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Organic matter imports to the Atacama Desert using polycyclic aromatic hydrocarbons as tracer

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ABSTRACT

The Atacama Desert, despite its extreme hostile conditions, still harbours traces of life in its dusty surface. Until now, it remains open whether organic molecules found in topsoils have their origin from in-situ biotic processes in the hyper-arid core, or whether and to which degree they stem from outside the desert and its border regions. In order to trace atmospheric organic matter input, we analyzed the concentration of polycyclic aromatic hydrocarbons (PAHs) which almost exclusively derive from combustion processes and compared this to total organic carbon (TOC) concentrations. We sampled topsoil (0–15 cm depth) in one coastal (Paposo) and one inland aridity transect (Aroma) and compared them to atmospheric dust collected over two months in the hyper-arid core of the desert (Yungay). We found TOC and PAHs in all desert soil and dust samples. The sum of 18 analyzed PAHs ranged from 0.001 to 0.31 ng g^{-1} in the Paposo transect and from 0.14 to 0.78 ng g^{-1} in the Aroma transect, with moderate correlation ($R^2 = 0.68$, $p < 0.05$) between TOC and PAHs. Measured PAH concentration in the dust was up to 5000 times higher and varied between 0.45 and 5.31 ng g^{-1}. PAHs contributed with up to 0.25% to the TOC in dust and with 0.1% to the TOC in soils. This indicates that not all organic matter found in deserts is a trace for life defying extreme conditions, as it can also stem from sources outside or at the borders of the desert. We hence recommend verifying the origin of organic matter found in regions at the dry limit and highlight the overall presence of atmospheric pollutants on earth.

1. Introduction

Hyper-arid areas such as the Atacama Desert in northern Chile accumulate significant amounts of dust, which also contains a suite of organic compounds (Ewing et al., 2007; Arenas-Díaz et al., 2022). Indeed, several studies found that molecular traces of life are abundant in this extreme environment (Kusch et al., 2020; Mörchen et al., 2019, 2021; Knief et al., 2020; Fuentes et al., 2022). However, the allocation of the sources of such organic matter (OM) and whether it derived from sources in- or outside the desert was not yet verified.

OM has been detected in the desert ranging between 1 and 50 g kg^{-1} in the upper centimeters of soils and strongly depending on gradients of aridity (Valdivia-Silva et al., 2012; Mörchen et al., 2019). In Yungay, a completely remote site which was intensively studied for years (e.g., Connon et al., 2007; Pfeiffer et al., 2018; Azua-Bustos et al., 2017; Fuentes et al., 2022), and also known as one of the driest regions of the desert, molecular analyses revealed the presence of formic acid and benzene (Navarro-González et al., 2003), as well as of amino acids (Buch et al., 2006; Amashukeli et al., 2007). Although it was already expected that the dispersal of OM in hyper-arid areas of the desert is partly controlled by aeolic activity (Valdivia-Silva et al., 2012), the origin of OM in the hyper-arid parts of the desert remained open.

Some organic molecules, such as polycyclic aromatic hydrocarbons (PAHs), are of global relevance, very stable over time and can be produced either by abiotic sources like combustion processes of anthropogenic emissions, wildfires (Simoneit and Mazurek, 1982; Wilcke, 2000; Lima et al., 2005; Patel et al., 2020) or by biotic sources like termites or plants (Wilcke et al., 2000, Wilcke et al., 2003, Kovacs et al., 2008).

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These are major sources for combustion-derived PAHs at the borders of the desert, the coast (Cabrera-Tobar et al., 2016) and the high Andean region (Puna, Paramos and Yungas) which are home to highly flammable ecosystems (Bradley and Millington, 2006; Roman-Cuesta et al., 2011, 2014). In the vegetation-free hyper-arid core of the desert only few roads, mining activities and thermoelectric power plants (formerly coal-fired, now oil and natural gas) represent sources for combustion-derived PAHs. Whether residues of combustion processes were imported into the central Atacama Desert is the main aim of our study.

Physico-chemical properties of PAHs control their fate in the environment and may also help attributing them to specific sources. Low molecular weight PAHs with 2 and 3 aromatic rings are preferably transported in the gaseous phase over long distances. With increasing molecular weight and ring number PAHs tend to bind to particles and transport distances decrease (Yang et al., 1991; Yunker et al., 2002). Many PAHs have source diagnostic potential. They are specific for traffic, industrial or biomass combustion, e.g., the ratio of fluoranthene to fluoranthene plus pyrene differentiates PAHs of vegetation fire from traffic emissions (Yunker et al., 2002). In the Yungay region of the central Atacama Desert, Stockton et al. (2009) already detected PAHs in the μg g^{-1} range. However, the sources of these PAHs and whether they were deposited ubiquitously throughout the Atacama Desert was not yet investigated.

The development of atmospheric dust, its composition and distribution along the west coast of Chile is controlled by a distinct climate and morphology. At the border to the Pacific Ocean, the Coastal Cordillera forms a prominent longitudinal orographic barrier separating the approx. 1–3 km wide coastal plain from the broad Central Valley of the Atacama Desert (Walk et al., 2020). The entire coast receives precipitation from fog (Cereceda et al., 2008) that can penetrate into the hinterland moving along topographic corridors. Vegetation growth, however, is mainly restricted to the region of the Cordillera (Ruhm et al., 2020; Mörchen et al., 2021) and fire frequencies are reported to be low for this area (Thonicke et al., 2010). Hence, anthropogenic activities such as thermoelectric power plants at the coast were expected to form the main source of PAHs.

We aimed at identifying the contribution of OM from external sources in the central Atacama Desert using PAHs as tracer. We hypothesized that PAHs from natural fires and anthropogenic combustion processes indicate whether OM was brought into the central desert via atmospheric transport and that such external inputs contribute ubiquitously to OM findings in the Atacama Desert. We analyzed PAHs in desert topsoils along two west-east transects in the south-western (Paposo) and north-eastern (Aroma) part of the Desert. This was complemented by atmospheric dust sampling in the hyper-arid core of the Atacama Desert (Yungay).

2. Material and methods

2.1. Topsoil and dust sampling

Topsoil sampling was performed in October 2016 and March 2017 as described by Mörchen et al. (2019). The study sites were selected within two gradients: one to the south of the desert at Paposo covering the orographic transition from the coast over the cordillera and into the hyper-arid region of the desert, the other to the north at Aroma reaching from the foot slopes of the Andes into the central depression of the Atacama Desert. Both sample sets covered gradual changes in aridity and vegetation cover, and were distant from anthropogenic emissions such as roads and mines (Fig. 1). The Paposo transect (25.1°S) is located at the west-coast of Chile, close to the small village Taltal, ranging from 800 to 2000 m a.s.l (Mörchen et al., 2019, 2021). Here, the formation of fog over the Pacific's cold water spends moisture into the Atacama Desert (Rundel and Mahu, 1976). Mean annual temperature (MAT) varied from 16.7 to 18.1 °C and mean annual precipitation (MAP) ranged from 0 to 6.7 mm (Quade et al., 2007). This transect also displays a distance to typical PAH emission regions such as roads with permanent traffic of cars and trucks like the Panamericana, dirt roads leading to mines, and ship emissions which are all situated at the coast (Fig. 1). The Aroma transect is located further north (19.4°S). MAT varied from 13.5 to 17.7 °C and MAP from 0 to 43.6 mm (Gobierno de Chile, Ministerio de obras publicas, direccion general de aguas, division de estudios y planificacion, 2013). With increasing distance to the coast, distance to Andes and respective wildfires becomes closer.

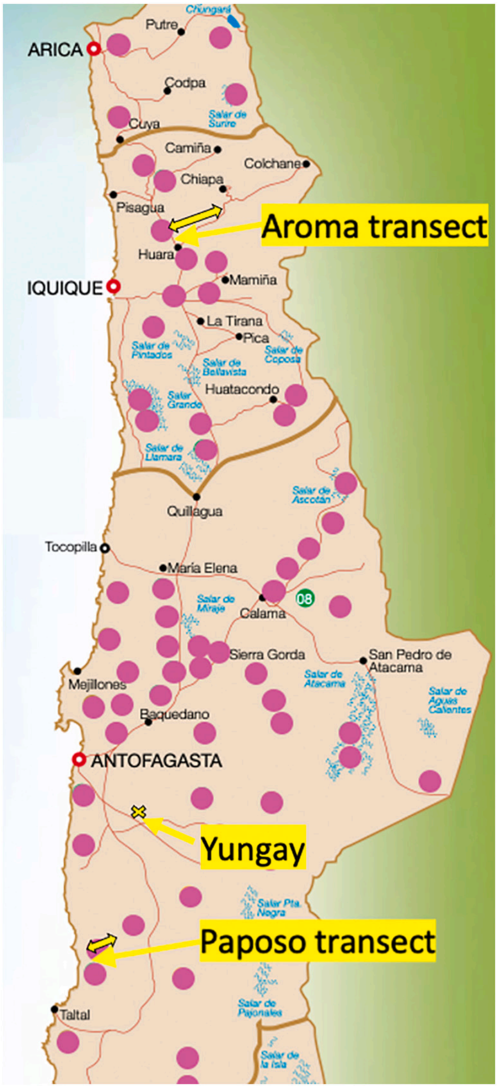

Fig. 1. The map is showing the mining areas (purple circles) near our transect sites (Paposo and Aroma) and the dust sampling site (Yungay) (Map modified after: https://biblioteca.sernageomin.cl/opac/DataFiles/mapa-minero-de-chile.pdf). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Degree of soil formation was characterized by humus accumulation, mineral weathering and salt precipitation (sulfates, carbonates) at the coastal sites and the foot slopes of the Andes according to the World Reference Base for Soil Resources (World Reference Base for Soil Resources, 2022). In the coastal area (Paposo) Cambisols and Gypsisols, were formed under sparse vegetation within 23 km from the coast, and at Aroma, from 86.8 km from the coast on, Cambisol formation has taken place under the first thin vegetation of the Andes (see Mörchen et al., 2021). All other surfaces at arid and hyper-arid sites showed no vegetation but only varying degrees of gypsum precipitation and abiotic surface crusts. The sampling was performed according to a nested design, as originally described in Youden and Mehlich (1937) and already applied to the sample set published in Mörchen et al. (2019). Briefly, the Paposo transect consisted of 10 sites and the Aroma transect of 12 sites. At each of these sites we took 6 topsoil samples (0–15 cm) in a random design approach with maximum 50 m to minimum 2.5 m distance between samples. We pooled these samples and homogenized them for PAH analysis. In addition, we sampled dust from November 2018 until January 2019 in Yungay (24.6°S), located in the hyper-arid core of the Desert (Fig. 1). Here, it has been shown that hyper-arid conditions have been prevalent since at least 15 million of years (Ewing et al., 2006). MAT is around 16 °C, and MAP is <2 mm (Ewing et al., 2006). Dust collection was done using a glass marble collector (Supplementary Information Fig. S1, S2; Goossens and Rajot, 2008). Marbles prevented already accumulated dust from being blown out. Four dust samples were collected at this site by emptying the collector four times every two weeks.

3. Analytical methods

3.1. Analyses of topsoil samples

3.1.1. Total organic carbon

TOC concentrations in soils were detected by temperature-dependent differentiation of total carbon (DIN 19539, Soli TOC, Elementar Analysensysteme, Hanau, Germany) and were published in Mörchen et al. (2019). Briefly, the dynamic temperature ramping method started at 100 °C and was ramped with a rate of 90 °C / minute to 400 °C for the determination of labile organic carbon (LOC). The temperature was then kept at 400 °C for 430 s. With 90 °C / min the sample was heated to 600 °C for detection of residual organic carbon (ROC) and to 900 °C for total inorganic carbon. Total organic carbon was calculated as the sum of LOC and ROC. Dust samples were analyzed by dry combustion in an elemental analyzer (Micro Cube, Elementar Analysensysteme, Hanau, Germany). All values were reported as the mean of duplicate analysis. To check for optimum performance of the instrument a standard of calcium carbonate (Calciumcarbonat, Elementar Analysensysteme, Hanau, Germany) and a soil standard (Bodenstandard Nr. 2, HEKA tech GmbH Analysentechnik, Wegberg, Germany) were analyzed prior, between, and after each run for Soli TOC and Acetanilide (Elementar Analysensysteme, Hanau, Germany) for the Micro Cube. All samples had a precision of >90%.

3.1.2. Polycyclic aromatic hydrocarbons

Soil and dust samples were extracted using accelerated solvent extraction (BÜCHI, E-916, Switzerland) with dichloromethane and methanol (DCM: MeOH, 9:1 *v*/v, extraction parameters: static time 40 min at 120 °C, 110 bar, 2 extraction cycles). Extracts were purified using solid-phase extraction (SPE): glass cartridges were filled with 2 g activated silica gel (1 g of 0.063–0.2 mm coarse mesh and 1 g of 0.015–0.04 mm fine mesh silica gel) and conditioned with *n*-hexane. Extracts were loaded and consecutively eluted with 4.5 ml *n*-hexane, 4 ml DCM: *n*-hexane (2:1, *v*/v), and 5 ml MeOH, yielding aliphatic, aromatic and hetero compound fractions, respectively. PAH recovery after sample clean-up was low but reproducible at 38 ± 9% and tested using perlyene-D12 and pyrene-D10 as internal standards. We decided to not correct for this as our research question aimed at identifying presence or absence of PAH in the desert rather than at their quantification.

Quantification of PAHs was carried out using gas chromatography coupled with mass selective detection (Agilent Technologies GC7890B, Germany, MS/MS, EVO3, Chromtech, Germany) in selected ion mode (SIM *m/z* 128, 152, 154, 166, 178, 192, 202, 206, 228, 252, 264, 276, 278) and equipped with a HP-5MS column (30 m × 250 μm × 0.25 μm, injection mode: 250 °C, 54 ml/min inlet flow, 9.4 psi pressure on column, helium as carrier gas; oven program: 80 °C held for 2 min, to 250 °C at 10 °C/min, to 320 °C at 3 °C/min). An external standard containing 16 PAHs with increasing molecular weight (QTM-PAH mix, Sigma-Aldrich, Germany) was used for compound identification and quality control. Up to seven parent and four alkylated PAHs were reliably detectable in samples (Supplementary information, Table S1).

3.2. Statistical analyses

The Shapiro-Wilk test was selected for assessing normal distribution within the sample sets. Plots and "Kendall" correlations were performed using R-Studio. Linear regression was calculated using Sigma Plot 13 for Windows.

4. Results

Organic carbon concentrations were already published in detail in Mörchen et al. (2019). In brief, they were low in the central part of the desert ranging between 0.4 and 0.7 mg g^{-1}. At the beginning of the two aridity gradients that have been sampled, i.e., were vegetation growth was still visible, TOC was higher around 16 mg g^{-1} close to the coast (Paposo transect) and 1.2 mg g^{-1} close to the Andes (Aroma transect).

In Paposo, we found acenaphthylene, acenaphthene, fluorene, phenanthrene, fluoranthene, pyrene, perylene, 3-methylphenanthrene, 2-methylphenanthrene, 9-methylphenanthrene, 1-methylphenanthrene. The concentration of individual PAHs, which are higher than the limit of detection (phenanthrene, sum of methyl-phenanthrenes, fluorene, pyrene, perylene), ranged between 0.001 and 0.31 ng g^{-1} (Fig. 2). The decrease in PAH concentrations was paralleled by a decrease in MAP from 7 to 2 mm (2 to 7 km distance to coast). However, also where no precipitation was reported, PAHs were detectable and the sum of PAH concentrations had a mean of 0.2 ± 0.1 ng g^{-1} (from 10 to 23 km distance to the coast) (Fig. 2). The PAH composition at all sites was dominated to 35–60% by phenanthrene, with two exceptions where pyrene had higher contributions (34% pyrene at <0.1 mm MAP and 15 and 23 km distance to coast), followed by fluoranthene (15–25%), the sum of methyl-phenanthrenes (0–10%), and perylene (0–5%), respectively. The portion of total PAHs on organic carbon (OC) varied between 1 and 664 μg g^{-1} with lowest values at sites with highest TOC concentrations, i.e., where some vegetation was growing and lead to a weak humus formation (7 to 2 mm MAP in 2 to 7 km distance to coast) (Fig. 2).

Within the Aroma transect, we detected the same PAHs as for Paposo. Starting at the slope of the Andes, PAH concentrations were overall higher than in Paposo and the sum of PAHs ranged between 0.14 and 0.78 ng g^{-1} (Fig. 2). Within this transect there was high variation as displayed in Fig. 2. Concentrations were lower at sites with 47, 53 and 62 km distance to the coast and strikingly higher at 58 km distance to the coast (all sites with MAP <0.1 mm) (Fig. 2). The individual PAH concentrations differed as well, so these sites were dominated to 32–70% by phenanthrene with four exceptions where pyrene had higher contributions of 32–46% (at 47, 73, 86 and 87 km distance to coast), followed by fluoranthene (5–31%), the sum of methyl-phenanthrenes (3–28%), and perylene (0–7%) respectively. In Aroma the portion of total PAHs on OC was generally higher than in Paposo and varied between 280 and 1128 μg g^{-1} (Fig. 2). Here, with some exceptions (at 50, 57 and 59 km distance to coast; all MAP of <0.1 mm) an increasing trend of the portion of total PAHs on OC was visible up to a distance of 81 km distance to coast (11 mm MAP). Then at 85 and 87 km distance to coast corresponding to a MAP of 43 mm, the portion of total PAHs on OC decreased again to a range comparable to the beginning of the transect (Fig. 2).

In the Paposo transect, there was no significant correlation between TOC and PAHs in soil samples. However, a moderate correlation of TOC to fluoranthene and perylene was noted (R^2= 0.55 and 0.60, respectively, Fig.S4, supplementary information). In the Aroma transect, there was a moderate correlation between TOC and fluoranthene and pyrene ($R^2 = 0.70$ and 0.63, respectively). PAHs showed a covariation between each other in the entire desert (Fig. S4, supplementary information). For instance, the R^2 of fluoranthene and pyrene compounds varied between 0.82 and 0.93 ($p < 0.05$) in Aroma and Paposo (Fig. S4, supplementary

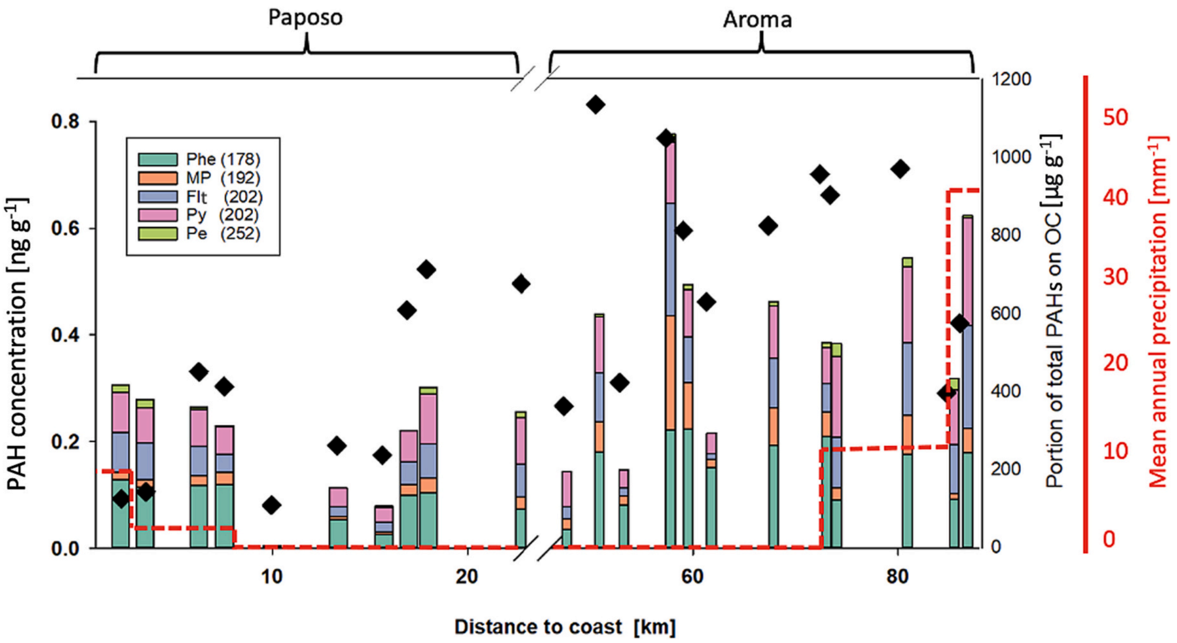

Fig. 2. Polycyclic aromatic hydrocarbon (PAH) concentrations and the portion of total PAHs on organic carbon (OC) (black rectangles) as related to mean annual precipitation (red, third y-axis) and distance to coast (x-axis) in soils from Paposo (2 to 23 km from coast) and Aroma (47 to 87 km from coast). Phe = phenanthrene, MP = sum of methyl-phenanthrenes, Flt = fluoranthene, Py = pyrene, Pe = perylene. Note i) the gap in the x-axis which displays the gap in distance to coast between the two transects and ii) that only data is listed which is above the detection limit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

information). At the same time, phenanthrene and sum of methyl-phenanthrenes had a R^2 value of 0.66 and 0.78 ($p < 0.05$) in Paposo and Aroma, respectively (Fig. S4, supplementary information). There was no correlation between the sum of phenanthrene and sum of methyl-phenanthrenes and fluoranthene and pyrene ($R^2 = 0.34$). However, TOC did very weakly correlate with PAHs in Paposo ($R^2 = 0.44$ ($p < 0.05$) and ($R^2 = 0.52$ ($p < 0.05$) in Aroma.

In dust samples we detected phenanthrene, acenaphthene, pyrene, fluoranthene, fluorene, chrysene, benzo(*b*)fluoranthene, benzo(*a*)pyrene, 3-methyl-phenanthrene, 2-methyl-phenanthrene, 9-methyl-phenanthrene, and 1-methyl-phenanthrene. PAH concentrations in atmospheric dust were nearly 10 (but up to 5000) times higher than in topsoils (Fig. 3). Dust samples were all dominated by pyrene (contributing 43–50% to total PAHs), followed by fluoranthene (25–30%), chrysene, benzo(*b*)fluoranthene and benzo(*a*)pyrene. The ratio of phenanthrene/pyrene in dust was 4 times lower than in Aroma and 5 times lower than in Paposo, which is mainly related to significantly higher pyrene concentrations in dust (up to 8 times more in Aroma and

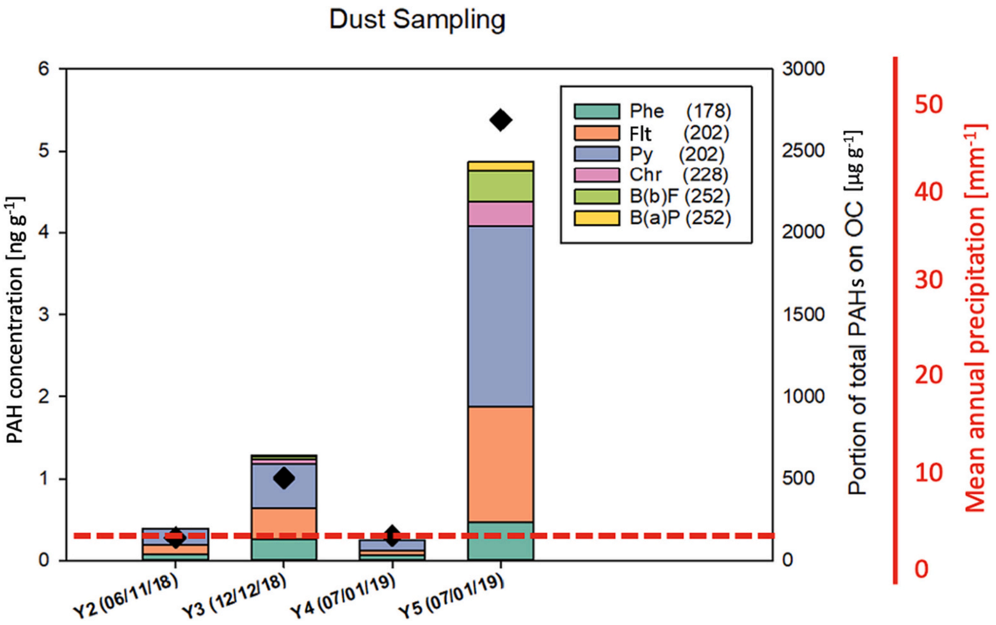

Fig. 3. PAH concentrations and portion of total PAHs on OC in dust collected in Yungay (central Atacama, Phe = phenanthrene, Flt = fluoranthene, Py = pyrene, Chr = chrysene, B(*b*)F = benzo(*b*)fluoranthene, B(*a*)P = benzo(*a*)pyrene). Displayed in red is mean annual precipitation in Yungay. Please note that only data is shown which is above the detection limit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

14 times more than in Paposo). Dust samples generally contained higher relative proportions of high molecular weight PAHs compared to topsoils. Portion of total PAHs on OC in atmospheric dust varied between 140 and > 2500 μg g^{-1} within the 2 months of sampling (Fig. 3).

5. Discussion

5.1. Abundance of PAHs in the Atacama Desert

The concentrations of PAHs in soils can vary in the order of several magnitudes, depending on climate, land-use, soil biogeochemical properties, and distance to individual emission sources (Wania and Mackay, 1996; Meharg et al., 1998; Bandowe et al., 2021). Regarding the distance, it was found that in sites which are far from any combustion source like Greenland ice or in Argentinian Pampa soils contained only around 0.001 ng g^{-1} PAHs (Taffrezo et al., 1994), or concentrations varied between 2.4 and 38 ng g^{-1} (Wilcke et al., 2014), respectively. Concentrations were higher in other more semi-arid regions like in Tajikistan (17 to 258 ng g^{-1} PAHs) or Kyrgyzstan (52 to 9440 ng g^{-1}; Li et al., 2020). Under different climates in less remote sites, soils can also contain up to 500 ng g^{-1} PAHs (as shown for Europe, see Bläsing et al., 2016, 2017) or up to 2670 ng g^{-1} when they are close to a copper smelting site (as shown for arid areas in Uzbekistan; Bandowe et al., 2021). Our results from the Atacama region were just in between the PAH concentrations reported for remote areas and ranged between 0.001 and 0.78 ng g^{-1} with an average of 0.3 ± 0.2 ng g^{-1} PAHs, i.e., being two magnitudes higher than in Greenland ice cores and two magnitudes lower than in remote European soil. While in all mentioned publications it was found that PAHs systematically decrease with distance to the emission sources (Wania and Mackay, 1996; Meharg et al., 1998), we did not observe this for the transects directing into the hyper-arid core of the Atacama Desert. However, in-situ biological production of PAHs in the hyper-arid part of the desert seems unlikely, as this process has so far been described mainly for higher organisms (e.g., Wilcke et al., 2000, Wilcke et al., 2003, and Wilcke, 2007, Kovacs et al., 2008), which are not present in the hyper-arid core. Microbes seem to be limited to forming smaller aromatic moieties such as aromatic amino acids (e.g., Noda and Kondo, 2017). Hence, the partially elevated abundance of PAHs in hyper-arid regions may be related to import processes and accumulation of PAHs in the absence of life.

5.2. Degradation and preservation of PAHs in the Atacama Desert

The Atacama Desert was said to have, in contrast to more moist regions, a very much reduced potential for microbial degradation of OM due to the lack of moisture (Wilhelm et al., 2016; Mörchen et al., 2021). Within two east-west transects through the desert we observed a clear decline of TOC, paralleling moisture and vegetation loss (Mörchen et al., 2021; Sun et al., 2023). It seems plausible that TOC concentrations in the desert were mostly controlled by vegetation growth. PAHs, instead, did not follow the vegetation gradient, indicating that other processes such as atmospheric transport and deposition as well as preferential preservation and accumulation played a role (Arenas-Díaz et al., 2022) .

The excellent preservation of PAHs in the hyper-arid core of the desert is of little surprise. However, some PAHs are prone to photooxidation, such as anthracene, benzo(*a*)anthracene and benzo(*a*)pyrene (Behymer and Hites, 1985), and these were completely lacking in Atacama soil samples. Only the atmospheric dust contained benzo(*a*)pyrene (Fig. 3) indicating rather recent sources or an effective stabilization of photo-sensitive PAHs when adsorbed to particles as previously observed by Krauss and Wilcke (2003). Also, the concentration of TOC in dust was about 4 times higher compared to soils, but the portion of total PAHs on OC was comparable (Fig. 3). This indicated that once deposited rather a dilution with minerogenic particles appeared, and that organics, including PAHs, remain stable in the Atacama Desert for long periods of time.

5.3. Sources of PAH and organic matter in the Atacama Desert

As PAHs derive almost exclusively from combustion processes, we expected that they reflect overall atmospheric transport into the Desert. At the coast, fog transports and deposits aerosols from the Pacific Ocean some kilometers into the Desert up to an elevation of about 1000 m a.s.l. of the Precordillera mountains (Rundel et al., 2007). This so-called fog barrier is reached in our study at 10 km distance from the coast in the Paposo transect. Here we found constantly elevated levels of PAH concentration (Fig. 2), indicating that fog supported the transport of PAHs associated to aerosol particles. But also, within the hyper-arid, central parts of the desert, colloidal particle composition has been related to atmospheric inputs (Moradi et al., 2020), indicating that also longer distance transport processes may play a role for bringing materials into deserts.

In order to get insights into the source regions of PAHs deposited into the Atacama Desert, we investigated gradients from potential sources into the central desert. We found no systematic decrease with distance to typical PAH emission regions such as the more vegetated coast, the Panamericana, towns like Taltal, and dirt roads leading to mines. Sampling sites at Aroma, however, being closer to the Andes and its wildfire activities, had up to two times more PAHs compared to the coast at Paposo (Fig. 2). Residues from wildfires, or anthropogenic biomass burning seemed to contribute most to the soils in the eastern Atacama Desert.

The composition of PAH can be related to its sources, most prominently to anthropogenic activities. We found high portions of perylene, pyrene and fluoranthene (Fig. 2) in the first kilometers from the coast, which have been related to diesel combustion (Cadle et al., 1999; Schauer et al., 2001; Riddle et al., 2007; Wei et al., 2015). The ratio of fluoranthene/fluoranthene+pyrene was around 0.5 ± 0.1 and indicated that PAHs most likely originate from a mix of petroleum and biomass combustion (Yunker et al., 2002; Abakumov et al., 2020). This agrees with Li et al. (2020) who found that in remote areas contribution of PAHs from anthropogenic activities are much likely, indicating that also in the Atacama Desert at least these PAHs were transported inside the desert.

Wildfires contribute up to 14% to the global PAH budget, from which at least 51% derive from wildfires in South America alone (Shen et al., 2013). Abakumov et al. (2020) showed that wildfire-affected soils in Siberia contributed to phenanthrene, fluoranthene, pyrene, and chrysene contents in soil. The deposition of these PAHs was also found in other high-altitude locations such as in Greenland and the Canadian Arctic (Halsall et al., 2001; Patton et al., 1991; Taffrezo et al., 1994). Therefore, we assume that PAHs found in the Atacama are most likely derived from both, long-distance transported anthropogenic emissions and from wildfires.

Some PAHs such as naphthalene, perylene and phenanthrenes may also have biological origin. Higher land plants from trees to orchids, as well as insects like ants and termites were shown to contain PAHs (Krauss et al., 2005, Kovacs et al., 2008). Also, bacteria are able to produce aromatic compounds (Gosset, 2009), however, a clear attribution to individual parent and methylated PAHs was not made. As the majority of PAH studies in soil science assume that PAHs stem from fossil and combustion sources, we expect that the contribution of biologically produced PAHs in the Atacama Desert should be low, although it cannot be fully excluded. In our study we found that perylene contributed 4% to soil PAHs and 0% to PAHs associated to atmospheric dust particles and take this as indication that in hyper-arid regions PAHs of biological origin may occur in small amounts and an in-situ formation of these cannot be fully excluded. In comparison, the majority of PAHs found in hyper-arid regions should stem from the desert margin and other sources outside the desert.

The primary origin of OM in dust and soils of the Atacama Desert may at least partially be located outside the hyper-arid regions of the Atacama Desert. Indeed, Arenas-Díaz et al. (2022) could show that the main source of primary aerosols was outside the desert pointing at a considerable contribution of OM and PAH from external sources. All atmospheric dust we sampled contained about 4 times more TOC compared to the soils in the hyper-arid core of the desert, while PAH concentrations were up to 5000 times higher. We take this variability in PAHs as further indication for their anthropogenic origin and conclude that also OM stemmed mostly from regions outside the central Atacama Desert.

6. Conclusion

By using PAHs as a tracer for atmospheric particle inputs into the desert, we were able to show that organic matter from outside the desert is deposited in the whole Atacama Desert region. We found organic carbon concentrations of 2.2 ± 0.6 mg g^{-1} in atmospheric dust and 1.6 ± 0.14 mg g^{-1} in desert soils. Up to 0.25% of this consisted of PAHs, most of them stemming from sources outside the desert. Although an in-situ biological formation of a small portion of PAHs such as perylene cannot be fully excluded. We finally show that PAHs enter hyper-arid regions via the atmosphere and contribute to organic matter accumulation in mostly bare regions.

Declaration of Generative AI and AI- assisted technologies in the writing process

During the preparation of this work the author(s) used no generative AI or AI- assisted technologies.

CRediT authorship contribution statement

Ramona Mörchen: Resources, Visualization, Writing – original draft, Writing – review & editing. **Humay Rahimova:** Data curation, Formal analysis, Investigation, Methodology, Writing – review & editing. **Barbara Fuentes Siegmund:** Conceptualization, Resources, Visualization, Writing – original draft, Writing – review & editing. **Franco Arenas Diaz:** Conceptualization, Resources, Writing – original draft, Writing – review & editing. **Bol Roland:** Conceptualization, Investigation, Writing – original draft, Writing – review & editing. **Eva Lehndorff:** Conceptualization, Data curation, Methodology, Project administration, Supervision, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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