae in mountain lakes. TUs calculated from water concentrations stated in various studies (Table 1, except Machate et al. [19]) investigating the chemical pollution of mountain lakes from different parts of the world. "Mixture" displays the predicted mixture toxic risk of all compounds that have been measured at a given site (∑TUs). Below, the individual toxic risks caused by each individual compound is displayed (TU) and they were sorted according to their median TUs (increasing from bottom to top). Thresholds for chronic and acute toxic risk are displayed in the form of a dashed and solid grey line, respectively

pollutants associated to them [42, 50, 54]. This way snow leads to an increased transport and build-up of organic pollutants in the mountain environment. As these pollutants are then released into downstream lakes quicker than the time it took for them to build up—either during the melting of seasonal snowpacks [54–56] or the climate change-driven ice melt of glaciers [42, 57, 58]—this leads to a higher exposure and hence risk for mountain lakes by organic chemicals than during other precipitation events (e.g., rain and fog). For glaciers, for example, it has been shown that the organic chemicals contained in their meltwater can pose a toxic risk to aquatic invertebrates [59]. This can be assumed to occur in the same or even higher extent during mountain spring-season, when pollutants accumulated in seasonal snowpacks are released within a relatively short period of time and thus more rapidly than from glaciers, thereby endangering biodiversity.

Various factors influence how exposed a certain mountain lake is towards atmospheric pollutants, whereat the proximity towards pollution sources is the most important [50]. Since pollution levels in mountain lakes are closely related to those in the atmosphere [60] and air quality is worse in closer proximity to source regions [50], mountain slopes and lakes adjacent to regional source regions show considerably higher levels of pollution compared to slopes averted or remote to source regions [61–63]. In addition, also climatic parameters are of importance when trying to understand the exposure of mountain lakes. Generally, lowering temperatures and rising precipitation with increasing altitude (Fig. 2) favour the deposition and accumulation of chemical pollutants at higher elevations, which is suggesting increasing chemical concentrations towards the mountain summit. In the actual mountain environment, however, this pattern has not been found consistently [20]. Likely source proximity, the complex mountain terrain, which leads to an uneven distribution of precipitation and incoming air within a given mountain range [64, 65], together

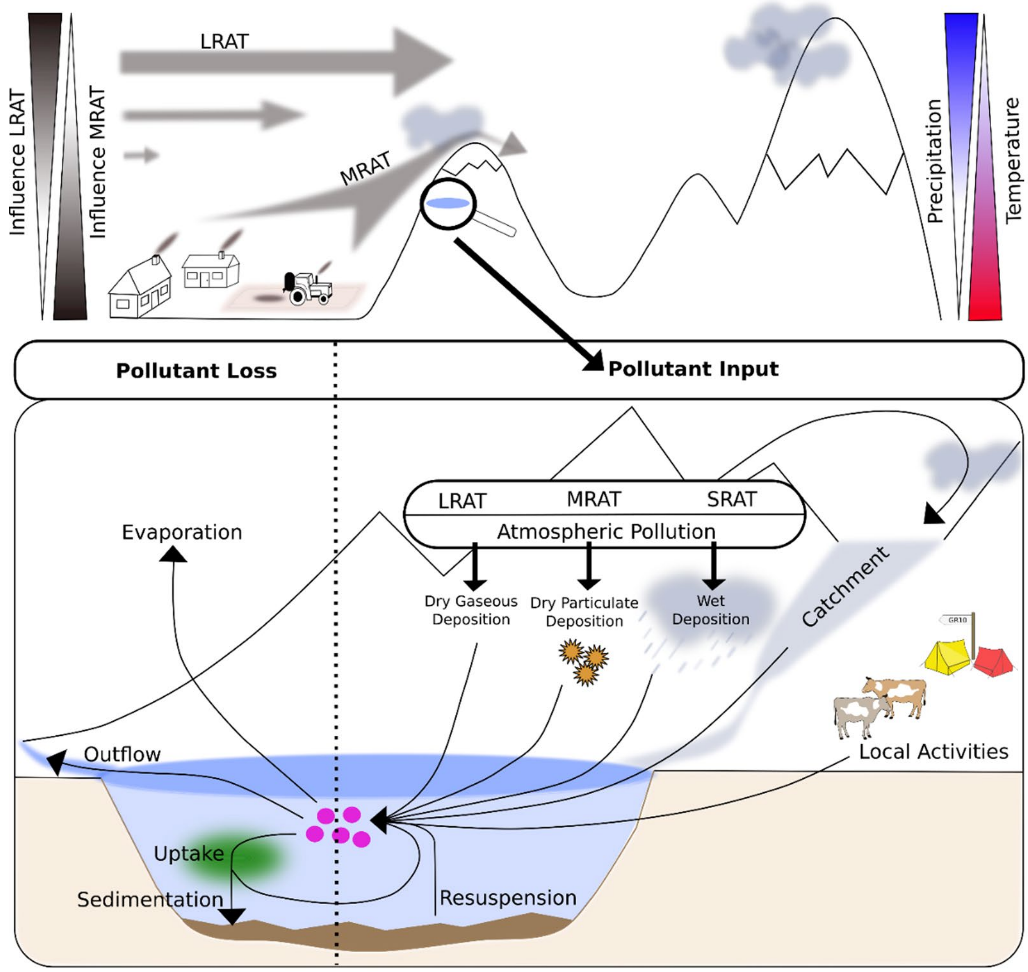

Fig. 2 Atmospheric transport of organic pollutants into mountain environments, and the input and loss of organic chemicals from the waters of mountain lakes. Included pollutants sources are atmospherically deposited pollutants that directly or indirectly introduced into mountain lakes, as well as pollutants from local activities like pastoralism and tourism. Not displayed are degradation processes or topographic and climatic features that can also affect pollutant transport and deposition, as well as their accumulation in lakes. *LRAT* long-range atmospheric transport; *MRAT* medium-range atmospheric transport; and *SRAT* short-range atmospheric transport (e.g., local wildfires)

with the compound and lake properties [20, 66], lead to complex and variable pollutant distribution. Current knowledge [67, 68] suggests that lower altitudes are more influenced by regional emissions (e.g., current-use pesticides) via regional air circulations that are facilitating medium-range atmospheric transport. Alpine regions, in contrast, are predominantly affected by long-range atmospheric transport that is introducing more persistent substances or particle bound compounds. Accordingly, in most cases mountain lakes at high altitudes, and/or located in mountains remote to urban activities, can be expected to be less polluted by atmospheric pollutants than mountain lakes at lower altitude and/or greater proximity to human activities [33, 69]. While this suggests a higher toxic risk for mountain lakes at lower altitudes, this also implies that even remote (alpine) mountain lakes are exposed to and potentially at risk by organic chemicals released at lower altitudes. In addition to the spatial differences, temporal fluctuations of atmospheric transport and pollutant levels are further complicating the pollution patterns in mountain areas. Seasonal shifts in climate and air circulation, as well as pollutant source activity, cause temporal variations in atmospheric pollutant levels and deposition processes [20]. Furthermore, seasonal and climate change-driven temperature fluctuations also cause events like the seasonal snowmelt and influence the properties of the lakes (e.g., no interaction with the atmosphere while being covered with ice). As a result, pollutant levels in mountain lakes fluctuate over time and are difficult to predict. To correctly evaluate the risk for aquatic habitats in mountains by the atmospheric deposition of organic chemicals is therefore challenging.

Based on the risk assessment by Machate et al. [19]—which provided new insights into the diversity of organic chemicals that can be found in mountain lakes by using a time-integrated sampling method and covering a large spatial extent of the French Pyrenees—the levels of organic chemicals introduced via atmospheric deposition are sufficiently high to cause chronic toxic effects towards crustacea in all lakes under investigation. Also when looking at the results of the here performed risk assessment, almost one third of the investigated lakes experience a chronic toxic risk. Taking into account that most of the considered studies only included a relatively small set of target pollutants (mostly POPs) and utilized grab samples, thereby neglecting relevant drivers of toxicity (e.g., chlorpyrifos) (Fig. 1) and temporal variations, it is very likely that the actual toxic risk is higher than predicted here. Furthermore, water concentration data are missing for mountain lakes and regions that are known to be considerably polluted with organic chemicals (Fig. 3, details in Additional file 1). This applies for example to the Tatra mountains. Studies investigating their pollution history revealed sediment burdens with PAHs and DDT that are similar to those of urban areas and higher than what has been discovered in other mountain lakes [70–72]. Therefore, it cannot be ruled out that regions exist where pollution is more severe than suggested based on the risk assessment performed here or by Machate et al. [19]. Generally, the current data suggest that organic pollutants introduced via atmospheric deposition are likely to cause widespread chronic toxic risks in mountain lakes. However, the currently available monitoring data likely underestimate the toxic risks, also due to the fact that accurate chemical monitoring strategies are difficult to perform in such a remote and difficult to access habitat.

Local sources cause acute toxic risks

Local emission sources of organic chemicals in mountain areas have been widely neglected, as urban settlements and conventional agricultural practices are mostly absent at high altitudes. However, local anthropogenic activities like mining, charcoal production, tourism, and pastoralism are practised in mountains since a long time (Fig. 2) [73, 74]. While mining mainly introduced inorganic pollutants into the mountain environment [17], tourists can introduce contaminants like pharmaceuticals, personal care products and insect repellents [15], plastics [75], and PAHs (e.g., through car travel) [58, 76]. Although not investigated yet, it is to be expected that the input of chemicals by tourists is increased especially where there are mountain refuges and huts, as these frequently attract higher amounts of tourists and often only have a rudimentary wastewater treatment system. Additionally, also other local activities, like the practices of pastoralism (releasing of cattle into the mountains over the summer), laying fires to keep landscapes open, and the introduction of fish, are known to introduce pollutants into mountain lakes [16, 19, 77, 78].

Only few studies provide insights into the risk and impact by local pollution sources on mountain lakes. For the French Pyrenees, Machate et al. [19] report an acute toxic risk for and impact on crustacean communities in mountain lakes, due to the introduction of the highly toxic compounds diazinon and permethrin through local sources (livestock and tourism). Via the here performed risk assessment it was possible to identify another study, performed in different mountain lakes of the Sierra Nevada in California, where insecticides (malathion and diazinon) could be detected at concentrations that suggest local sources and cause an acute toxic risk for the present crustacean communities (Fig. 1) [33]. Acute toxic risks from the input of insecticides by local sources are therefore not only a phenomenon of the Pyrenees, but likely also affects mountain lakes in other mountain ranges of the world. While more data are needed to fully comprehend the extent of this risk, this demonstrates that acute toxic risks for water bodies cannot only be found in populated lowland regions, but also in waterbodies of remote mountain areas.

Evidence for toxic impacts by chemical pollution in mountain lakes

Various studies demonstrated that pollution-induced toxicity causes biological responses in aquatic mountain organisms and their populations. Their results showed: (1) a widespread estrogenic activity in sediments of 83 European mountain lakes, ranging from Norway to the Pyrenees and Eastern Europe [79]; (2) a background fish feminization in a lake from the Tatras and the Pyrenees, identified by investigating the expression of oestrogen receptors and zona radiata genes in male fish [80]; (3) an expression of the Cytochrome P450 1A enzyme in fish of European mountain lakes, indicating a widespread impact by pollution on aquatic organisms of European mountains [81]; (4) altered zooplankton communities with reduced numbers of crustacea in mountain lakes of the French Pyrenees [19]; (5) in situ effects in frogs from the Sierra Nevada Mountains (California, USA), including an increased mortality, slower developmental rates and lowered cholinesterase activities in more polluted mountain regions [82], and (6) multiple studies suggesting that pesticide pollution drives frog population

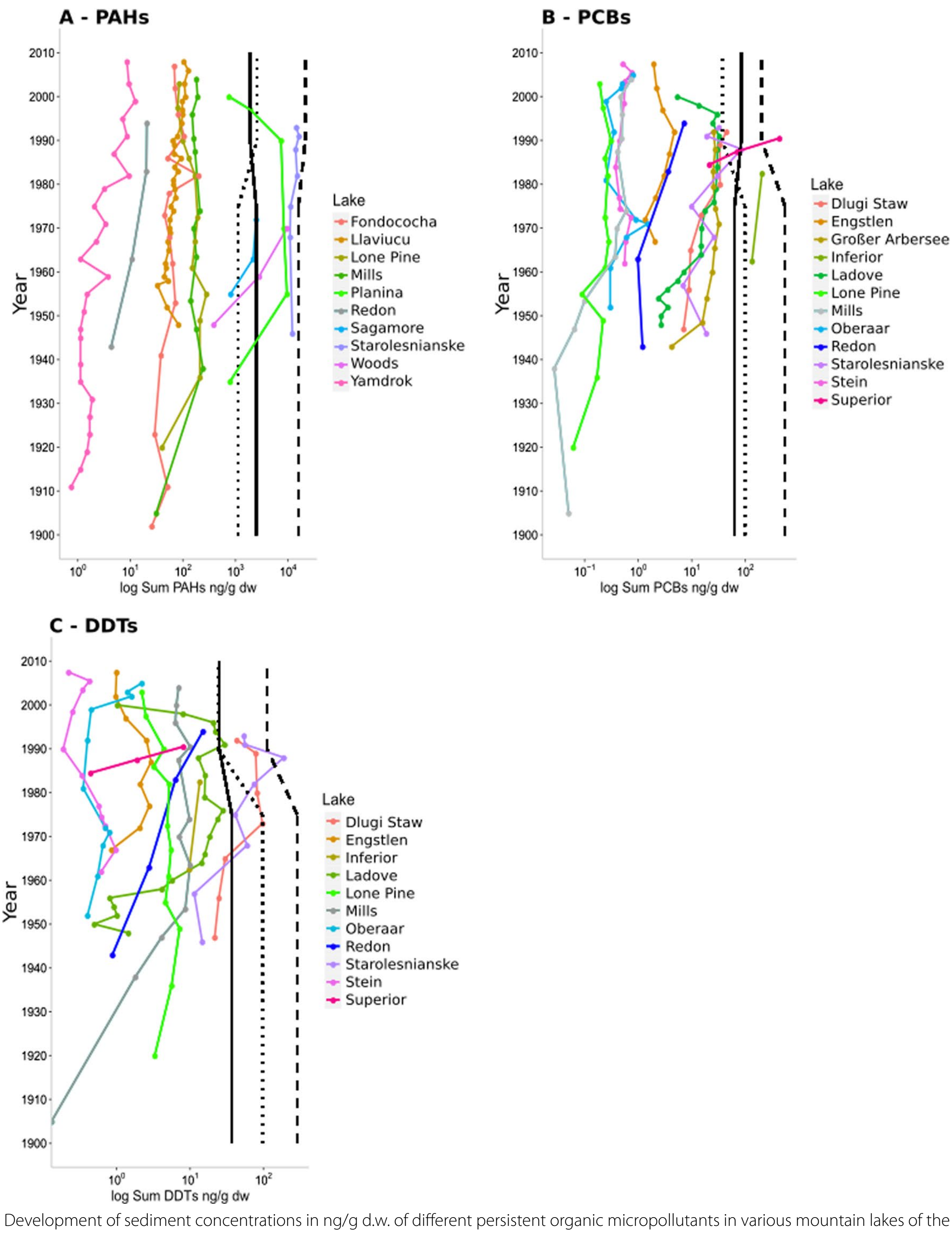

Fig. 3 Development of sediment concentrations in ng/g d.w. of different persistent organic micropollutants in various mountain lakes of the world over time. Development of **A** ∑PAH concentrations, **B** ∑PCB concentrations and **C** ∑DDT concentrations. Vertical lines represent points of reference and show average concentrations of the 1970s and − 90 s in lakes of the U.S. in urban (dashed line), light urban (dotted line) and remote areas (solid line) (Additional file 1: Table S2)

declines in the Sierra Nevada Mountains [83–87]. It is now known that the fungal pathogen *Batrachochytrium dendrobatidis* (Bd) caused the observed amphibian decline in the Sierra Nevada and many other parts of the world [88]. However, considering the number of studies suggesting a cause–effect relationship between pesticide pollution and declining frog populations at high altitudes, pollution and *Bd*-driven declines are likely linked. Chemical pollution might have increased the Bd infection rate by weakening the immune system of frogs and reducing the abundance of aquatic predators that otherwise consume Bd zoospores and, thereby, reduce infection pressure on amphibians [89, 90]. This notion is supported by other studies, which confirm that an increased success of infectious diseases might be linked to pollution [91]. However, more studies on the cause–effect relationships between pathogen outbreaks and pollution are required to confirm this observation.

Therefore, similar to the results of the here performed TU-based risk assessment, also studies that looked at biological indicators support the idea of widespread toxic effects, and especially chronic toxic effects, on aquatic organisms in mountains. While also in this regard more studies will be required to allow for a reliable conclusion, these results agree with findings made in the polar regions. Similar to the Arctic, also mountainous areas have been shown to act as focusing regions for POPs with reported concentrations being similar or even slightly higher than in the Arctic [21]. Considering that similar concentrations of POPs cause severe adverse effects on the wildlife at the top of the arctic food-chain (e.g., immune system suppression, endocrine disruption, and carcinogenicity as well as impacts on behaviour, reproduction, and development) [13, 14], we may assume that aquatic organisms in the mountain environment are similarly impacted by pollution-induced adverse effects. A negative effect on the health of aquatic mountain organisms due to chemical pollution is therefore very likely.

Habitat characteristics affect vulnerability towards and fate of chemical pollution

Mountain habitats exhibit unique characteristics. While currently little explored, these characteristics have the potential to cause aquatic ecosystems to be more vulnerable to pollution. The harsh living conditions include cold temperatures, thin air, low nutrient levels, high UV radiation, as well as long winters and short summers [6], which are known to strongly enhance the sensitivity of organisms to pollution by a factor of up to 100 [92–95]. Furthermore, increased UV radiation and clear mountain lake waters may enhance toxicity of compounds that are susceptible for photoactivation, such as PAHs [96, 97]. To endure the harsh living conditions in the mountain environment, organisms have developed certain traits such as increased longevity, higher lipid contents and slower growth rates [21]. While necessary to survive at high altitudes, these traits favour the bioaccumulation of organic chemicals in the organisms and increase internal concentrations [21, 55, 98–100]. This effect is probably magnified by the typically oligotrophic conditions in mountain lakes. Under such conditions, (planktonic) organisms represent the highest amount of total organic matter in the lake and nutrient cycling is highly efficient. This causes an increased and extended exposure of present organisms while sedimentation is reduced [101, 102]. In addition, mountain lake food webs are typically short and unbranched with highly specialized top predators [21]. In such food webs, biomagnification of POPs in top predators is particularly severe, resulting in a great ecotoxicological risk for those species [13]. Similar risks were reported for organisms that live close to or temporarily in mountain lakes (amphibians and birds of prey) [66, 103] and likely also apply for waterborne organisms [21, 43]. However, some studies also obtained results that deviated from this assumption [104, 105]. Something that is likely caused by differences in feeding habits and the zone of the lake organisms inhabit (pelagic vs benthic), which lead to a more complex exposure scenario of the present organisms and may make it difficult to demonstrate the effect of biomagnification.

Furthermore, the very secluded and patchy landscape of mountains may cause a higher sensitivity of mountain organism populations to pollution events. For example, disconnected mountain lakes form insular ecosystems [54], where a re-colonization from intact populations of neighbouring water bodies is hampered. Thus, the recovery of a mountain lake after a toxicity event is strongly hindered [106]. The ability to recover is likely further reduced by the slow growth rates that prevail at high altitudes [21]. Consequently, recovery processes are likely to be slow or even incomplete in case a species has been lost. Considering that mountain lakes constitute relatively simple systems with a low functional redundancy, the introduction of organic chemicals poses a great risk for their overall functioning and health [107].

The special habitat characteristics prevailing in mountain landscapes might not only impact the vulnerability of their aquatic ecosystems, but also cause pollutants to accumulate in mountain lakes. Mountain watersheds are often characterized by a very limited soil development and generally extremely low organic carbon contents [6]. This reduces the ability of the catchment to effectively retain organic chemicals and support their mobilization and accumulation in downstream mountain lakes [10, 20, 56]. Especially mountain lakes with longer water residence times or a closer ratio between lake size and the size of their watershed, have a higher potential to accumulate pollutants [66]. That potential may be further enhanced by the prevailing low temperature that reduces the volatilization of compounds into the atmosphere [50]. In addition, low temperatures also reduce the efficacy of hydrolysis and biodegradation processes [108], likely increasing the build-up of chemical pollutants in mountain lakes. However, as the levels of UV radiation increase with increasing altitude and due to the fact that the water of mostly oligotrophic mountain lakes is very clean, which allows the water to be penetrated to a greater depth by the incoming radiation, this may lead to an increased efficacy of photodegradation processes [6, 109], which might partly counteract the tendency of chemical accumulation. Nevertheless, all in all the prevailing environmental conditions render mountain lakes as a focusing point and effective sink for organic chemicals in the mountain environment. Something that has also been observed by few studies [41, 54, 70].

Conclusion and research needs

Currently, the knowledge on the risk for mountain lakes by chemical pollution is limited. Neither their exposure nor their sensitivity is explored sufficiently. Similar to lowland waterbodies, mountain lakes are exposed to a complex mixture of organic chemicals via local activities and atmospheric deposition. This chemical cocktail can cause acute or chronic toxic risks for aquatic organisms with cascading effects potentially affecting the whole mountain environment, including, e.g., water quality. These risks may be enhanced by the specific habitat conditions and the possibly greater vulnerability of mountain organisms to chemical pollution. Considering the importance of aquatic mountain lake ecosystems for the global supply with drinking water, more efforts are needed to monitor chemical pollution and its impact on mountain lake ecosystems on a regional, continental, and global scale. Only then will it be possible to close the existing knowledge gaps and preserve these ecosystems and their services for future generations.

The assessment of chemical risks for mountain lakes is hampered most by the lack of monitoring data and hence exposure information (Table 1). In a mountain context, only few lakes, regions and compounds have been investigated so far. Future chemical monitoring needs to go substantially beyond POPs well-known for atmospheric transport, since recent studies indicated that unexpected compounds from local sources may have the potential to drive toxic risks in mountain lakes [19]. State-of-the-art LC–HRMS and GC–HRMS target, suspect and non-target-screening [110, 111], together with effect-based monitoring [112, 113], are needed to allow for a comprehensive monitoring of complex mixtures and help to better characterize chemical pollution and possible toxic risks and effects in mountain lakes. Thereby, low chemical concentrations, which demand for a sample enrichment, and logistic challenges of transport and preservation of large water volumes in remote areas, need to be overcome by utilizing passive sampling techniques. These techniques allow for time-integrated sampling of a broad range of pollutants, including substances with effect concentrations in the nanogram per litre range [114–117].

The results obtained by these studies need to be used to better understand the spatial distribution of chemical pollution. To this end, it would be helpful to advance existing exposure models. The currently most progressive model is the MCMPOP model, which considers air, soil, precipitation (rain and snow), vegetation, and ice cover in order to understand the transport processes of POPs across the central Himalayas [118]. Future models should include information on short- to long-range atmospheric transport, thereby also considering the mountain-specific distribution of air masses and precipitation, together with local sources and the characteristics of lakes and catchment, asking for high-resolution remote sensing data. By developing such an advanced model, it would be possible to predict those lakes that are most exposed to and hence at the highest risk by chemical pollution. This is currently still difficult, which makes it difficult to plan meaningful exposure monitorings and fully understand the existing toxic risk by chemical pollution. Furthermore, having such models would help to develop a better understanding of how global climate and land use changes will alter the future exposure and toxic risk for mountain lakes by organic chemicals [119]. Knowledge that is still lacking, but crucially needed in order to be able to effectively protect and maintain the health of mountain lakes. Another obstacle that needs to be overcome in order to fully understand the risk for mountain lakes by chemical pollution is the uncertainty around the sensitivity of their organisms. Based on their characteristics, organisms in mountain lakes are likely to be more vulnerable to chemical pollution. Standard toxicity tests are, however, done under laboratory conditions, which is likely leading to an underestimation of the prevailing toxic risk when using EC to assess the prevailing risk. Therefore, improved exposure modelling and monitoring needs to be complemented by a better understanding of the impact of chemical pollution on mountain lake ecosystems and their biodiversity under multiple stress conditions. Therefore, investigations on the ecotoxicity of relevant pollutants, and mixtures thereof, under realistic stress conditions are urgently needed. Relevant stressors include traditional environmental stressors like high ultraviolet radiation, cold temperatures and food shortage [96, 120, 121], but also pathogens [122, 123] and climate change scenarios [124], and should be supported by predictive models on multiple stresses [95]. Also paleolimnological studies could be useful, since they allow to understand the impact of chemical pollution over longer periods of time and help to reconstruct how mountain lake ecosystems looked like before the introduction of anthropogenic organic chemicals [125, 126].

Abbreviations

POPs	Persistent organic pollutants
TU	Toxic unit
SI	Supplementary information
BDEs	Brominated diphenyl ethers
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
AF	Additional file
OCs	Organochlorine pesticides
CUPs	Current-use pesticides
PAHs	Polyaromatic hydrocarbons
PCBs	Polychlorinated biphenyls
EC	Effect concentration
NOEL	No observable effect levels
LOEL	Lowest effect levels
NOEC	No observable effect concentration
LOEC	Lowest observable effect concentration
Kwa	Water–air partitioning coefficient
Bd	*Batrachochytrium dendrobatidis*

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s12302-022-00710-3.

Additional file 1: Figure S1: Comparison of freely dissolved equilibrium water concentrations, predicted based on sediment concentrations, towards water concentrations measured in other monitoring studies of mountain lakes. **Table S1:** Sediment core data for mountain lakes from different mountain ranges of the world containing concentration information on sums of polyaromatic hydrocarbons (∑PAHs), polychlorinated biphenyls (∑PCBs) and DDTs (∑DDTs). Where certain compounds were not measured, they are marked as not assessed (NA). **Table S2:** Reference values for mountain lake sediment cores derived from Van Metre et al (2004) in ng/g d.w. derived by investigating 38 U.S. lakes and reservoirs. Values displayed are means of values stated for the respective lakes and reservoirs. Urban = In densely populated region; Light Urban = In less densely populated regions; Remote = In scarcely populated regions.

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Author contributions

OM gathered the data and information presented here, performed the toxicity risk assessment and drafted the manuscript. DSS initiated the manuscript and provided guidance on the ecology and vulnerability of mountain lakes ecosystems. TS provided the toxicity data and guidance on their application. WB supervised the work on this manuscript, helped to structure its content and contributed text passages to the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

The datasets supporting the conclusions of this article are included within the article and its additional file. Furthermore, they can also be found in the data repository Zenodo (https://doi.org/10.5281/zenodo.7013722).

Declarations

Ethics approval and consent to participate

No reporting on any animal or human data or tissue is included within this manuscript. Following no approval or consent is required.

Consent for publication

All data that have been presented throughout the manuscript or are attached within the additional file have either been publicly available or if not, the originator of the data has agreed to it being used in this manuscript.

Competing interests

The authors declare they have no competing interests.

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