Soil's Hidden Power: the stable soil organic carbon pool controls the burden of persistent organic pollutants in background soils

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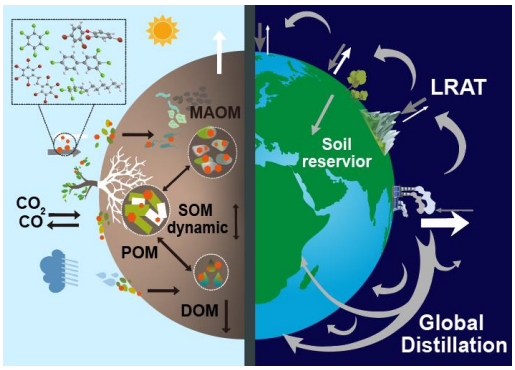

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43 **Graphic for Table of Contents**

Abstract

Persistent organic pollutants (POPs) tend to accumulate in cold regions by *cold condensation* and *global distillation*. Soil organic matter is the main storage compartment for POPs in terrestrial ecosystems due to deposition and repeated air-surface exchange processes. Here, physicochemical properties and environmental factors were investigated for their role in influencing POPs accumulation in soils of the Tibet Plateau, Antarctic and Arctic regions. The results showed that the soil burden of most POPs was closely coupled to stable mineral-associated organic carbon (MAOC). Combining the proportion of MAOC and physicochemical properties can explain much of the soil distribution characteristics of POPs. The background levels of POPs were estimated in conjunction with the global soil database. It led to the proposition that the stable soil carbon pools are key controlling factors affecting the ultimate global distribution of POPs, so that the dynamic cycling of soil carbon acts to counteract the *cold-trapping* effects. In the future, soil carbon pool composition should be fully considered in multimedia environmental model of POPs, and the risk of secondary release of POPs in soils under conditions such as climate change can be further assessed with soil organic carbon models.

Keywords

Persistent organic pollutants, soil organic matter, background soil burden, mineral-associated organic matter, Tibetan Plateau, Antarctic and Arctic regions

Synopsis

This work suggests that SOM, particularly stable components, plays a crucial role in burden of POPs in the global background soils.

1. Introduction

Persistent organic pollutants (POPs) are now found in all of the Earth's environments mainly following their long-range atmospheric transport (LRAT)[1-5]. *Global distillation*[6, 7], in response to repeated air and surface temperature changes, has been proposed to drive POPs towards a kind of *global equilibrium* steady state. As a result, both the Arctic[7, 8] and Antarctic[9, 10] have been considered as the important receiving environments for POPs, due to the cold condensation effect. At the same time, the Earth's surface environment can also affect the LRAT of POPs and retard their re-emission, which will confound such global cold condensation. Surface soil is likely to be of critical importance to the global budget and cycling in this respect, because soil organic matter (SOM) has a very high capacity to store and interact with POPs[11, 12]. Therefore, the LRAT of POPs could be reduced by surface absorption and retention, limiting repeated alternation of warm volatilization and cold deposition. These processes are complex, because they depend on the properties of the chemicals, the surface environment, and the ambient conditions. These ideas have been proposed before[13, 14], but no study has systematically addressed the links between POPs and SOM pools/dynamics on a global scale.

As the largest organic carbon (OC) pool in the terrestrial ecosystem[15], SOM is of key importance for POPs storage, cycling and possible biodegradation, acting as a buffer retaining POPs and retarding their re-emission to the atmosphere[16]. The effect of SOM on the fate of POPs in soils and sediments has been widely studied[17-20]. However, these early studies largely considered SOM as a single compartment. In reality, SOM is much more complicated - a continuum of organic materials from granular organic debris to small organic molecules[15]. As such, SOM comprises various OC pools that are sensitive to changes in the environment and dynamic, with different turnover times[21-24]. POPs demonstrate high persistence in the environment, having long half-lives in soils[25]. So, for these long-lived chemicals, coupling to the OC cycle will determine their long-term fate and environmental significance [26]. In recent decades, the importance of mineral protection in promoting soil organic carbon (SOC) preservation has been widely recognized[27, 28]. Growing studies suggest that SOC turnover and

response to climate change can be better described if SOM is broadly divided into a particulate organic matter (POM) and a mineral-associated organic matter (MAOM) pool, which are fundamentally different in terms of their formation, persistence, and functioning[22, 29, 30]. In general, POM is largely made up of lightweight fragments of plant material, while MAOM mainly consists of small compounds associated with soil minerals[31,32]. So that MAOM is a more beneficial form of long-term soil OC storage than POM[22, 29]. Therefore, we hypothesized that the burden of POPs in soils will be greatly affected by the the proportion of MAOM/POM.

On a global scale, all surface soils receive POPs from atmospheric deposition, while only 0.04% of the soil burden also comes from receiving direct inputs of significant industrial and man-made emissions[16]. Thus, background soils (i.e. those only receiving atmospheric inputs of POPs) are essential to comprehensively represent and investigate the global fate of POPs. Soil samples in this study were collected from the Tibetan Plateau (TP) and the polar regions, which remain relatively pristine with less interference from local emissions. Highly diverse types of soils are included in these regions[18, 33], including oligotrophic to carbon-rich soils. The SOM in sub-regions of the polar and TP are vulnerable to ongoing climate changes, which in turn affects the behavior of POPs in soil[9, 34]. On the other hand, POPs stored in soil with different SOC contents will take different time to reach air-soil steady-state. For low-SOC soils, equilibrium conditions may already exist for some POPs, while not yet attained for high-SOC soils. As a key sink for POPs under the global distillation hypothesis[35, 36], the TP and polar regions with a wide range of SOC contents are envisaged as ideal areas to study the background soil burden of POPs through LRAT[7, 37], as well as the effects of the combination of climate perturbation and soil carbon cycling.

In this study, a total of five classes of POPs were analyzed, including legacy (organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs)) and emerging POPs (polybrominated diphenyl ethers (PBDEs), and short-chain chlorinated paraffins(SCCPs)) as well as emerging contaminants including novel brominated flame retardants (NBFRs), medium- and long-chain chlorinated paraffins (MCCPs and

LCCPs). Globally, these POPs are emitted from different source areas, with different usage histories, covering a wide range of physicochemical properties as well as different timescales for global re-distribution and to approach global equilibrium. Different soil OC fractions were also determined in the soils. Commencing with an examination of the interplay between the dynamic evolution of SOM and distribution of POPs by using synthesized statistical analyses, we compared the main mechanisms influencing the burden of POPs in background soils. Through a meticulous analysis at both microscopical and macroscopic levels, this study allows us to assess: i. the large-scale distribution and retention of POPs in background soils across an unprecedented range of environmental gradients; and ii. the pivotal role of the stable fraction of SOM on the accumulation of POPs in background soils.

2. Experimental section

The description of materials, sample preparation, and analytical quantification are shown in Supporting Information (SI) (Text S1).

2.1 Study area and sampling sites

Eighteen sites were selected in Ny-Ålesund and London Island, Svalbard in the Arctic (A1−7), and King Georgia Island (KGI), Fildes Peninsula in Antarctica (S1−11) (Figure S1). During the Chinese scientific research expeditions to the Arctic from 2011 to 2017 and the Antarctic from 2012 to 2018, surface soils were collected covering tundra areas and coastal mudflats.

In July 2019 − July 2021, soil samples were collected from 23 sites along a 457 km transect (covering Nyingchi, and Lhasa) across the TP region (Table S1). Therein, Shergyla Mountain, Nyingchi, holds a comparatively complete elevational belt spectrum that comprises a variety of climatic zones ranging from the mountainous tropics to the alpine cold zone. Fourteen sites were evenly distributed at 200−300 m intervals on the east (TS1−8) and west (TS9−14) slopes, covering an elevation gradient ranging from 2440 m to 4580 m. Nine points were located in Lhasa, five of which were evenly placed in urban areas (TS15−19), while four sites were located around Namsto

(TS20−23). The distribution of five POPs in surface soils (0−10 cm) across the TP region was analyzed in conjunction with SOM analysis covering different lands. Particle size fractionation was used to separate SOM into MAOM and POM[23, 38] (Figure 1). MAOM is defined as the soil organic matter fraction <53 μm and POM is 53–2,000 μm, after aggregate dispersion. The OC content of above two types SOM were regarded as mineral-associated organic carbon (MAOC) and particulate organic carbon (POC), respectively. Published data of the other four POPs classes in polar soils (Specific data and references are in Table S2 and S3) were compiled with data on chlorinated paraffins (CPs) conducted in this work. Basic meteorological data of the TP region was obtained from the South-East Tibetan Plateau Station of the Chinese Academy of Sciences[39].

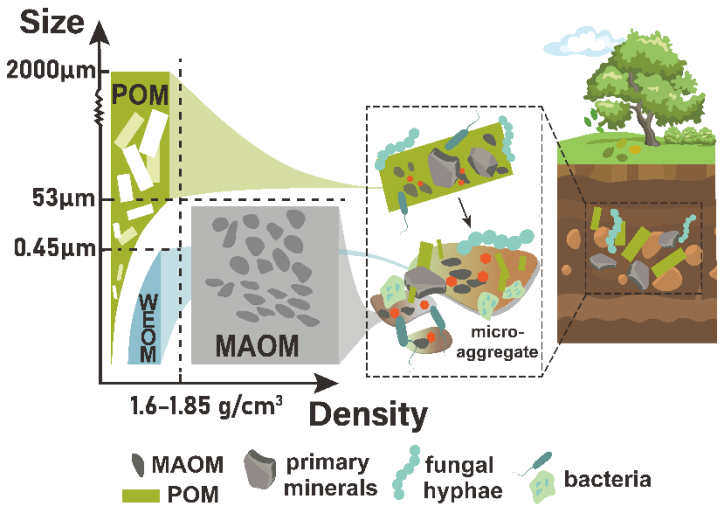

Figure 1. Conceptual representation of major terrestrial pools of organic carbon (OC) developed by Lavallee et al., (2019) and Witzgallin et al., (2021) in this study. These OC components are physically defined based on size and density, shown on the y and x axes, respectively.

2.2 Quality Assurance and Quality Control

To minimize contamination during analytical procedures, strict quality assurance and control measures were conducted which were reported in previous studies[4, 40, 41]. Method detection limits (MDLs) of contaminants were determined as three times the standard deviation (SD) of method blank concentrations. For those contaminants with no signals in the blanks, MDLs were determined as the lowest standard in the

calibration curve. In this study, the MDLs for PCBs, PBDEs, NBFRs, and OCPs were in the range 0.09–0.20 pg g^{-1}, 0.01–0.38 pg g^{-1}, 1.00–4.00 pg g^{-1}, 0.02–0.29 ng g^{-1}, respectively. The MDLs for SCCPs, MCCPs, and LCCPs were 1.16, 0.35, and 0.36 ng g^{-1}. The surrogate standard recoveries were in the range of 53.0–128% for CPs, 59–104% for PCBs, 64–92% for PBDEs, 53.1–148% for OCPs, and 79.3%–101% for NBFRs. Results reported in the study are expressed on a dry weight basis and corrected for recovery results. And for the sake of further calculation, data below the detection limit were replaced by half of the MDLs.

2.3 Statistical analysis

The physicochemical properties were obtained from the U.S. EPA as a part of the Estimation Program Interface (EPI) Suite. In which, an improved MCI-K_{OC} relationship for both polar and nonpolar chemicals by using a combination of the first-order MCI and a series of statistically derived fragment correction factors. This alternative to the op-LFER approach has been encoded into a computer program, PCKOCWIN.

Differences in nonparametric grouped data were analyzed using Kruskal-Wallis (K-W) analysis of variance (ANOVA) tests by IBM SPSS Statistics 26. Considering greatly different sample numbers of soil, the K-W test could provide more robust results than parametric methods when comparing the POPs level and pattern among sample types.

Network analysis has been used extensively in ecological studies to visualize the underlying associations between environmental factors and complex contaminant components. Here, a correlation matrix was constructed by calculating all pairwise Spearman's correlations among physicochemical properties and distribution characteristics of POPs with the package in OriginPro 2024. The correlation that occurred in P-value > 0.05 of all samples was discarded to reduce the complexity of computation and avoid spurious correlation bias. To quantitatively analyze the effects of various factors on the distributional characteristics of POPs, sensitivity analyses of the variables were conducted with other factors held constant in the model.

Structural equation models (SEMs) were adopted to evaluate the direct and indirect effects of environmental factors (i.e. meteorological factors, geographical conditions, and soil components) and POPs patterns. SEM is an a priori approach with the capacity to identify causal relationships between variables by fitting data to the models representing causal hypotheses. In this paper, α-coefficient (Cronbach's Alpha) was used to measure the reliability of the seven factors (α=0.788 with 95% confidence interval range from 0.599 to 0.900). SEMs are tested based on robust maximum-likelihood evaluation with IBM SPSS Amos 22. For the first SEM of environmental factors and POPs patterns, χ^2=35.3, p<0.001, CMIN/DF (ratio of χ^2 and degrees of freedom) =6.31, GFI (goodness-of-fit index) =0.994, CFI (comparative fit index) =0.998, RMSEA (root mean square error of approximation) <0.05. The SEM for five separate categories POPs distribution was also validated and yielded a good model fit, including PBDEs (χ^2=5.32, p=0.378, CMIN/DF = 1.063, GFI = 0.992, CFI=1, RMSEA < 0.01), PCBs (χ^2=4.10, p=0.092, CMIN/DF = 1.026, GFI = 0.996, CFI=1, RMSEA < 0.01), CPs (χ^2=6.16, p<0.05, CMIN/DF = 1.54, GFI = 0.996, CFI=0.999, RMSEA < 0.01), OCPs (χ^2=8.28, p=0.218, CMIN/DF = 1.38, GFI = 0.995, CFI=0.998, RMSEA < 0.05), NBFRs (χ^2=7.61, p=0.179, CMIN/DF = 1.06, GFI = 0.992, CFI=1, RMSEA < 0.01).

Radial Basis Function (RBF) networks represent a type of artificial neural network (ANN) with an input layer, an output layer, and one hidden layer of radial units, each modeling a Gaussian response surface. This hidden layer of the RBF act based on the radial basis activation function. The RBF models developed in this study were divided into two categories distinguished by different roles. In this work, RBFANN is conducted by IBM SPSS Statistics 26 to predict the global distribution based on the SOM database and empirical data. Three variables (TOC, MAOC, and POC) were used as inputs for RBFANN. The numbers of hidden layer and output units vary with different POPs, with specific descriptions in Table S6.

Uncertainty analysis was performed using Monte Carlo simulation assuming that the parameters in the above model obey a lognormal distribution, the output was

calculated by repeating the simulation 2000 times to calculate the coefficient of variation(C_V) and the interquartile differences(Q) (Text S1).

3. Results

3.1 Spatial characteristics of legacy and emerging POPs in TP and polar soils

In this work, the ΣCPs in the polar soils and the soil burden of the five POPs classes from the TP region were analyzed. The soil characters and concentrations of Σ_{25}PCBs, Σ_{27}PBDEs, Σ_{9}NBFRs, Σ_{13}OCPs, and Σ_{89}CPs are listed in Table S1–4 and Figure S1–S3.

In this study, the difference between the highest and lowest levels of POPs was nearly three orders of magnitude. ΣOCPs, ΣPBDEs and ΣNBFRs from the TP region were higher than those in the Antarctic[42,43] and Arctic[44] (Figure S3). This is the first report of the soil burden of LCCPs (in a range of <MDL to 14.4 ng g^{-1}), and they occur at a higher level than the other four POPs in some remote areas (Figure S1 and S2). Contrary to the other target POPs in this study, ΣPCBs were significantly lower on the TP than those in the polar regions, especially the Arctic.

The more barren and frozen environment of the polar regions can result in lower degradation rates and is more affected by the historic source regions of PCBs (e.g., Europe and North America[6]). As legacy POPs, decreasing primary emissions (since the peak in the 1960/70s[6]) and long timescales may explain the low soil burden of PCBs in remote regions. However, diffusive emissions still occur from ongoing use in legacy stocks and waste streams[45], which contribute to ongoing releases and deposition in remote regions via LRAT. Similar but more time-compressed situations have occurred for some of the new POPs, such as PBDEs[46]. The generally lower production[46] of PBDEs compared to PCBs and higher bio-degradation rates[47] likely contribute to this lower soil burden of PBDEs. Emerging replacement/alternative chemicals, NBFRs and CPs, have been globally mass-produced and used (especially in Asia[48, 49]), resulting in their higher occurrence in these soils, since they have only recently been restricted. Despite the global restriction, some OCPs are still in use across South Asia (notably India)[50]. Driven by the Indian monsoon, these POPs will enter the Tibetan region via

air mass movement and accumulate in soils following deposition[51].

In regions devoid of direct contamination by POPs, the soil's capacity for accumulation emerges as a pivotal determinant shaping the distribution and concentration of these pollutants. In this work, our subsequent analysis and discussion section are predicated on the premise that disruptions in POPs dynamics across the aforementioned remote environments primarily stem from the transboundary transport of pollutants via LRAT originating from lower latitudes. The Tri-polar regions stand out as one of the rare biospheric realms characterized by limited human presence, facilitating the identification of pristine zones largely untouched by anthropogenic pollutants. While localized anthropogenic sources of pollution exist in remote polar regions, which may increase the uncertainty of discussion within the purported "last clear background".

3.2 Interactive impacts of SOM fractions on soil distribution of POPs

To identify the main environmental factors associated with POPs distribution, separate linear regression analyses were performed between the POPs concentrations and all available factors. These independent variables include longitude, latitude, elevation, ambient temperature, humidity, and SOM variables (i.e., SOC, POC, MAOC, and water extractable organic carbon (WEOC)) (Table S1).

Throughout the large-scale meta-analysis (Figure S4), the SOC components showed a significant positive relationship ($p \leq 0.05$) with 41.7% of the target POPs components (e.g., PCBs). For some components (e.g., Aldrin, Isodrin, and Nona/DeBDEs), indistinct or opposite correlations with SOC were observed. These results indicate that the spatial distribution of POPs in the soil may be affected by a combination of various environmental factors or competing processes (e.g., soil storage, degradation, and losses[16]). PCBs have been banned for a long time, so have had longer to reach dynamic equilibrium between soils and air[14]. Therefore, the distribution of these legacy POPs is more dependent on the soil OC pool. With mass manufacture and use in the regions bordering the TP[50], the distribution of some POPs, notably OCPs, was also affected by the monsoon from these areas[52], overlaying cumulative

atmospheric deposition to the soil with trace levels of these compounds. Additionally, the lack of correlation could be related to the relatively low concentrations of some compounds in the soil samples (e.g. low amounts of DeBDEs reaching remote soils due to its high molecular weight/low volatility and limited LRAT)[53].

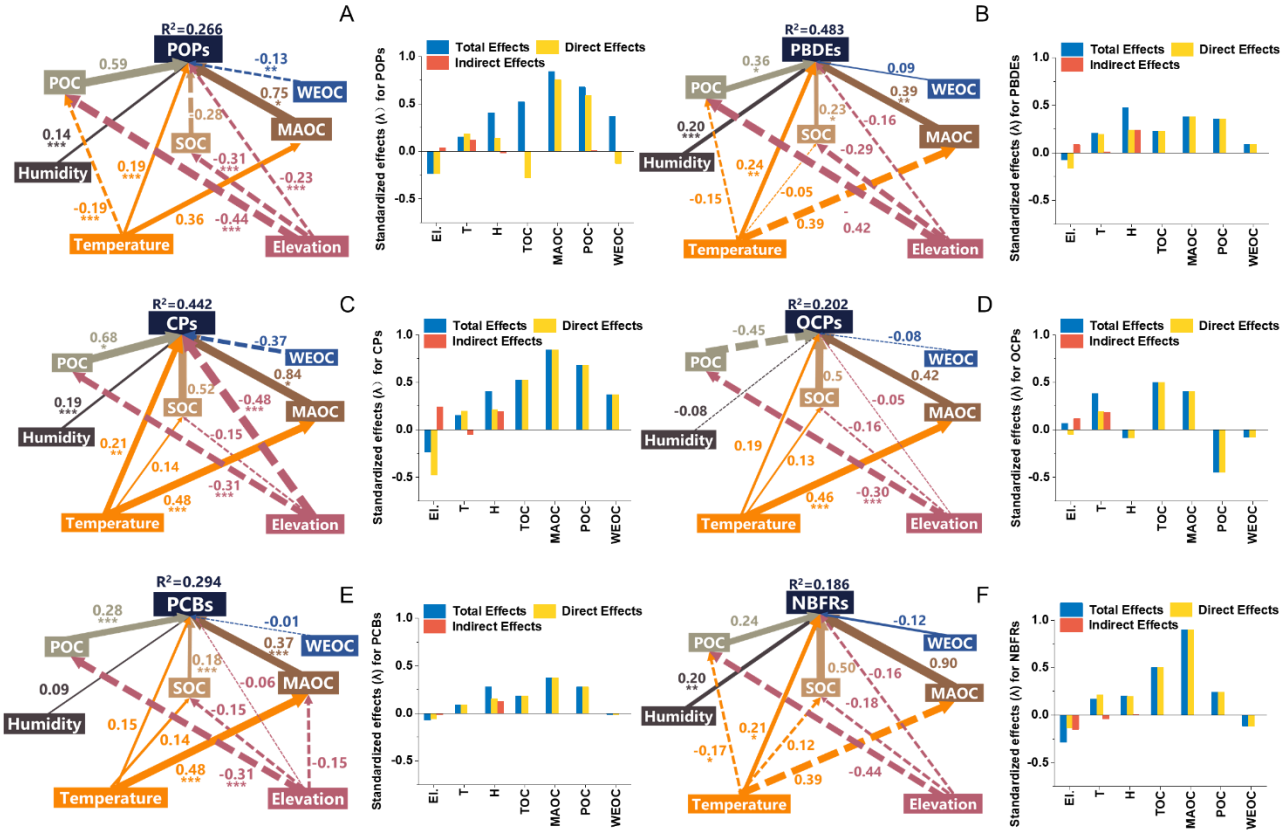

Figure 2. Structural equation model (SEM) demonstrated and quantified the direct and indirect effects of environmental factors that influenced the occurrence of five groups of POPs in background soils. The width of each arrow is proportional to the standardized direction coefficients (λ), shown as numbers on the arrows. The solid and dashed lines indicate positive and negative flows of causality, respectively. R^2 represents the variance of factors explained for each endogenous variable. The total effects were the sum of direct and indirect effects. El, T, and H are abbreviations for elevation, temperature, and humidity, respectively.

To delineate the cascading relationships between the POPs distribution and environmental factors, a structural equation model (SEM) was introduced to quantify these complex mutual effects (Figure 2). The model consisted of seven factors with high impacts based on the aforementioned test. The interactions between environmental factors and POPs distribution were quantitatively described as direct and indirect effects of soil properties and geographic variables on ΣPOPs. The combination of climatic

316 factors and SOM components explained 26.6% of the variance in five POPs (variation
317 ranging from 18.6% to 48.3%). MAOC and POC were the two major factors
318 influencing the POPs distribution. This dominant influence of SOC was supported by
319 the aforementioned partial correlations. The effects of elevation, temperature, and
320 humidity on POPs occurrence represented a combination of direct effects and indirect
321 effects through soil properties, and these direct effects of climatic factors on POPs were
322 less pronounced than indirect effects. The correlation degree for NBFRs and OCPs were
323 lower. Discrepancy of internal monomeric component (e.g., different production and
324 scale of use) may influence the overall relationship between these two POPs and SOC
325 components.

326 The results of the Monte Carlo simulation show that the C_V and Q value for
327 elevation are overall higher than the other inputs (Figure S5), indicating a high level of
328 uncertainty in the extent to which the parameter of elevation affects POPs. In addition,
329 the uncertainty of SOC components varies for different POPs, so it is necessary to
330 further confirm the altitude and each SOC parameter to improve the accuracy of the
331 calculation results.

332 Besides, the persistence and accumulation of POPs in soils are governed by a
333 multitude of factors, including partition properties and degradation processes within the
334 soil matrix. While our study primarily delves into the characterization of SOM
335 properties, it is also important to acknowledge the intricate interplay between various
336 degradation mechanisms and environmental factors. Indeed, biodegradation,
337 photodegradation, and chemical degradation also represent key pathways through
338 which POPs transform within soil matrices[54] , influenced by parameters such as SOC
339 composition, temperature, moisture levels, and the intrinsic properties of the POPs
340 under investigation. Of particular significance is the role of microorganisms in
341 facilitating the biodegradation of POPs, resulting in a very complicated process. In
342 order to emphasize our concern about the role of SOC, we only considered the net
343 amount of accumulated POPs in soil while still capturing the overall impact of soil
344 fraction properties, particularly SOM, which serves as a carbon and energy source for

345 microorganisms, influencing their community structure and abundance.

346

347 **3.3 Exploring the links between compound properties and factors influencing the**
348 **soil burden**

349 To identify the critical internal influences, an interaction network was conducted
350 using the physicochemical properties of the POPs compounds. The 35 sets of POPs in
351 the soil sample data were divided into four groups, according to the SOC contents
352 (TG1–4) (TG1<1.5%, TG2=1.5%–2.5%, TG3=2.5%–8.5%, TG4>8.5%, Kruskal-
353 Wallis (K-W) test, $p<0.05$). Nine properties were found to be significantly correlated
354 with the distribution of the target POP congeners (Figure S6a). Of these, the
355 octanol−water partition coefficient (K_{OW}), organic-carbon-based partition coefficient
356 (K_{OC}), and octanol−air partition coefficient (K_{OA}) were strongly related to all four
357 groups (Figure S6a). The receiver operating characteristic (ROC) curves were plotted
358 to compare the performance for each property, with $\log K_{OW}$ had the highest
359 performance with an ROC area of 0.927, followed by $\log K_{OA}$ (0.863), and $\log K_{OC}$
360 (0.701) (Figure S6b). This highlights the importance of considering these compound
361 properties as potential driving factors affecting the distribution of POPs in soils.

362 To investigate the integrated effect of these drivers promoting compound retention
363 in soils after LRAT/deposition inputs, the distributions of the five POPs groups with a
364 MAOC-normalized concentration were analyzed alongside corresponding
365 physicochemical properties (Figure 3). Most previous studies focused on fixed pair
366 parameters to evaluate the persistence and potential of LRAT[55, 56], (i.e., K_{AW} and K_{OA}
367 [57, 58]) with a small range of domains tested with legacy POPs[3, 35] (shown inside the green
368 and violet box in Figure 3). With other emerging chemicals being manufactured and
369 used, it is difficult to reflect the environmental fates of a widened class range using
370 existing criteria. In this work, a widened range of POPs also have the potential to
371 survive after LRAT and enrich in the remote regions, including highly adsorbed and
372 relatively hydrophobic components ($\log K_{OA}>10$ and $-4<\log K_{AW}<0$), such as LCCPs
373 (Figure 3A). These low-volatility and highly adsorbed components may have a notable

374 impact on the local ecosystem as they correspondingly were estimated to have high
375 bioaccumulation potential ($5<\log K_{OW}<8$)[35]. After the MAOC/SOC (M/S) ratio
376 correction, the distribution was flipped along the diagonal, and ΣPOPs with $\log K_{OA}>10$
377 were more abundant (Figure 3B). This finding indicates that soils with high proportions
378 of MAOM are more likely to bind low-volatility POPs which are more efficiently
379 deposited by precipitation scavenging ($\log K_{AW}<-4$, $\log K_{OW}<5$)[35]. This increases the
380 retention of these POPs in soils. More volatile POPs with low air surface media partition
381 coefficients (alternatively expressed by $\log K_{OA}<10$) will more readily move towards
382 equilibrium with the OM burden in soils on a global scale, independent of the specific
383 components.
384 After reducing the differences in soil types, the level of more hydrophilic POPs
385 ($\log K_{OW}<4$, Figure S7A, B) and those with a higher migration potential ($\log K_{OC}<6$,
386 Figure S7C, D) decreased, while that of more hydrophobic POPs with high sorption
387 characteristics increased. This finding indicates that the more hydrophilic POPs
388 entering soils via wet deposition become more easily bound to MAOM and are then
389 stored in microaggregates. Soil mineral protection may enhance the retention in the
390 soils of those POPs which have undergone LRAT. The gas-particle partition coefficient
391 (K_P) was further introduced into the system with K_{OC} to investigate the dominant dry-
392 deposition conditions. (Figure S7E, F). The overall reduction in the easily transportable,
393 high particle binding fraction suggests that contaminants that settled into the soils via
394 the atmospheric particle phase may be retained in the soils for a long time, due to
395 mineral protection. POPs may be sequestered by soil due to the presence of sinking
396 particulate matter in dry/wet deposition during LRAT.

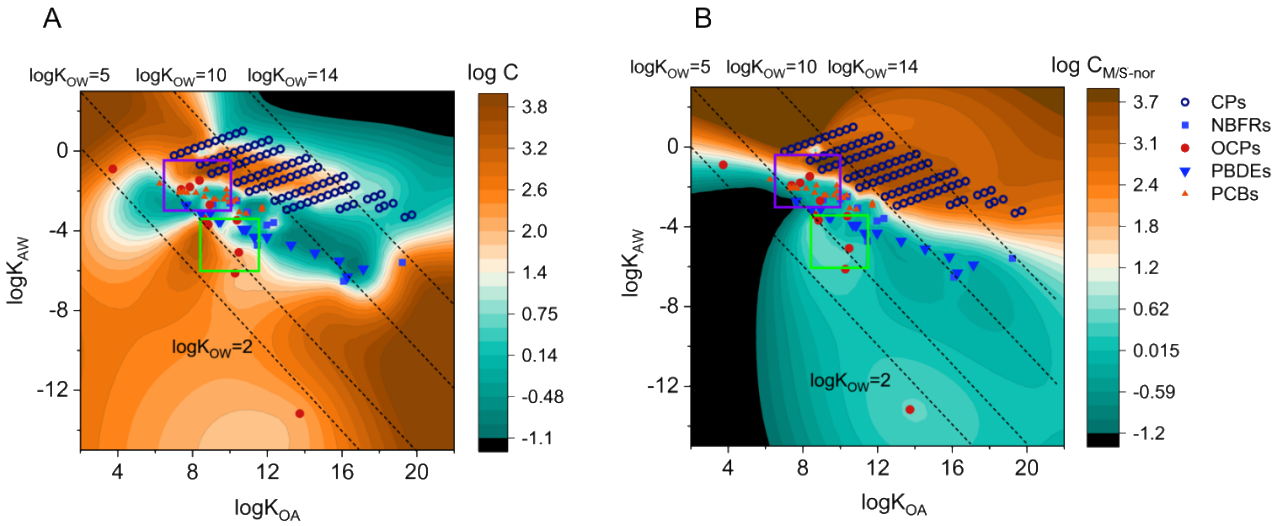

Figure 3. The matrix of chemical space diagrams for the effect of the MAOC/SOC (M/S) ratio and the combination of K_{OA} and K_{AW} on POPs distributions. The steady state processes were characterized by the original (A) or M/S-normalized (B) plots. The diagonal lines indicate a log K_{OW} of 2, 5, 10, and 14, delineating the region of compounds with progressively higher bioaccumulation potential. The green ($8.5 < \log K_{OA} < 11.5$ and $-6 < \log K_{AW} < -3.5$) and violet ($6.5 < \log K_{OA} < 10$ and $-3 < \log K_{AW} < -0.5$) boxes were the estimated combination of properties, in which chemicals can migrate and accumulate in alpine and polar regions, respectively.

3.4 Estimation of the global distribution of POPs in background soils

Based on the above studies, the global burden of legacy and emerging POPs in background soils were estimated using the global soil database[59] (Figure S8). The predicted concentrations of POPs were derived solely based on LRAT inputs, without any consideration of the complex dynamic balance between the soil and atmosphere or any possible influence of nearby point sources. Specifically, the SOC components (MAOC, POC and SOC) were used in a trained RBF model (Table S6) for this estimation on a larger scale (Figure 4). The estimated background soil capture capacities of OCPs, NBFRs, PCBs, PBDEs, and CPs were 0.69–4.57 ng g^{-1}, 43.7–1349 pg g^{-1}, 15.5–257 pg g^{-1}, 8.91–79.4 pg g^{-1}, and 3.80–91.2 ng g^{-1}, respectively. A higher importance of MAOC (0.387) than SOC (0.317) and POC (0.296) was observed (Table S6), proving again that MAOC is key to the soil retention of POPs. The results of the

Monte Carlo simulations show that a high level of uncertainty in the extent to which the parameter of TOC affects estimated result (Figure S9). It is necessary to further confirm the TOC parameter to improve the accuracy of the calculation results. Given the hypothesis mentioned above and some uncertainties (i.e., the deficiency and uneven distribution of SOC data, different soil types[16], and the reliability of SOC mapping efforts under point-/area-source effect interference[60]), these simulated values serve only as rough estimates within a given range for POPs accumulated in idealized background soils.

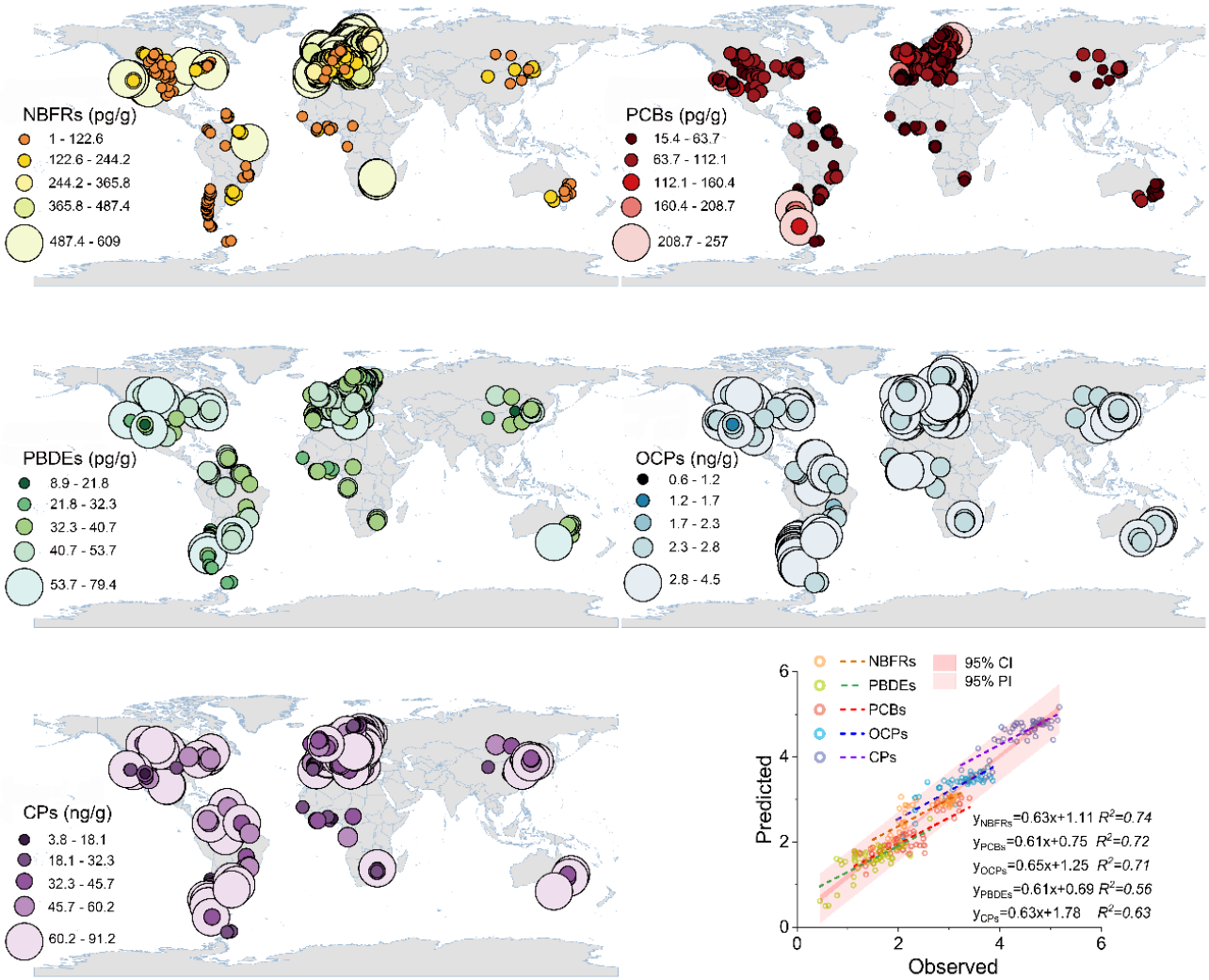

Figure 4. The estimated soil storage capacities of five chemical classes of ΣPOPs at the global scale are based on the RBF model results. The bottom right corner shows the fit

between a combination of the observed and predicted logarithm concentrations of Tri-pole data for the five classes of POPs in this study. The zoom part of Europe and North America can be found in Figure S10

Overall, the estimated capture capacity of soils for the five classes of POPs is concentrated in the mid-latitudes, generally distributed at the junction of wind bands. The stronger soil capture capacities, superimposed on deposition or air-surface exchanges during LRAT, may reduce the total amount of POPs migrating to higher latitudes, and increase the soil burden of POPs at mid-latitudes to a certain extent. Compared with data previously reported, the simulated concentrations of the five POPs classes are generally lower than those observed in the above areas (Table S6 and Figure S11). In other words, the accumulation of POPs at mid-latitudes is higher than the total soil burden that can be estimated using SOM data. This also suggests the presence of POPs from other input pathways, in addition to LRAT input. In particular, active local industrial activities occur in temperate regions (e.g., CPs[49], PBDEs[46]) where unintentional emissions are also continuously released from legacy POPs stocks (e.g., PCBs[61]), making these areas the main distribution of POPs point sources. Based on these results, the burden of POPs in soils may be more severe and direct in temperate-region ecosystems than in high-latitude regions, which therefore weakens the migration of POPs to higher latitudes. Further, the simulated sequestration capacities of soils were separated by soil types (Figure S12). Grasslands and cropland fields show the presence of a high ratio of MAOC/POC in soils, i.e. the storage of carbon in MAOM dominates. However, it saturates after a certain proportion (MAOC/POC about 2–3), and the estimated reserves of POPs no longer occur with it (Figure S12) . Among seven diverse agrotypes, grasslands overall hold higher capture capacities for the five POPs classes, which we refer to here as natural ecosystems and are more carbon-saturated than agricultural systems. The proportion of MAOC in grasslands is relatively high as grasslands have a much higher proportion of below-ground inputs, relatively lower inputs from above-ground fallout, and below-ground inputs, which are mainly root secretions, are more likely to form MAOC. This may allow them to act as substantial C sinks, which can be ascribed to the larger MAOC storages in grasslands[30]. Another

area of concern is the tropical countries, where MAOC is low and the soil capture capacity of POPs is low. E-waste/waste due to human economic activities may exceed the burden of POPs in the soil of this region, thus increasing the risk of secondary migration of POPs.

4. Discussion

With widespread production and use of new chemicals, emerging POPs (e.g., CPs) show a higher concentration range[13], while some legacy POPs still have a global soil burden, despite being regulated or banned for decades[62]. The *source-sink process* mechanisms of POPs burden in background soils are dominated by dynamic processes (i.e., monsoon-driven atmospheric transport, forest filter deposition, and residual soil sinks). Thermodynamically driven *global distillation* and *cold condensation* play secondary roles that may be attenuated with repeated exchange and deposition processes during LRAT[57], and the soil OC pool plays a more and more important role in determining the secondary transport of POPs as time passes.

Soil OC dynamic and ambient meteorological conditions (e.g., the dry and wet deposition rates) can be potential influences that mask global cold condensation trends (Figure 5). Our data suggested that the stabilized soil C pool (MAOC) is a kinetically controlled POPs reservoir in global soils, rather than a thermodynamically equilibrium-driven, secondary source of emissions. Notably, only a small range of organic compounds are considered to undergo cold-trapping[3,35] during LRAT. Here, we suggested that a relatively wide range of contaminants can be captured in remote areas, due to their sequestration by MAOM. Globally, the current soil mineralogical C capacity was 899 Pg C to a depth of 1 m[59], which was estimated to be approximately 600 Gt C retained by reactive minerals, accounting for one-quarter of the total SOC pool[63]. In addition, MAOC is generally less susceptible than POC [31], with turnover even slowing under climate change[32]. Therefore, this high percentage of MAOM in soils can impede the diffusion and secondary release of POPs with media turnover after intercepting them during LRAT. Due to the low microbial utilization of MAOM[22], the delayed biodegradation of POPs may lead to a longer half-life than the model estimates.

In this way, the persistence of POPs in soils is enhanced by MAOM sequestration. Thus, at least for some POPs, the MAOM - as well as subsequent role conversion between sources and sinks - may represent a mechanism for driving their environmental fate under the global fractionation process in soils. With the global climate continually changing[64], weight-of-evidence methods integrating physicochemical factors with stabilized SOM are suggested in the future assessment of persistence and LRAT potential of chemicals. These modifications can better explain the potential of some POPs to undergo LRAT diluted by soil retention, as well as help to understand the processes affecting their fate and transport at regional and global scales.

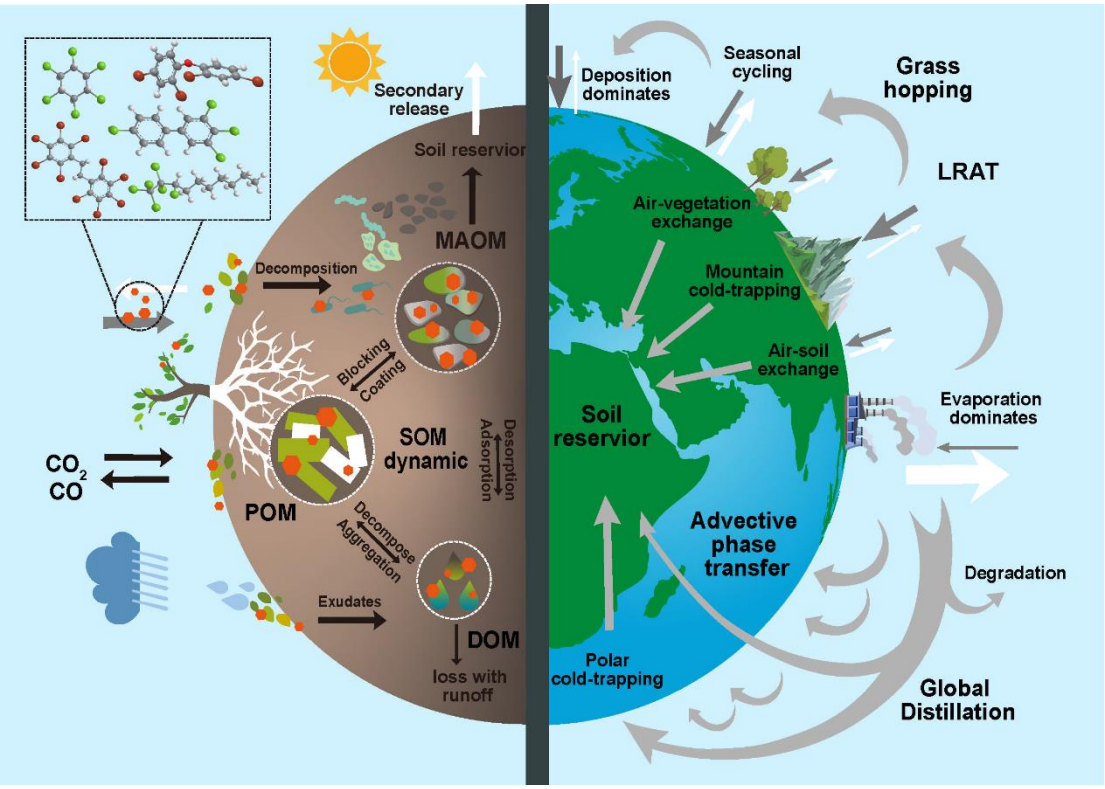

Figure 5. The coupling effects of SOM dynamics and the global cycling of POPs. The right side of the figure is developed by Wania and Mackay (1996), while the left side elaborates on some of the complex processes discussed in this paper, which will strongly influence the pool of POPs available to participate in the air-surface exchange processes shown on the right side.

In addition, SOM is a dynamic OC pool, of which the fraction changes under perturbations of human activities (e.g., implementation of *double carbon* and *dual control* policies) or climate crisis[64], especially in ecologically vulnerable areas. Sub-regions of the polar areas and the TP are vulnerable to ongoing climate changes and

have been reported to be experiencing the most rapid rates of warming worldwide[65, 66].Wang et al found that a large amount of halogenated pollutants which are difficult to extract are stored in frozen soil, and most of them are stored in the form of physical closure[67]. Combined with their work, we hypothesize that MAOM carbon banks may store more POPs in non-extractable states.

In the context of global change challenges, future studies are needed to integrate POPs into soil carbon cycle models. This will enable a more holistic and sophisticated approach to the risk for soil sequestration and release of POPs from changes in soil carbon pools.

Currently international mechanisms, including the Stockholm Convention, have been working on interactions between POPs' fate and climate change, which were included in the scope of topics since 2011[68]. Until now the role of soil has been limited to considering a single long-term reservoir [69-71] , and soil OC turnover is not included in the scope of discussion. Better regulatory policies and decision-making are greatly facilitated by the construction of global POPs estimations, and ecosystem models describing the transport, deposition, and exposures in different media. Based on this work, we suggest the future inclusion focusing on the dynamics of SOM fractions induced by climate change as a topic of discussion on the relevance for the environmental fate and changes of POPs.

Description of Supporting Information

Supporting Information includes one text, twelve figures and six tables.

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Supporting Information.

The following files are available free of charge.

Detailed description of materials, pretreatment process, and instrumental analysis;

Distribution of the sampling sites and characteristics of the soil samples across Tibet Plateau (Table S1, S4), Antarctic (Table S2) and Arctic (Table S3) regions;

Comparison between the present study (pg/g) and various studies from remote areas (Table S5);

Model details and validation for RBFANN (Table S6);

Spatial distribution and Logarithmic concentration of legacy and emerging POPs in Antarctic (a) and Arctic (b) regions (Figure S1);

Spatial distribution and homologous characteristics of the 5 POPs in soil of Tibetan Plateau (Figure S2);

Comparison of the minimum and maximum concentrations of ΣPOPs in the Tri-polar regions (Figure S3);

The relationship between environmental factors and POPs level in soil (Figure S4);

(a) The coefficient of variation (C_V) of different environmental factors from structural equation model (SEM) model, (b-f) the Monte Carlo simulation results of model calculation (Figure S5);

(a)The co-occurrence patterns between the distribution of target POPs components and physicochemical properties of each component; (b) The receiver operating characteristic (ROC) curves of each property (Figure S6);

The matrix of chemical space diagrams showing how the MAOC/SOC (M/S) ratio and properties affect ΣPOPs (Figure S7);

Special pattern of normalization SOM data obtained from the Soil Profile Database (Figure S8);

(a) The coefficient of variation (C_V) of different SOC fractions from RBF model, (b-f) the Monte Carlo simulation results of model calculation (Figure S9);

The enlarged picture for estimated soil storage capacities of five ΣPOPs in the Europe and North America based on the RBF model results (Figure S10);

(a) Spatial and concentration distributions of target POPs level reported previously and (b) estimated five POPs by RBF model in this work. The data sources of (a) in this figure are shown in Table S5 (Figure S11);

Separation of simulated storage capacities for five target POPs by soil types (Figure S12).

AUTHOR CONTRIBUTIONS

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Writing—original draft: L.J.

Writing—review & editing: J.T.L., K.C.J., Y.W.W., and G.B.J.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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