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Temporal trends of persistent organic pollutants (POPs) and perfluoroalkyl substances (PFAS) in Adèlie penguin (*Pygoscelis adeliae*) eggs from the Ross Sea (Antarctica), including their relationship with climate parameters☆

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A R T I C L E I N F O

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A B S T R A C T

Temporal trends of contaminants represent an important tool to evaluate the effectiveness of chemical restriction measures. In this work, 50 eggs of Adèlie penguin (*Pygoscelis adeliae*) collected along the Ross Sea coasts from 1997 to 2021 were analysed for polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), hexachlorobenzene (HCB), *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE), perfluoroalkyl substances (PFAS). Some PCB congeners showed a significantly decreasing trend, whereas HCB and *p,p'*-DDE indicated decreasing but not significant trends, potentially related to the unintentional production of HCB and ongoing use of DDT, even if a contribution from climate-driven remobilisation mechanisms may also play a role. PBDE-47 also indicated a decreasing but not significant trend, which might be explained by the more recent global restriction. PFAS trends agreed with what has been previously observed in the Arctic, i.e. significantly decreasing perfluorooctane sulfonate (PFOS) according to its global ban and increasing long-chain perfluorinated carboxylic acids (PFCAs). Correlations with selected climate parameters showed an association between PBDE-47 and sampling year precipitations. To our knowledge, this work represents the longest time trend study of pollutants in penguins from the Ross Sea and the first one reporting PFAS. It highlights the importance of global regulations for the contaminant developments in polar ecosystems.

1. Introduction

Chemical contamination has been studied in Antarctica since the 1960s (Risebrough et al., 1968) and it is recognised as one of the most important threats for Antarctic wildlife and biodiversity (Bengtson Nash et al., 2023; Brooks and Ainley, 2022; Chown et al., 2015; Convey and Peck, 2019). Despite these efforts to establish levels of organic pollutants (Lewis et al., 2022; Morales et al., 2022; Wild et al., 2022), a coordinated monitoring program is lacking (Bengtson Nash et al., 2023; Corsolini and Ademollo, 2022) and only few temporal trend studies are available for Antarctic biota, e.g. Cincinelli et al. (2016), Corsolini et al. (2011), Ellis et al. (2018), Goerke et al. (2004), Markham et al. (2018), van den Brink et al. (2011). However, the scientific community widely recognises the importance of monitoring chemical contamination in Antarctica to improve our insights on the global distribution of pollutants (Bargagli, 2008; Bengtson Nash, 2011; Corsolini and Ademollo, 2022; Ebinghaus et al., 2023; Grotti et al., 2016; Kallenborn et al., 2013) and to guide regulatory decisions, providing data on long-range transport and bioaccumulation of toxic substances (Bengtson Nash et al., 2023; Ebinghaus et al., 2023).

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Temporal trend analysis is an important tool to evaluate the effectiveness of the global restriction measures applied to chemicals of concern and has been established in the Arctic for many years (Bengtson Nash, 2011; Cincinelli et al., 2016; Hung et al., 2016b; Rigét et al., 2019; Wong et al., 2021). Time trend monitoring can also empirically support and validate predictive modelling studies on contamination levels (Hansen et al., 2020). However, our ability to detect and interpret pollutant trends could be compromised by climate-driven confounding factors (AMAP, 2021; Hung et al., 2016b). As reviewed in Vorkamp et al. (2022), recent studies have shown correlations between climate change parameters and temporal trends of pollutants in Arctic biota. For example, Bustnes et al. (2010) found a significant association between Arctic Oscillation (AO) index and persistent organic pollutants (POPs) in glaucous gulls (*Larus hyperboreus*), and Foster et al. (2019a) observed a positive correlation between precipitation and POP concentrations in thick-billed murre (*Uria lomvia*). However, our understanding of the influence of climate change on the pollutant trends is still scarce (AMAP, 2021), and very few studies investigated climate-connected effects on pollutant distribution in Antarctica (Corsolini and Ademollo, 2022). Among them, Geisz et al. (2008) suggested that melting glaciers may have an influence on DDT concentrations in penguin eggs, and Cincinelli et al. (2016) hypothesised iceberg calving to cause concentration peaks in fish. To the best of our knowledge, a correlation between climatic parameters and pollutant concentration has not previously been tested in Antarctica. Changes in climate variables can affect pollutant concentrations in biota in several and unexpected ways. For example, shifts in diet due to alterations in sea ice coverage have been observed in penguins (Corsolini et al., 2003), with potential different exposure to contaminants. Additionally, rising temperatures may increase volatilization from water and reduce the bioavailability of pollutants in marine food webs (Vorkamp et al., 2022). Thus, correlation between pollutants and climate parameters deserve to be studied and, since the Ross Sea Marine Protected Area (RS-MPA) was established in 2017 (CCAMLR, 2016), monitoring studies will be even more useful to understand whether and how the establishment of regional protection measures may affect the contamination burden in Antarctic biota.

Adèlie penguin (*Pygoscelis adeliae* Hombron & Jacquinot, 1841) represents a good sentinel species for monitoring pollutants on a regional scale, as shown for example in Brasso et al. (2012), Dos Santos et al. (2006), Lewis et al. (2022). Moreover, abandoned eggs are considered a valuable tool for non-destructive monitoring purposes (e.g. Cipro et al., 2010; Corsolini et al., 2002; Mello et al., 2016; Morales et al., 2022; Schiavone et al., 2009a, b) and the use of archived samples allows retrospective analysis for detecting temporal changes (Bianchini et al., 2022; Grotti et al., 2016; Vorkamp et al., 2018). Following the aims of the Antarctic Monitoring and Assessment Program (AnMAP, https://scar.org/scar-news/anmap-scar) of the Scientific Committee on Antarctic Research (SCAR), endorsed also by the United Nations Ocean Decade Action (https://www.oceandecade.org), this study attempted to fill the above-mentioned knowledge gaps and to compare results with those reported for the Arctic by the Arctic Monitoring and Assessment Programme (AMAP) (Reiersen et al., 2024).

The main goal was to evaluate the temporal trends of selected POPs, and some currently unregulated perfluoroalkyl substances (PFAS) over more than two decades (1997–2021) in samples of Adèlie penguin eggs collected along the Ross Sea coasts and provide a first assessment of the relationships between chemical contamination and climate parameters.

More specifically, we aimed: i) to evaluate the temporal trend of pollutants such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), organochlorine pesticides (OCPs) and perfluoroalkyl substances (PFAS) in eggs of Adèlie penguin collected from three colonies along the Ross Sea coasts; therefore we could evaluate the effectiveness of the global restrictions on the use of legacy pollutants (e.g. PCBs and OCPs) and yet provide data on still not regulated substances (e.g. long-chain perfluorinated carboxylic acids, PFCAs) that will be useful for future regulatory purpose; ii) to assess the correlation between pollutants and some climate parameters (such as temperature, precipitation, sea ice coverage and oscillation indices) providing suggestions on how climate may indirectly affect the contamination in Antarctic biota.

We expect to observe temporal trends similar to those observed years or decades ago in the northern hemisphere, and specifically in the Arctic, due to the overall delayed transport of pollutants to Antarctica from primary source regions.

2. Materials and methods

2.1. Sampling, species and study area

A total of 50 Adèlie penguin unhatched eggs (Table S1) were collected at the end of the hatching period (December and beginning of January) during seven Antarctic expeditions (1997/1998, 2000/2001, 2002/2003, 2004/2005, 2010/2011, 2018/2019, 2021/2022) in the framework of the Italian National Research Program in Antarctica (PNRA). The samples were collected from three colonies laying within or nearby Antarctic Specially Protected Areas (ASPAs), specifically at: Inexpressible Island (74°54 S, 163°39 E), Adèlie Cove (74°46 S, 164°00 E) and Edmonson Point (74°20 S, 165°08 E) (Fig. S1). Eggs were stored in polyethylene bags and kept at −20 °C until laboratory analysis.

The three colonies belong to the same metapopulation (Ballerini et al., 2015) and, as reported in Pala et al. (2024), eggs collected in those colonies did not show significant differences in the contamination levels. Thus, they were combined to study the contamination trends.

Adèlie penguins are capital breeders as resources they allocate for the egg formation are stored before the breeding period (Astheimer and Grau, 1985; Meijer and Drent, 1999; Mello et al., 2016); moreover it is known that, together with necessary resources (e.g. lipids and protein), they transfer contaminants to the eggs (Schiavone et al., 2009a, b; Brasso et al., 2015; Mello et al., 2016). Therefore, it is suggested that their eggs reflect the female pollutants intake during the late winter to pre-breeding period (Brasso et al., 2012; Mello et al., 2016; Padilha et al., 2022). Thanks to tracking studies conducted on other Ross Sea colonies (Ballard et al., 2010; Davis et al., 1996), we can estimate the wintering area and the pre-breeding tracks of the individuals of the study, assuming that they behave in the same way as southern Ross Sea colonies, i.e. moving northward to pursue better sea-ice and sunlight conditions to survive (Ballard et al., 2010). Consequently, we assume that the eggs reflect the contamination of those areas during the late winter to pre-breeding period.

Accordingly, for climate and weather parameters we chose a reference area (Fig. 1), and the period mid-July (late winter) to mid-October, when the penguins start to arrive at their breeding colonies (Olmastroni et al., 2022) and begin the pre-laying fast. The selected period should conservatively cover the resources storing period, including the time in which the penguins increase their foraging effort (September–October; Takahashi et al., 2018; Thiebot et al., 2019), and the beginning of the egg formation (Astheimer and Grau, 1985; Meijer and Drent, 1999).

2.2. Chemicals and residue analysis

2.2.1. Polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs)

The samples were analysed for seven PCB congeners (IUPAC numbers 28, 52, 101, 118, 138, 153, 180), hexachlorobenzene (HCB), *p,p'*-dichlorodiphenyldichloroethylene (DDE) and twelve PBDE congeners (IUPAC numbers 17, 28, 47, 49, 66, 85, 99, 100, 153, 154, 183, 209).

The analytical method was previously described in Pala et al. (2024). The whole egg content was homogenised and approximately 2.5 g were spiked with a known amount of the recovery standards PCB-40 and PCB-198. The extractable lipid content was determined gravimetrically.

PCBs and OCPs were identified and quantified on an Agilent 7890A gas chromatography (GC) system coupled to a micro electron capture detector (Agilent, Palo Alto, CA, USA).

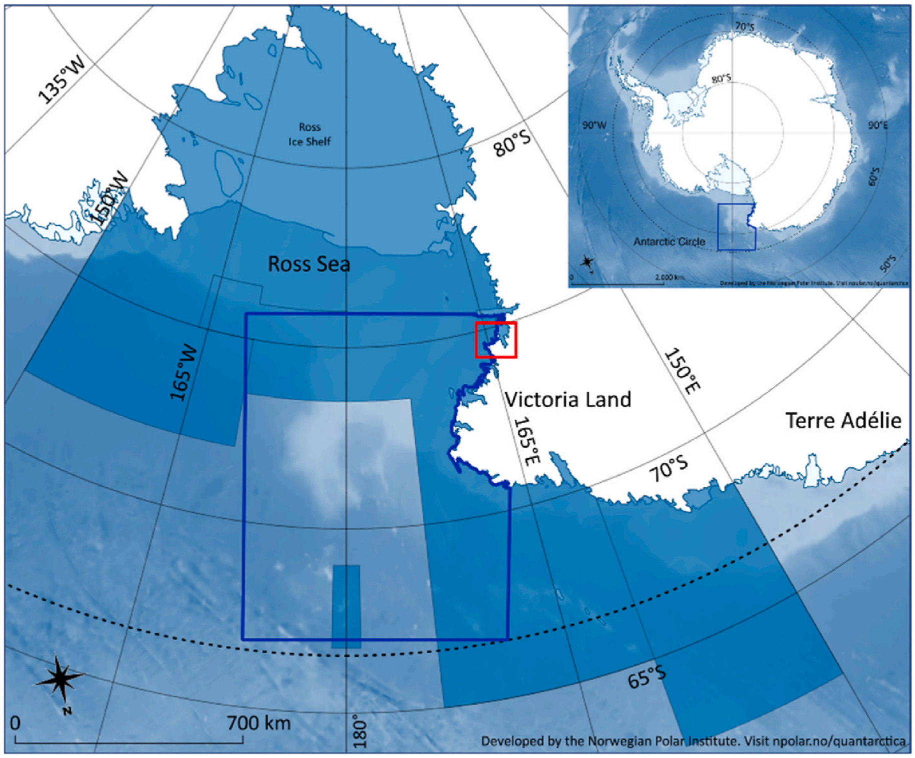

Fig. 1. Map showing the approximate penguin late winter/pre-breeding living area (blue open square) for which climate data were collected. The studied colonies lie within the red squared area (focusing closely on Fig. S1). The light blue shaded area indicates the Ross Sea Marine Protected Area (RS-MPA). The dashed line represents the Antarctic Circle. The map was created using Quantarctica 3.2 (Matsuoka et al., 2021). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

PBDEs were analysed on an Agilent 7890B GC system coupled to a 5977B mass spectrometer (Agilent, Palo Alto, CA, USA). PBDE-209 was analysed on an Agilent 6890 GC, coupled with a 5973N mass spectrometer (Agilent, Palo Alto, Ca, USA). Analyses were performed in selected ion monitoring (SIM) mode with electron capture negative ionization (ECNI).

Quantification was based on the construction of linear duplicate nine-point calibration curves for PCBs, OCPs and PBDE-209 (ten-point for the other PBDEs).

All the target compounds showed a significant statistical correlation (Pearson correlation test, $p < 0.05$) between log-transformed wet weight (ww) concentration and lipid content (Table S2) thus, results were presented on lipid weight (lw) basis. Lipid contents are reported in Table S1.

The quality assurance and control (QA/QC) results of the analyses were given in Pala et al. (2024). Briefly, all recovery rates were between 82 and 110 % and thus deemed acceptable. Maximum deviation between duplicates was 17 %. Blank values were below the method detection limit (MDL) in all the samples. The data are also provided in the Supplementary Information (Tables S1 and S3).

2.2.2. *Ionic perfluoroalkyl substances (PFAS) and perfluorooctane sulfonamide (PFOSA)*

The PFAS analysed in the samples were identical with those in Pala et al. (2024) and included five perfluorinated sulfonic acids (PFSAs), ten PFCAs and perfluorooctane sulfonamide (PFOSA).

PFAS extraction was based on the method described in Pala et al. (2024). Aliquots of around 1 g of the previously homogenised whole egg content were spiked with a mix of ^{13}C-PFAS as internal standards (Table S4). The instrumental analysis was performed on a high-performance liquid chromatography (HPLC) system Agilent 1290 Infinity II, coupled with an Agilent 6495 triple quadrupole mass spectrometer (Agilent, Palo Alto, CA, USA). The analysis was performed in multi reaction monitoring (MRM) mode with negative electrospray ionization (ESI).

Quantification was based on the construction of a five-point calibration curve in duplicate. Monitored ions and their corresponding ^{13}C-labelled standards are reported in Table S4. Results are reported in ng/g ww. The QA/QC measures were described in detail in Pala et al. (2024). Briefly, deviations from the assigned values in reference materials were acceptable for all compounds except perfluorotetradecanoic acid (PFTDA), that was accordingly excluded. Relative percentage difference between duplicates was <10 % for the included compounds, and procedural blanks did not show detectable values. MDLs are given in Table S3.

2.3. *Climate and meteorological parameters*

The selected climate parameters were: total precipitations, 2m-temperature (defined as the air temperature at 2 m above the sea surface), sea-ice coverage and Antarctic Oscillation (AAO) index. Selection of parameters was based on what had been previously concluded in reviews of similar Arctic studies (de Wit et al., 2022; Vorkamp et al., 2022).

Daily values for AAO were available from 1979 to 2024 at https://www.cpc.ncep.noaa.gov/.Precipitation and temperature data (1996–2021) were obtained from the ERA5 product (the fifth generation of ECMWF reanalysis data)and were downloaded as hourly values from the Copernicus Climate Data Store (https://cds-beta.climate.copernicus.eu/), computed as daily aggregated values and averaged for the region of interest. Sea-ice data were estimated from Moderate Resolution Imaging Spectroradiometer - MODIS - product MOD291PD 6.1 version (https://nsidc.org/data/data-access-tool/MOD29P1D/versions/61), which provides daily sea-ice cover at 1 km spatial resolution since 2000. Here, also the sea-ice cover in percentage was calculated as the ratio of sea-ice and total non-cloud cover areas by means of Terra library in R software.

Table 1
Results of the time trend analysis for the selected chemicals detected in Adèlie penguin eggs from the Ross Sea over the period 1997–2021, based on annual geometric means. Annual change (%) with ±95 % confidence interval (CI); coefficient of determination (r^2) and p-value for the linear regression (lr) are reported along with the non-parametric Mann-Kendall tau and its respective p-value. Significant results ($p < 0.05$) are indicated with bold.

Compound	n	Annual change (%)	CI	r^2	P(lr)	tau	P(tau)
PCB-28	29	−1.43	(-2.7, −0.1)	**0.16**	**0.03**	**−0.41**	**0.01**
PCB-118	50	−1.36	(-2.4, −0.3)	**0.12**	**0.01**	−0.17	0.11
PCB-138	50	−1.30	(-2.3, −0.3)	**0.12**	**0.01**	−0.18	0.10
PCB-153	50	−1.24	(-2.4, 0.0)	**0.08**	**0.04**	−0.12	0.27
PCB-180	50	−2.03	(-3.3, −0.8)	**0.18**	**0.00**	**−0.26**	**0.02**
$\sum_4$PCB[a]	50	−1.40	(-2.5, −0.3)	**0.11**	**0.02**	−0.16	0.13
PCB-28/PCB-180	29	−0.11	(-1.7, 1.5)	0.00	0.89	−0.08	0.60
PCB-118/PCB-180	50	0.68	(0.18, 1.2)	**0.13**	**0.00**	**0.31**	**0.00**
p,p'-DDE	50	−0.38	(-1.3, 0.6)	0.01	0.43	−0.01	0.94
HCB	29	−0.86	(-1.9, 0.1)	0.10	0.09	−0.17	0.28
PBDE-47	50	−0.38	(-1.3, 0.6)	0.01	0.43	−0.01	0.95
PFHxS	29	−3.07	(-6.3, 0.3)	0.11	0.07	−0.25	0.11
PFOA	29	−0.17	(-2.2, 2.0)	0.00	0.87	−0.04	0.76
PFOS	29	−2.37	(-3.5, −1.2)	**0.39**	**0.00**	**−0.40**	**0.01**
PFNA	29	2.00	(0.7, 3.4)	**0.26**	**0.01**	**0.38**	**0.02**
PFDA	29	3.96	(2.5, 5.5)	**0.54**	**0.00**	**0.57**	**0.00**
PFUnDA	29	3.52	(2.2, 4.8)	**0.54**	**0.00**	**0.53**	**0.00**
PFDoDA	29	0.98	(-1.1, 3.1)	0.03	0.35	0.08	0.60
PFTrDA	29	2.83	(0.8, 4.8)	**0.24**	**0.01**	**0.42**	**0.01**
$\sum$PFAS	29	0.38	(-0.8, 1.6)	0.01	0.53	0.09	0.56
$\sum$PFCA	29	2.45	(1.0, 3.9)	**0.31**	**0.00**	**0.41**	**0.01**
PFOS/$\sum$PFCA	29	−4.70	(-5.5, −3.9)	**0.83**	**0.00**	**−0.60**	**0.00**

[a] $\sum_4$PCB = sum of four PCBs, PCB-28 excluded.

Sea-ice coverage data refers to September because absent or very limited sunlight does not allow reliable satellite measurements during the previous months.

2.4. Data analysis

Statistical analyses were performed through the software PIA (http://www.amap.no) and GraphPad Prism 5.01 (GraphPad Software). Values below MDL were substituted with MDL/$\sqrt{2}$. Compounds below the MDL in more than 50 % of the samples were excluded. Concentrations were not blank corrected. Correlations with lipid contents were evaluated through Pearson correlation test on log-transformed data after checking the normality assumption with D'Agostino-Pearson test. Temporal trend analysis was performed by log-linear regression using yearly geometric means. In a conservative approach, linear regression results were compared to the non-parametric Mann-Kendall trend test results to avoid conclusions on significant results that may be caused by exceptionally high (or low) endpoints (at the end or beginning of the time period). Thus, cases of significant linear regression and non-significant Mann-Kendall results could reflect the low power of the non-parametric test or the above-mentioned leverage of the endpoints.

Correlations between chemicals and climate parameters were evaluated through Pearson and Mann-Kendall tests using median concentrations for the year of sampling and applying a time-lag of one year. Significance level was established at 5 %.

Some available samples had already been freeze-dried for another study, as discussed in Pala et al. (2024). Based on evaluations of which data obtained from dried and wet sample analyses could be combined (Pala et al., 2024), the number of samples included in the time trend varied for different compounds (Table 1).

3. Results and discussion

The concentrations of the studied compounds in recent samples (2018–2021) were described in detail in Pala et al. (2024). Concentrations over the entire study period (1997–2021) are summarised in Table S5. Individual egg concentrations are reported in Tables S6a and S6b.

PCB congeners 52 and 101, all PBDE congeners except PBDE-47, three PFSAs and three PFCAs were excluded from the time trend analysis due to >50 % of the samples below MDLs. (for PFTDA see section 2.2.2). PFOSA was excluded because it was only detected in samples from the 2021/2022 expedition (Pala et al., 2024).

3.1. Temporal trends

Table 1 reports the results for the time trend analysis. The linear regression results and the non-parametric tests agreed in most of the cases, except for PCB congeners 118, 138, and 153 (and consequently the $\sum_4$PCB). For those compounds the linear regression results showed significant decreasing trends but the lack of significance of the Mann-Kendall test results seemed to suggest that linear regression results may be due to the influence of a single egg collected in 1997, that showed values two to three times higher than the other from the same year (Table S6a). However, the low power of the non-parametric test as a possible explanation cannot be excluded. There were not known analytical issues with that specific egg sample, thus, a casual higher exposure for the laying penguin or other individual factors may have been hypothesised. Considering all that said, as reported in Section 3.1.1 we assume no significant trend for PCB-118, 138 and 153.

3.1.1. PCBs

Among the seven PCB congeners analysed, five were detected with a 100 % detection frequency (IUPAC numbers 28, 118, 138, 153, 180) and all of them indicated a slightly decreasing trend with an annual change ranging from 1 to 2 % (Table 1). This result was expected and likely reflects how PCBs are responding to regional restriction measures applied since the end of the 1970s and the following international ban through the Stockholm Convention (https://chm.pops.int/) in 2004. However, only PCB-28 and PCB-180, which also showed the highest annual change, significantly decreased over time (Fig. 2).

To the best of our knowledge only three studies have evaluated the time trend of PCBs in penguins so far. Among these, Corsolini et al. (2011) and van den Brink et al. (2011) focused on Adèlie penguins. While Corsolini et al. (2011) did not find differences over the period 1995–2005 in eggs from the Ross Sea (considering either, individual PCBs 28, 52, 101, 118 + 149, 153, 138, 180 or the $\sum_{54}$PCBs), van den Brink et al. (2011) reported a significant decreasing trend for PCB-153 in preen oil taken from Hop Island penguins between 1994 and 2004. The third available study reported an increase for PCB isomeric average and

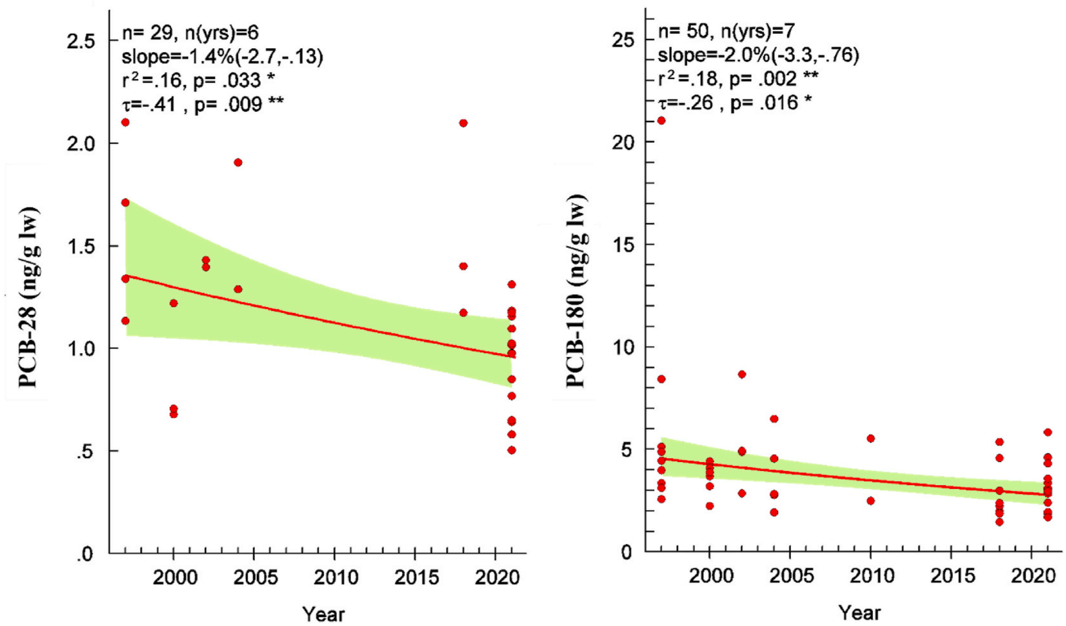

Fig. 2. Time trend of PCB-28 (left) and PCB-180 (right) in Adèlie penguin eggs collected along the Ross Sea coasts over the period 1997–2021. n = number of samples; n(yrs) = number of years. Slopes indicate the annual percent change with 95 % confidence interval in brackets (green area); r^2 = coefficient of determination of the regression line (in red) with *p*-value; τ = Mann-Kendall tau with *p*-value. * Indicates a significant trend. Temporal trends of the other PCBs are reported in Fig. S3a. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

∑PCBs (with no information on individual congeners) between the 1960 and 2010 in a circumpolar study based on literature results (Ellis et al., 2018). However, data originated from seven penguin species pooled together and with a wide geographical range, thus making the comparison with our results difficult. In general, also considering temporal trend studies conducted on other organisms, e.g. fish (Cincinelli et al., 2016; Goerke et al., 2004) or molluscs (Grotti et al., 2016), results were not consistent as PCB bioaccumulation seemed to be highly dependent on species and site (Corsolini, 2009) and was reported to show wide fluctuations over time (Ellis et al., 2018; Grotti et al., 2016), as well as different bioaccumulation patterns between benthic and pelagic species (van den Brink et al., 2011). The paucity of data is also reported to be an important limitation for time trend studies in Antarctica (Corsolini, 2009; Cincinelli et al., 2016; Corsolini et al., 2017; Ellis et al., 2018).

An overall decreasing trend was observed in Baltic Sea biota (1969–2012), including seabirds and fish (Nyberg et al., 2015). Concerning the Arctic, Rigét et al. (2019) summarised the circumpolar results from time trend studies conducted over more than three decades (1980s–2010s) and reported an overall decrease for PCBs. A PCB decrease over four decades (1975–2015) was also reported in seabird eggs collected in the Canadian Arctic (Braune et al., 2019) as well as in ringed seals from the same area and period (Houde et al., 2019). However, some time series indicated stable results for some congeners, for example CB-153 in ringed seals in the Canadian Arctic and East Greenland as well as in common eiders from Svalbard (Bustnes et al., 2024; Houde et al., 2019; Vorkamp et al., 2011). Rigét et al. (2016) reported for polar bears from East Greenland that recent increases of PCBs had been observed. Possible explanations for stable or even increasing concentrations might include climate change-related factors, such as remobilisation from secondary sources and changes in ecosystem structures, or emissions from local sources (Houde et al., 2019; Rigét et al., 2019; Vorkamp et al., 2022).

It is interesting to note that the most reliable decreasing trends were detected for the two congeners with the highest (PCB-180) and the lowest (PCB-28) chlorination degree, despite their different chemical-physical properties. Moreover, the rate of decrease was faster for the more recalcitrant PCB-180 (Table 1), contrary to what may be expected. In fact, the tri-CB-28, being more volatile, can easily reach polar regions (Wania and MacKay, 1996; Wania and Su, 2004) and having entered the trophic web could be more readily metabolised (Bengtson Nash et al., 2008; Wild et al., 2022), thus a faster decrease over time may be hypothesised for PCB-28 in biota. In a warming climate, on the other hand, PCB-28 could be more prone to remobilisation from pollutants reservoir like ice, snow or water (Hung et al., 2022; Vorkamp et al., 2022), thus becoming available again for bioaccumulation in the trophic webs (Potapowicz et al., 2019). Looking at our results, the hypothesis of a remobilisation from secondary sources seemed to be likely. Years after the PCB ban, the trend of the ratio PCB-28/PCB-180 would be expected to decrease (Nyberg et al., 2014) due to the higher persistence of the PCB-180. However, such a ratio was stable over decades (Table 1), suggesting that new emissions of PCB-28 (i.e. remobilisation from secondary sources) may have been occurred.

In addition to these secondary sources, new PCB inputs to Antarctica may originate from PCB-containing e-waste disposal practices in the southern hemisphere, possibly with enhanced long-range atmospheric transport due to global warming (Breivik et al., 2014; Ellis et al., 2018). This could be especially relevant for volatile PCB congeners, such as PCB-28 (Wania and MacKay, 1996). Furthermore, although the PCB pattern apparently showed an overall stable contribution of the detected congeners over time (Fig. S2), the ratio PCB-118/PCB-180 showed a significant increasing trend (Table 1), potentially indicating new sources for the penta-chlorinated PCB-118 and rising toxicological concern about exposure to this dioxin-like congener. Along with the above-mentioned potential sources, a long-range delayed transport from source regions through ocean or atmospheric currents may have also influenced the PCB-118 trend. However, the low detoxifying activity of the penguins enzymatic system for this specific congener was reported (Corsolini et al., 2011; de Boer and Wester, 1991; Wanwimolruk et al., 1999) and may have caused a slower decreasing rate for PCB-118 compared to PCB-180.

3.1.2. OCPs

HCB and *p,p'*-DDE, both with a 100 % detection frequency, indicated decreasing but not significant trends over the studied period (Table 1; Fig. S3b). In the case of *p,p'*-DDE this result could be related to the ongoing use of its precursor *p,p'*-DDT for malaria control (Pozo et al., 2017; Zanardi-Lamardo et al., 2019), combined with its high persistence in the environment. Moreover, remobilisation from melting ice has been hypothesised to be a source of DDTs to the Antarctic marine ecosystem, possibly contributing to the exposure of the penguins along the Antarctic Peninsula (Geisz et al., 2008). For HCB, ongoing sources might be related to its unintentional production as impurities in pesticides formulations or an industrial by-product (AMAP, 2021; Bailey, 2001; Barber et al., 2005). Given its high volatility and persistence, it can reach the Antarctic efficiently (Bailey, 2001; Barber et al., 2005). HCB remobilisation from Antarctic benthic biota, following increasing temperatures, had been hypothesised by Bates et al. (2017) and could have made it available for bioaccumulation in the pelagic trophic web, thus affecting its trend in penguin eggs. However, such a hypothesis was based on an ecosystem model outcomes and potential mechanisms were not explained.

Other Antarctic studies reported not significant (Corsolini et al., 2011, 1995–2005) or decreasing (van den Brink et al., 2011, 1993–2004) trend of HCB and not significant trend for *p,p'*-DDE (Corsolini et al. (2011), 1995–2005; van den Brink et al. (2009), 1994–2004; in the latter case the authors indicated a decreasing trend, however the reported *p*-value was actually above the selected significance level) over time in Adèlie penguins. Ellis et al. (2018), reported for the same compounds a decreasing trend in pooled penguin species since the 1990s, followed by stable concentrations at present. Thus, even considering all the limitations in comparing trend studies (e.g. different periods, areas, and number of samples) those results seemed to agree with our observations.

In the Arctic, *p,p'*-DDE were mostly reported as significantly decreasing in biota (1975–2015, Braune et al., 2019, 1972–2016, Houde et al., 2019; Rigét et al., 2019). However, while *p,p'*-DDE seemed to show the same slowly decreasing trend as PCBs, HCB was occasionally reported as increasing, including in seabirds from Svalbard (2007–2017, Bustnes et al., 2024), or showed slower decreasing rate in biota compared to other POPs (Rigét et al., 2019). Same observations were made for HCB in Arctic air (Hung et al., 2016a). Besides the above-mentioned still ongoing HCB emissions as possible causes, climate change-related effects have been discussed for temporal trends of HCB, since its volatility might make it most susceptible to temperature changes (Rigét et al., 2019; Vorkamp et al., 2022), including volatilization from secondary sources as hypothesised by Ma et al. (2011). Glacial run-off was also suggested to affect the HCB trend in the fjord environment of Svalbard (Bustnes et al., 2024; Pouch et al., 2018).

3.1.3. PBDE-47

PBDE-47 was detected in all samples and indicated a decreasing but not significant trend over the studied period (Table 1; Fig. S3c). PBDE-47 is one of the major congeners in the Penta-BDE technical formulation (La Guardia et al., 2006), which was internationally banned in 2009 (UNEP, 2009a), i.e. later than PCBs, HCB and DDTs (restricted). Thus, this result could indicate a delayed effect of the global phase-out, compared to the chlorinated POPs, that may become significant in the future. Markham et al. (2018) suggested a delayed arrival of PBDEs in the southern hemisphere by approximately a decade after a time-lag due to the long-range transport. Moreover, Chiuchiolo et al. (2004) reported melting sea-ice as a possible source for PBDEs to enter Antarctic trophic webs, thus, as previously hypothesised for other POPs, the warming climate may have influenced the observed trend.

PBDEs in Antarctic biota were less studied than the other classes of POPs (Mello et al., 2016; van den Brink et al., 2011; Wolschke et al., 2015; Yogui and Sericano, 2009) and data on temporal trends are lacking. To the best of our knowledge only two studies have been published and none deals with penguins. Markham et al. (2018) showed stable or increasing trends (2000–2014) for PBDEs in Antarctic biota at different trophic levels (phytoplankton, krill and fur seal milk), collected in the Ross Sea and along the Western Antarctic Peninsula, highlighting how a decreasing trend was still lacking. Cincinelli et al. (2016) observed a decreasing trend (2005–2011) for PBDEs in *Trematomus bernacchii* from the Ross Sea after a peak in 2005. These authors hypothesised melting icebergs in the previous years as a possible explanation for the highest concentrations in 2005 and suggested that stable or slight increasing levels may have occurred without the additional influence of the icebergs. However, the paucity of data was pointed out as a limitation for the interpretation of the trend.

In the Arctic, PBDEs were reported to decrease having peaked in the beginning of the 2000s in Canadian seabirds (Braune et al., 2015), ringed seals from East Greenland (Vorkamp et al., 2011) and overall, in Arctic biota, as analysed in Rigét et al. (2019). Most of the studies agreed on the explanation that these trends are likely to reflect the bans and voluntary phase-outs of Penta- and Octa-BDE in the European Union and the USA, followed by the global ban through the Stockholm Convention (Braune et al., 2015; Rigét et al., 2019; Vorkamp et al., 2011). Other authors did not observe a decline yet, for example in Canadian ringed seals (1981–2015) (Addison et al., 2020).

3.1.4. PFAS

Eight PFAS were detected (Table 1), of which perfluorohexane sulfonate (PFHxS) showed a 90 % detection frequency, while the others were found in all the samples. PFHxS, perfluorooctanoic acid (PFOA) and perfluorododecanoic acid (PFDoDA) indicated a not significant trend (Table 1; Fig. S3d). Perfluorooctane sulfonate (PFOS) was decreasing while all the remaining long-chain ($\geq C_9$) carboxylated compounds showed increasing trends (Table 1; Fig. 3, S3d and e). The significant results were also confirmed by the Mann-Kendall test, enhancing the reliability of the observations. These results may be expected as they reflect the PFAS restrictions timeline. PFOS was voluntarily phased-out from the beginning of the 2000s and globally restricted in 2009 (UNEP, 2009b), while PFOA and PFHxS, which only indicated a slight and non-significant decreasing trend, were banned in 2019 (UNEP, 2019) and 2022 (UNEP, 2022), respectively. Moreover, the long-chain carboxylates are currently under review by the Stockholm Convention, but not yet regulated (UNEP, 2021), which agrees with their increasing trend. ∑PFAS showed no significant trend (Table 1), likely because of the opposite trends of PFOS, the compound with the highest percentage contribution (from 33 to 48 %; Fig. S2b) between 1997 and 2004, and PFUnDA (Fig. 3). In fact, PFUnDA, being the second prevailing compound (except in 2000; Fig. S2b) and exceeding PFOS contribution since 2018 (accounting for up to 30 % of ∑PFAS; Fig. S2b), strongly leveraged the overall trend. ∑PFCA significantly increased (Table 1; Fig. 3), with PFOA as the only decreasing compound. Accordingly, the ratio PFOS:∑PFCA significantly decreased (Table 1), well describing the relative changes over time and likely reflecting the shift to fluorotelomer-based industrial processes after the phase-out of PFOS and POSF, as previously suggested (Ahrens et al., 2011; Haque et al., 2023).

Among the few studies reporting PFAS in Antarctic biota (Alfaro Garcia et al., 2022; Padilha et al., 2022; Roscales et al., 2019; Schiavone et al., 2009b; Tao et al., 2006; Wild et al., 2022) none deals with their temporal trends. In Pala et al. (2024) recent PFAS levels in the Ross Sea were discussed: it was highlighted that the only study analysing PFOS in samples from the same area (Tao et al., 2006) did not detect it and reported detection limits ten times higher than ours, making it difficult to assess the temporal trend.

PFAS trends from Arctic biota were inconsistent (Butt et al., 2010; Muir et al., 2019), and dependent on site (e.g. Haque et al., 2023; Smythe et al., 2018), species (e.g. Routti et al., 2017) and compound. They included PFOS peaks, i.e. increases followed by decreases (Rigét et al., 2019). Focusing on bird eggs there seems to be a frequently found pattern of decreasing or stable PFOS concentrations over time together with increasing long-chain PFCAs, thus in line with our observations.

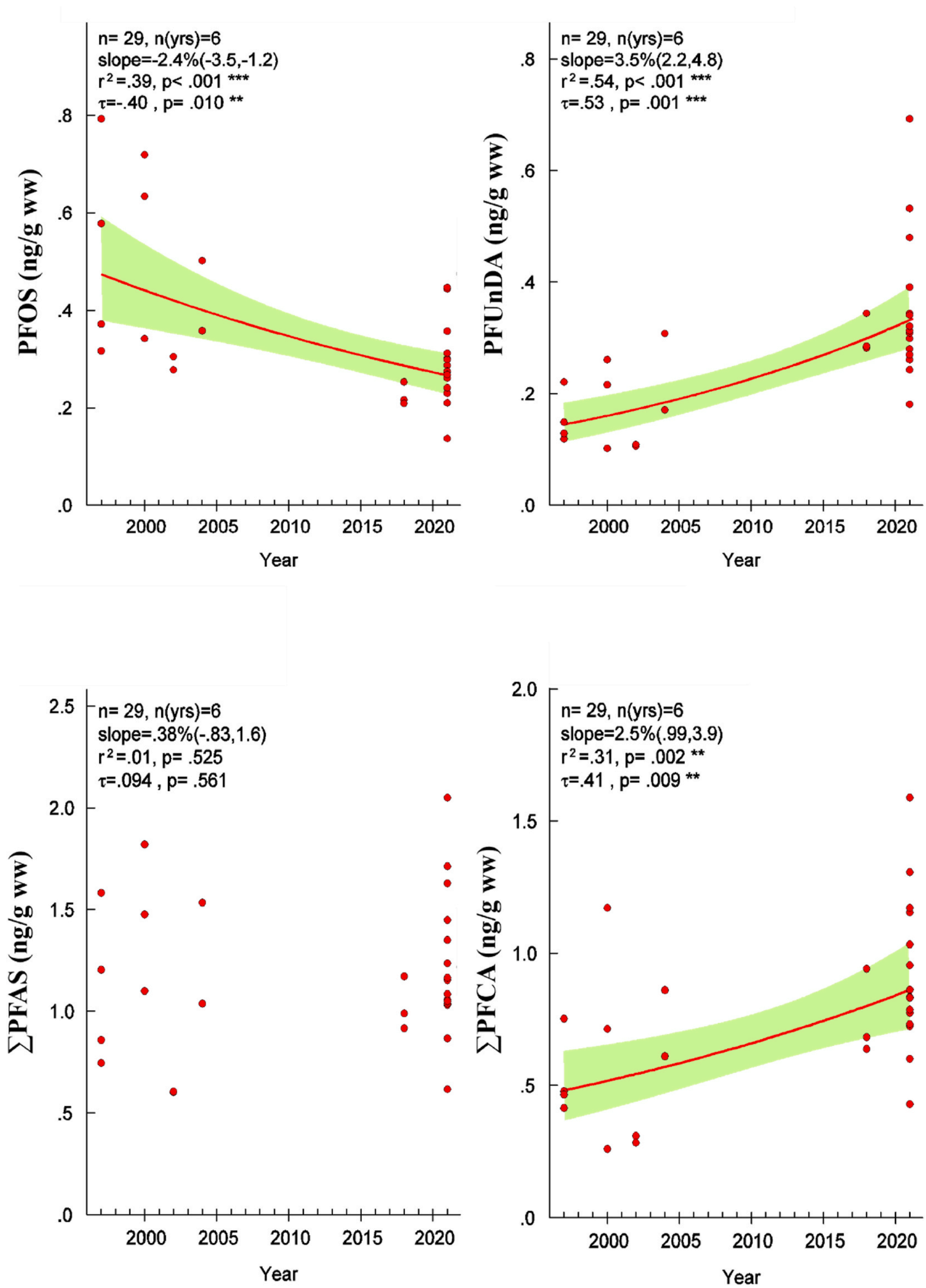

Fig. 3. Time trend of PFOS (top left), PFUnDA (top right), ∑PFAS (bottom left) and ∑PFCA (bottom right) in Adèlie penguin eggs collected along the Ross Sea coasts over the period 1997–2021. n = number of samples; n(yrs) = number of years. Slopes indicate the annual percent change with 95 % confidence interval in brackets (green area, when reported); r^2 = coefficient of determination of the regression line (in red, when reported) with *p*-value; τ = Mann-Kendall tau with *p*-value. * Indicates a significant trend. Temporal trends of the other PFAS are reported in Figs. S3d and S3e. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Such trends were reported in Canadian Arctic seabird eggs collected from 1975 to 2011 (Braune and Letcher, 2013) and in peregrine falcon eggs from Greenland (1986–2014) (Vorkamp et al., 2019). Similar results were shown in studies from the Scandinavian region, for example in Tawny Owl eggs from Norway (1986–2009) (Ahrens et al., 2011) and in peregrine falcon eggs from Sweden (1974–2007) (Holmström et al., 2010), as well as from the Great Lakes herring gull eggs (1990–2010) (Gebbink et al., 2011). Most of the discussions on PFAS trends were based on restriction timelines, changes in production processes and use (e.g. Ahrens et al., 2011; Braune and Letcher, 2013; Vorkamp et al., 2019). When stable PFOS trends were reported, continued emissions from Asian countries were suggested as possibly influencing trends in

Arctic birds (Braune and Letcher, 2013; Vorkamp et al., 2019). However, other factors, such as feeding habits and point sources (Ahrens et al., 2011; Smythe et al., 2018; Vorkamp et al., 2019) as well as differences in exposure pathways (Ahrens et al., 2011; Braune and Letcher, 2013) were also suggested to influence PFAS trends.

Haque et al. (2023) analysed white-tailed sea eagle eggs collected from 1969 to 2021 across Sweden and showed that, although PFCAs were overall increasing over the whole study period, some of them (C_{10}-C_{14}) started to significantly decrease in the last 15 years. The authors suggested that previous studies may have been conducted on too short time spans to observe such trends and related recent decreases to the U.S. phase-out of long-chain PFCAs, which started in 2006 and may have been adapted by other PFCA producing countries (Haque et al., 2023; Land et al., 2018). Similar observations were previously made by Ahrens et al. (2011) and by Muir et al. (2019).

In summary, our results are in line with those observed in the past (until around the mid-2010s) at Arctic and other northern hemisphere sites, while they were different from those found more recently, likely as a consequence of the delayed arrival of pollutants to Antarctica. Future monitoring studies will be useful to check if the trend recently observed by Haque et al. (2023) will be detectable in the Antarctic ecosystem.

3.2. Correlations with climate parameters

The results for the correlations between total precipitation, temperature, sea-ice coverage, AAO index and pollutants levels are reported in Tables S7–S11. Due to the low number of samples and to the limited number of studied seasons, the results should be used with caution, Conservatively, only correlations that were statistically significant and confirmed by the applied non-parametric test were discussed.

PBDE-47 showed a significant negative correlation with the year of sampling total precipitations and the result was confirmed by the Mann-Kendall test (Table S7). To the best of our knowledge the relationship between PBDE-47 and precipitation in seabirds was only reported in AMAP (2021) and was not significant. Our result was opposite to what may be expected since climate-related increasing precipitation was reported to increase POP deposition through scavenging (AMAP, 2021; Borgå et al., 2022; Cabrerizo et al., 2018), resulting in a potential increased bioaccumulation in aquatic trophic webs. In line with this, Foster et al. (2019a) found a positive relationship between HCB, DDE and most PCBs in eggs of thick-billed murre from the Canadian Arctic. The same authors found a negative correlation between PCB-170 and PCB-180 and precipitation, thus similarly to our result. However, the inverse relationship we found cannot be explained. Many factors may affect the observed relationship and comparison among studies, including those concerning similar compounds, is difficult because of the different species and time lags applied. Some authors pointed out the importance of long time series to allow understanding of the correlations (Foster et al., 2019a, b; Houde et al., 2019, 2020; Morris et al., 2022) and some also highlighted how differently long time series may lead to observe inverse relationships (Foster et al., 2019b; Morris et al., 2022).

Despite the limitations, our results, following what has been observed in the Arctic, may indicate climate variables as factors to account for when interpreting results on pollutants distribution and trend in biota and, considering the forecasted and expected global changes (e.g. precipitation are expected to increase in Antarctica (Chown et al., 2022; Lim et al., 2016) as also reported in Wille et al. (2024b, 2024a) after recent extreme heat waves in East Antarctica, including the coast of the Victoria Land), may support the need to include such data in time trend studies, with a particular attention for large-scale climate parameters such as oscillation indices, as previously suggested (De Wit et al., 2022; Vorkamp et al., 2022).

4. Conclusions

This study evaluated the temporal trends of PCBs, OCPs, PBDEs and PFAS in Adèlie penguin eggs collected along the Ross Sea coasts over a 25-year period, thus being the most long-term investigation on both legacy and newer pollutants in Antarctica. The regulation timeline of the included pollutants seems to be a governing factor for the observed trend, for example in the case of declining trends of PCBs and PFOS or increasing trends of long-chain PFCAs. These results highlight the importance of global regulations which are crucial for the protection of the polar environments. Mechanisms related to climate change may further influence the time trends, in particular those of the volatile compounds such as PCB-28. Overall, unhatched eggs of the endemic Adèlie penguin were confirmed to be a reliable tool to evaluate contamination trends of chemical pollutants within the RS-MPA and check the effectiveness of the applied restriction measures over time.

Moreover, in a first attempt to evaluate correlations between pollutants and climate parameters in Antarctica, some significant negative relationships were found with precipitation. Thus, although the paucity of data on pollutants trends and climatic parameters did not allow a clear analysis of the occurring relationships, our results represent a starting point in combining such data and may be useful in the future as time series grow and changes in climate-related parameters might become more pronounced.

CRediT authorship contribution statement

Nicolas Pala: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Katrin Vorkamp:** Writing – review & editing, Supervision, Methodology, Investigation. **Rossana Bossi:** Writing – review & editing, Supervision, Resources, Methodology, Formal analysis. **Anders Bignert:** Writing – review & editing, Supervision, Methodology, Formal analysis. **Giacomo Traversa:** Writing – review & editing, Methodology, Formal analysis. **Davide Fugazza:** Writing – review & editing, Methodology, Formal analysis. **Stefania Ancora:** Writing – review & editing, Supervision. **Nicoletta Ademollo:** Writing – review & editing, Supervision, Conceptualization. **Davide Baroni:** Writing – review & editing, Supervision. **Simonetta Corsolini:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2025.126130.

Data availability

Data will be made available on request.

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