



A ten-year surveillance of PCDD/Fs and PCBs in raw milk from bovine, ovine, and caprine farms in a peri-industrial area of southern Italy

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ABSTRACT

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) remain among the most persistent threats to food safety in peri-industrial agropastoral systems, yet long-term, multi-species surveillance data capable of disentangling environmental, biological, and regulatory determinants of contamination remain relatively scarce in the European literature. This study presents results from a ten-year monitoring program (2016–2025) of PCDD/Fs and PCBs in raw milk from bovine, ovine, and caprine farms reared within the industrially impacted territory of Taranto, southern Italy. A total of 671 samples were analysed by high-resolution gas chromatography–mass spectrometry, and contamination dynamics were modeled using linear mixed-effects regression accounting for farm distance from the industrial perimeter, animal species, and sampling year. PCBs exhibited a significant inverse relationship with distance, whereas pure PCDD/Fs showed no measurable spatial gradient, a pattern consistent with their greater soil persistence and likely reflecting decades-long environmental homogenization. Caprine and ovine milk consistently and substantially exceeded bovine concentrations across all contaminant classes, confirming small ruminants as the most sensitive sentinel species. A significant increasing temporal trend was attributable to sustained soil contamination and to the progressive, deliberate concentration of monitoring on sentinel high-risk holdings. These findings provide a transferable evidentiary basis for species-differentiated, soil-informed surveillance design in European territories affected by legacy industrial contamination.

1. Introduction

Long-term, multi-species monitoring of PCDD/Fs and PCBs in raw milk from extensively managed ruminants remains the exception rather than the rule across European agropastoral systems, and it is precisely this scarcity of longitudinal evidence that limits the capacity of regulatory frameworks to assess their own effectiveness. Yet the stakes are high. More than 80% of human PCDD/F and PCB exposure is dietary, with animal-derived foods, meat, dairy, and eggs, constituting the dominant intake vector (EFSA CONTAM Panel, 2018; Esposito et al., 2017; Güzel et al., 2020; Canlı et al., 2025). Commission Regulation

(EU) 2023/915 (European Commission, 2023) establishes binding maximum levels for these compounds across animal-derived matrices, including raw milk from bovine, ovine, and caprine species, and the scientific basis for these thresholds is under active revision: in 2018, the EFSA CONTAM Panel reduced the tolerable weekly intake (TWI) to 2 pg WHO2005-TEQ/kg body weight (Van den Berg et al., 2006), a sevenfold reduction from the 2001 standard on the basis of evidence linking chronic low-level exposure to reproductive toxicity (EFSA CONTAM Panel, 2018). In this context, a large-scale, multi-species, decade-long dataset from a high-risk contamination zone provides the type of longitudinal evidence required to assess the effectiveness of food safety

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governance, which remains relatively scarce in the European literature.

Extensively managed ruminants are among the most toxicologically exposed food-producing animals in peri-industrial landscapes (Weber et al., 2018), yet they remain among the least systematically monitored. Grazing sheep, goats, and cattle ingest contaminated soil at rates estimated between 2% and 20% of daily dry matter intake, a route of exposure invisible to feed safety controls designed around industrial ingredients (Scortichini et al., 2015). Absorbed lipophilic POPs bioaccumulate in adipose tissue and are excreted via milk fat, making raw milk a direct and measurable reflection of the contamination status of the land on which animals graze (Driesen et al., 2022). Controlled studies have demonstrated that soil-to-milk transfer alone is sufficient to generate exceedances of EU maximum levels at soil concentrations previously considered safe, an effect particularly marked in small ruminants due to their intensive grazing behavior (Weber et al., 2018). The regulatory implication is stark: soil quality standards and food safety thresholds for livestock products were not designed in concert, and this misalignment creates a systemic blind spot that is most dangerous precisely where industrial deposition has been degrading pasture quality for decades.

The province of Taranto, on the Northern Ionian coast of southern Italy, is the most extensively documented instance of this problem in Europe and, for that reason, its most instructive. Home to the continent's largest integrated steelworks and iron ore sintering complex since the 1960s alongside a refinery, thermoelectric plants, and cement works, the province has been subject to chronic PCDD/F and PCB atmospheric deposition across its agricultural hinterland for over half a century (Giannico et al., 2022; Leogrande et al., 2019). Repeated independent monitoring campaigns have confirmed contamination of agricultural soils within at least 15 km of the industrial perimeter (Liberatori et al., 2021), and when exceedances of EU regulatory limits were first detected in ovine tissues and caprine milk in 2008–2009 (Scortichini et al., 2015), the response of the competent authorities was legally extraordinary: a Regional Ordinance (n. 176, 2010) mandated the systematic destruction of small ruminant livers from farms within a 20-km radius and prohibited grazing on non-agricultural land (Regione Puglia, 2010). These measures, for which no direct precedent exists in the harmonised EU framework, indicate that the existing harmonised regulatory architecture was not designed to address a risk of this nature, which had been accumulating for decades. Taranto is not an anomaly. Comparable contamination dynamics and equivalent regulatory inadequacies have been documented in peri-industrial agricultural zones across Europe (Covaci et al., 2007; Turrio-Baldassarri et al., 2009; Weber et al., 2018; Canlı et al., 2022; Güzel et al., 2024). What distinguishes Taranto is not the problem, but the depth and continuity of the monitoring record that has been built around it.

It is precisely this record that the present study exploits. Here we report a ten-year systematic monitoring programme (2016–2025) of PCDD/Fs and PCBs in raw milk from sheep, goats, and cattle reared within 20 km of a major industrial complex in southern Italy, generating one of the largest longitudinal datasets of its kind assembled for any European peri-industrial agropastoral system. The scale and duration of the dataset allow us to pursue objectives that shorter or single-species studies cannot: (i) to characterise spatial and temporal contamination dynamics across three ruminant species simultaneously; (ii) to evaluate regulatory compliance against Commission Regulation (EU) 2023/915 (European Commission, 2023) across a decade of mandatory risk management; and (iii) to provide the first evidence-based appraisal of whether the extraordinary measures mandated by the 2010 Regional Ordinance produced a measurable effect on contamination levels in the food chain. The answers are discussed within the framework of EU food safety law. The Taranto experience, examined at this resolution, offers more than a local case study: it provides a methodological and evidentiary template for the design of long-term food safety surveillance in any European territory where extensive livestock farming intersects with the legacy of industrial contamination.

2. Materials and methods

2.1. Study area

The study was conducted in the province of Taranto, located on the Northern Ionian coast of the Apulia Region, southern Italy. The area is classified as a Site of National Interest (SIN) for environmental remediation and has been recognised as an Area of High Environmental Risk since 1991 (Leogrande et al., 2019), owing to the presence of one of the most extensive industrial complexes in Europe. The industrial pole encompasses an integrated steelwork and iron ore sintering facility, the largest of its kind on the continent, alongside a crude oil refinery, thermoelectric power plants, a cement manufacturing facility, and a major military-naval installation (Giannico et al., 2022; Leogrande et al., 2019). This complex has historically constituted the dominant source of PCDD/F and PCB atmospheric emissions in Italy, and the consequent deposition of halogenated compounds onto the surrounding agricultural and pastoral land has been extensively documented by independent monitoring campaigns (Leogrande et al., 2019; Liberatori et al., 2021).

The study area was defined as the agropastoral territory falling within a 20-km radius of the industrial perimeter, consistent with the risk zone formally established by Regional Ordinance n. 176 of 23 February 2010 (Regione Puglia, 2010). This perimeter encompasses several municipalities within which extensive livestock farming, principally free-range ovine, caprine, and bovine husbandry, has been practised continuously throughout the study period. All farms included in the monitoring program are registered in the Italian National Livestock Registry (Banca Dati Nazionale - BDN) and are characterized by extensive management systems with daytime grazing and overnight housing, a profile that maximizes soil and vegetation ingestion and, consequently, exposure to environmentally deposited PCDD/Fs and PCBs. The geographical extent of the study area and the spatial distribution of monitored holdings are illustrated in Fig. 1.

2.2. Sampling

Raw milk samples were collected within the framework of the Extraordinary Monitoring Plan for Dioxins and PCBs, applied since 2008 by the Prevention Department of the Local Health Authority of Taranto (ASL Taranto) in coordination with the Istituto Zooprofilattico Sperimentale dell'Abruzzo e del Molise (IZS Teramo), the designated Italian National Reference Laboratory for dioxins and PCBs in food and feed. Sampling activities were conducted in compliance with Regulation (EC) No 854/2004 (European Parliament and Council, 2004) for the period preceding 14 December 2019 and with Regulation (EU) 2017/625 (European Parliament and Council, 2017) thereafter, which repealed and replaced the former with effect from that date. The present study reports data collected during the ten-year period from 2016 to 2025.

Over the duration of the monitoring program, a total of 327 farms engaged in raw milk production were registered within the 20-km risk zone: 121 bovine, 105 caprine, and 101 ovine holdings. Annual sampling was organized on the basis of a risk-stratified plan, with a target of approximately 270 farms per year during the earlier implementation phases, distributed equally across the three species (90 bovine, 90 caprine, and 90 ovine) and progressively adjusted in subsequent years in accordance with the observed contamination trend. In the final triennium of the study period (2023–2025), the annual sampling numerosity was reduced to approximately 140 samples across all matrices, reflecting the progressive concentration of monitoring resources on a subset of holdings with historically elevated contamination profiles, which progressively assumed the operational role of sentinel farms, systematically included in each annual sampling cycle to ensure continuity of surveillance on the highest-risk stratum of the monitored population. Farm selection was based on distance from the industrial perimeter, risk categorization, and the results of preceding sampling cycles. Sampling activities were managed through the SINVA national

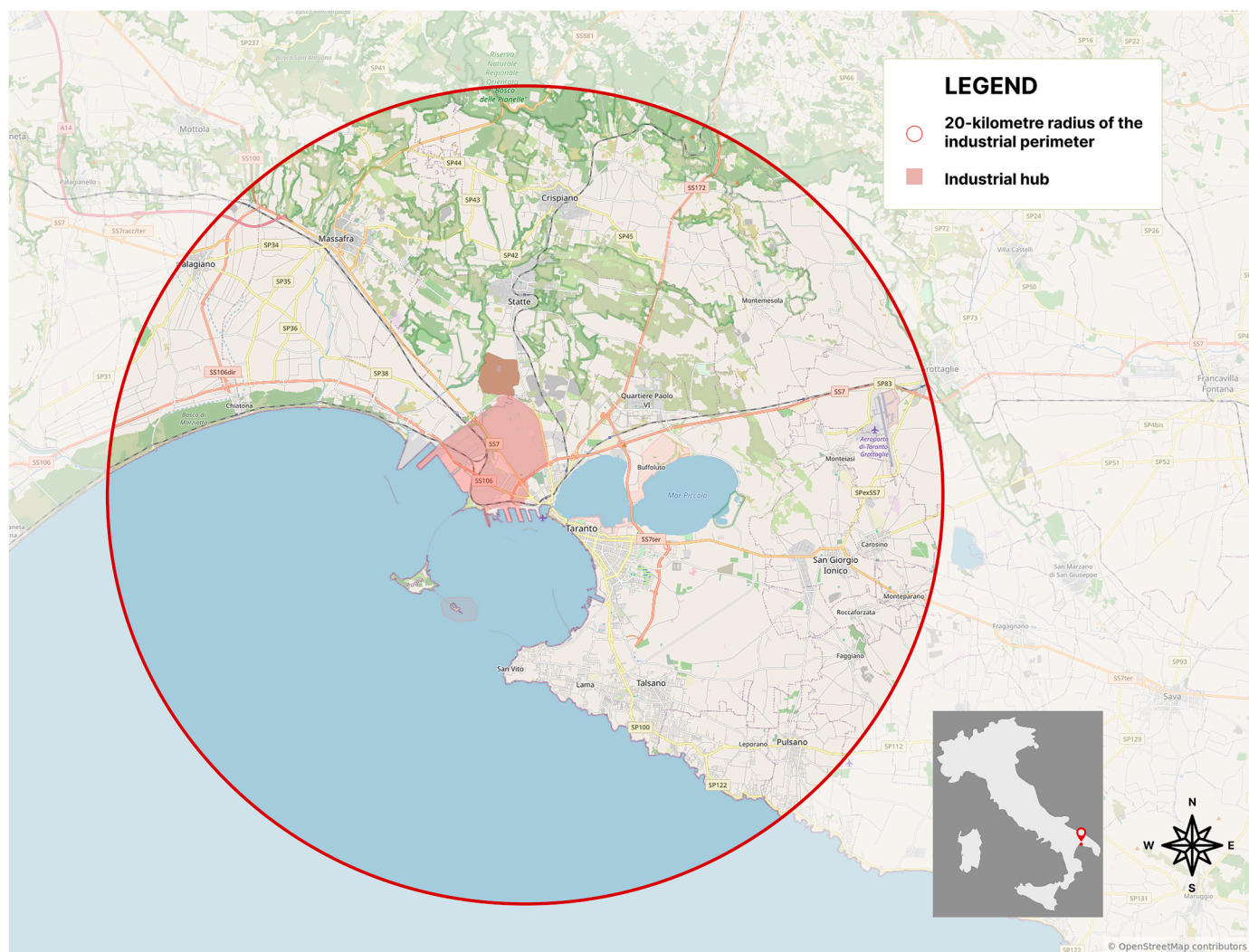


Fig. 1. Geographical extent of the study area. The map shows the agropastoral territory subject to the Extraordinary Monitoring Plan for dioxins and polychlorinated biphenyls within the province of Taranto (Apulia Region, southern Italy). The red-shaded polygon indicates the location of the industrial complex. The red circle delineates the 20 km risk perimeter formally established by Regional Ordinance n. 176 of 23 February 2010, within which all sampled holdings are located.

information system, made available by the *Ministero della Salute* through the national service center at IZS Abruzzo e Molise, which enabled harmonised recording of sampling activities and analytical outcomes.

Bulk raw milk samples of approximately 1 L were collected at farm level by veterinary officers of the ASL Taranto Prevention Department. Samples were collected as single monitoring aliquots and forwarded to the *Istituto Zooprofilattico Sperimentale della Puglia e Basilicata* to IZS Teramo for analysis. All samples were transported under cold chain conditions. Where a non-conformity was identified on a monitoring sample, a confirmatory official sample was collected; upon confirmation, official sampling of meat matrices (liver, muscle, and adipose tissue) was activated following the slaughter of a representative number of animals from the affected herd or flock, in accordance with the operational protocol of the Extraordinary Plan. The full operational protocol of the Extraordinary Monitoring Plan, from routine screening through to enforcement and activation of the Meat Protocol, is summarised in Fig. 2.

2.3. Chemical analysis and analytical methodology

All analytical determinations were performed at IZS Teramo, the Italian National Reference Laboratory for dioxins and PCBs in food and feed. The laboratory is accredited pursuant to ISO/IEC 17025 (ISO,

2017) and operates in full conformity with Commission Regulation (EU) No. 252/2012 (European Commission, 2012), which lays down the methods of sampling and analysis for the official control of PCDD/Fs and DL-PCBs in food.

For PCDD/F determination, all 17 2,3,7,8-chlorine-substituted congeners were analysed. For dioxin-like PCBs (DL-PCBs), the four non-ortho congeners (PCB-77, -81, -126, and -169) and the eight mono-ortho congeners (PCB-105, -114, -118, -123, -156, -157, -167, and -189) were determined. For non-dioxin-like PCBs (NDL-PCBs), the six indicator congeners (PCB-28, -52, -101, -138, -153, and -180) were quantified in accordance with Commission Regulation (EU) 2023/915 (European Commission, 2023). Confirmatory determinations were performed by high-resolution gas chromatography coupled with high-resolution mass spectrometry (GC-HRMS), the reference method mandated by Regulation (EU) No. 252/2012 (European Commission, 2012) for official control purposes.

Analytical performance complied with the requirements laid down in Commission Regulation (EU) No. 252/2012 for confirmatory methods. Quantification was carried out by isotope-dilution GC-HRMS, using $^{13}\text{C}_{12}$ -labelled analogues of the target congeners as internal standards added to each sample prior to extraction. Recoveries of the internal standards fell within the 60–120% range required by the Regulation; the limits of quantification were established well below the maximum and

OPERATIONAL PROTOCOL OF THE EXTRAORDINARY MONITORING PLAN

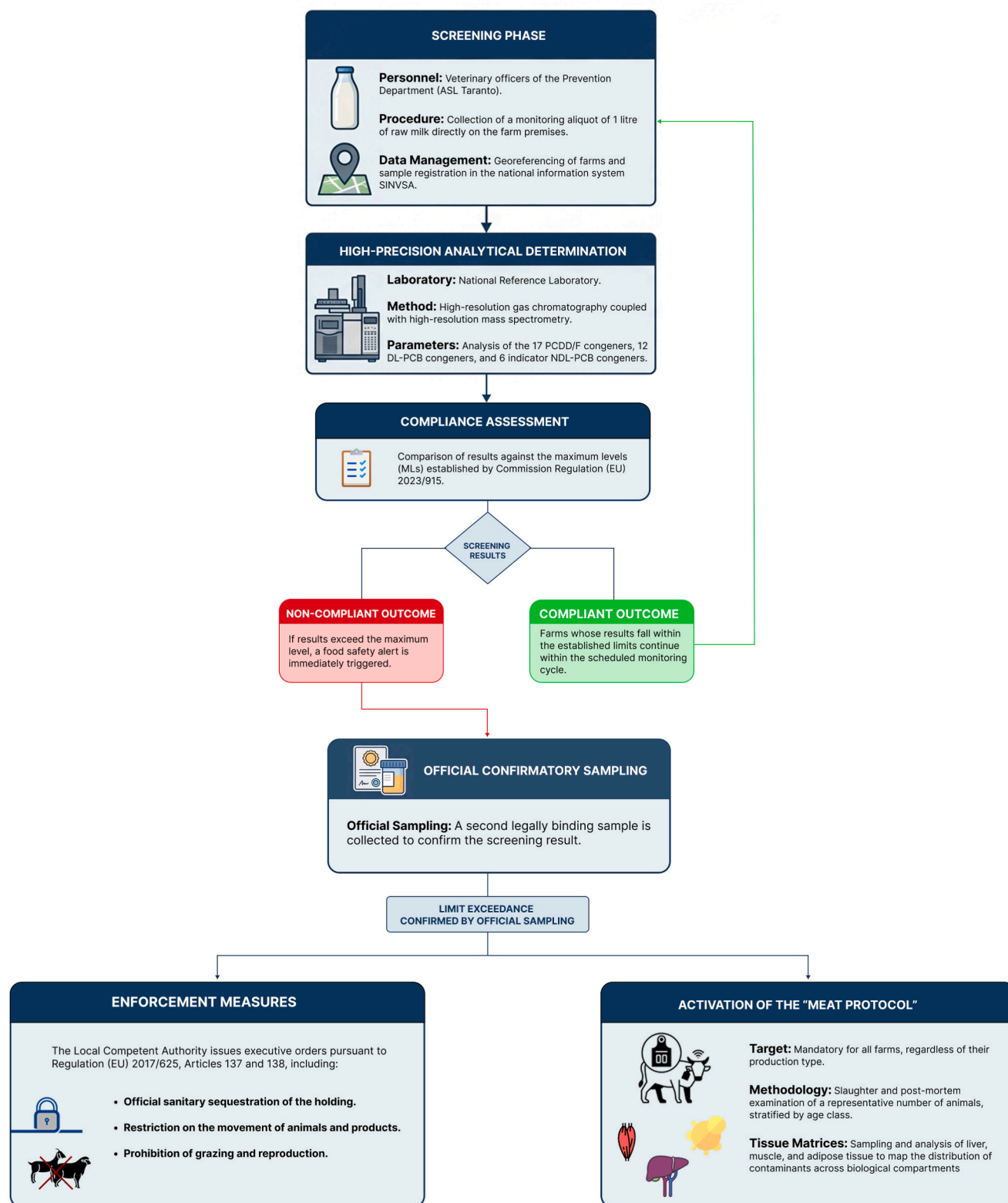


Fig. 2. Operational protocol of the Extraordinary Monitoring Plan for dioxins and PCBs implemented by the Local Health Authority of Taranto (ASL Taranto). The flowchart illustrates the sequential phases of the surveillance system: routine screening by collection of a raw milk monitoring aliquot and high-precision analytical determination by GC-HRMS at the National Reference Laboratory (IZS Teramo); compliance assessment against the maximum levels established by Commission Regulation (EU) 2023/915; and, upon confirmed non-compliance, activation of enforcement measures pursuant to Regulation (EU) 2017/625 (Articles 137 and 138). The "Meat Protocol" is activated conditionally upon legal confirmation of limit exceedance in milk.

action levels applicable to raw milk, allowing reliable quantification at the concentrations of regulatory interest; and the expanded measurement uncertainty of each determination was estimated and applied on an upper-bound basis. Calibration linearity, method precision, and procedural blanks were verified within the laboratory's ISO/IEC 17025 quality-assurance system, and analytical reliability was further supported by the continued accreditation of the National Reference Laboratory and its regular participation in international proficiency-testing schemes for PCDD/Fs and PCBs in food and feed. In accordance with this accredited procedure, each analytical batch included the analysis of procedural blanks and of certified reference material, evaluated against the acceptance criteria established by Regulation (EU) No. 252/2012, and analytical results were released only when these quality-control criteria were met.

Results were expressed as WHO 2005-TEQ values (Van den Berg et al., 2006) for both PCDD/Fs and DL-PCBs, and as the analytical sum of the six NDL-PCB indicator congeners, all reported on an upper-bound basis in accordance with Regulation (EU) No. 252/2012 (European Commission, 2012). Measurement uncertainty was accounted for in the reporting of all results. Compliance was assessed against the maximum levels (MLs) established by Commission Regulation (EU) 2023/915 (European Commission, 2023) and its predecessor Regulation (EC) No. 1881/2006 (European Commission, 2006), as progressively amended. Notably, Regulation (EU) 2022/2002 (European Commission, 2022), subsequently consolidated into Regulation (EU) 2023/915 (European Commission, 2023), introduced with effect from 1 January 2023 a reduction of the ML for the sum of PCDD/Fs and DL-PCBs in raw bovine, ovine, and caprine milk from 5.5 to 4.0 pg WHO-TEQ/g fat, a regulatory tightening of direct relevance to the interpretation of results obtained in the final years of the study period. The action levels applicable throughout the study period were those set by Commission Recommendation (2013)/711/EU, as amended (European Commission, 2013, 2014).

2.4. Statistical analysis

All statistical analyses and graphical representations were performed using R statistical software (version 4.6.0), with the threshold for statistical significance established at $\alpha = 0.05$.

To evaluate the factors influencing environmental contamination in milk, linear mixed-effects models were employed. Separate analyses were conducted for four distinct target variables: the combined total of dioxins and dioxin-like polychlorinated biphenyls (PCDD/Fs + DL-PCBs), pure dioxins (PCDD/Fs), pure DL-PCBs, and non-dioxin-like PCBs (NDL-PCBs). By treating individual farms as a random effect across all models, the analysis accounts for the fact that samples originating from the same holding over time are naturally related. This approach ensures that the final estimates are mathematically reliable and not falsely distorted by specific operational or geographical features unique to individual holdings. The dependent variables were log-transformed before analysis to stabilise variance and prevent mathematical bias from non-detectable values, employing a small precautionary offset where necessary. Cattle were selected as the baseline reference group to which the other animal species were systematically compared. Because the response variables were modeled on the natural-logarithm scale, the estimated fixed-effect coefficients (β) were back-transformed to obtain interpretable effect sizes: the percentage change in concentration per additional kilometre of distance and per calendar year was computed as $(\exp(\beta) - 1) \times 100$, whereas the species contrasts relative to cattle were expressed as multiplicative factors equal to $\exp(\beta)$; the corresponding 95% confidence intervals were obtained by applying the same transformations to the confidence limits of β .

To ensure the models were robust and reliable, the total variance was partitioned to evaluate whether contamination differences were primarily driven by permanent differences between individual farms or by random background variation. Furthermore, the validity of the final

estimates was verified by assessing the distribution of the model residuals to ensure they met the standard assumptions of normality and symmetry required for stable, unbiased predictions.

3. Results

The variance partition revealed distinct behaviours between the contaminants. For pure dioxins, the variation between individual farms was estimated at zero, indicating a singular variance components structure where the residual unexplained variation ($\sigma^2 = 0.565$) represents the predominant source of variance. Similarly, for the total combined contamination, the isolated DL-PCBs, and the NDL-PCBs, the variance between individual holdings was remarkably low ($\sigma^2 = 0.0009$, $\sigma^2 = 0.0083$, and $\sigma^2 = 0.0058$, respectively), leaving the residual terms as the main contributors to the total variance. Taken together, these findings indicate that once the primary factors of distance, species, and calendar year are accounted for, the remaining differences in contamination levels are predominantly driven by short-term, random environmental fluctuations or unmeasured local factors rather than permanent, systematic differences between the farms themselves.

The analysis revealed contrasting spatial patterns between the chemical groups regarding their distance from the industrial plant. A highly significant, negative relationship was confirmed for the total combined contamination ($\beta = -2.145 \times 10^{-5}$, $SE = 8.337 \times 10^{-6}$, $p = 0.010$), the isolated DL-PCBs ($\beta = -2.413 \times 10^{-5}$, $SE = 9.159 \times 10^{-6}$, $p = 0.009$), and the NDL-PCBs ($\beta = -4.018 \times 10^{-5}$, $SE = 8.594 \times 10^{-6}$, $p < 0.001$). After converting the mathematical units from metres to kilometres, the models show that for every additional kilometre of distance from the ILVA plant, total combined concentrations decrease by approximately 2.12% (95% Confidence Interval [CI]: 0.52% to 3.71% decrease) (Fig. 3), pure DL-PCB concentrations drop by 2.38% (95% CI: 0.62% to 4.13% decrease) (Fig. 4), and NDL-PCB concentrations systematically decrease by 3.94% (95% CI: 2.31% to 5.55% decrease) (Fig. 5). The concentration of pure dioxins was not significantly affected by distance from the plant ($\beta = -1.222 \times 10^{-5}$, $SE = 8.719 \times 10^{-6}$, $p = 0.162$), with a 95% confidence interval ranging from a 2.88% decrease to a 0.48% increase per additional kilometre (Fig. 6).

Regarding differences between species, small ruminants exhibited a consistently higher potential to accumulate all categories of pollutants compared to cattle. This inter-species hierarchy remained strong and significant across all models, even when controlling for spatial and temporal variables. For the combined total contamination, levels in goats were 2.07 times higher than in cattle ($\beta = 0.729$, $SE = 0.066$, $p < 0.001$), and sheep showed concentrations 1.47 times higher than cattle ($\beta = 0.388$, $SE = 0.069$, $p < 0.001$) (Fig. 3). This pattern was most pronounced for pure dioxins (Fig. 6), where levels were 2.35 times higher in goats ($\beta = 0.856$, $SE = 0.069$, $p < 0.001$, 95% CI: 2.06 to 2.69) and 1.94 times higher in sheep ($\beta = 0.663$, $SE = 0.073$, $p < 0.001$, 95% CI: 1.68 to 2.24) relative to cattle. For isolated DL-PCBs, goats displayed concentrations 1.96 times higher than cattle ($\beta = 0.675$, $SE = 0.072$, $p < 0.001$, 95% CI: 1.71 to 2.26), while sheep levels were 1.30 times higher ($\beta = 0.265$, $SE = 0.076$, $p = 0.0005$, 95% CI: 1.12 to 1.51) (Fig. 4). NDL-PCBs in the small ruminant group deviated significantly from the general pattern. Although both species showed significantly elevated concentrations compared to the cattle, their relative ranking was inverted (Fig. 5). Specifically, sheep demonstrated a stronger bioaccumulation potential than goats, with contamination levels 1.72 times higher than those found in cattle ($\beta = 0.541$, $SE = 0.071$, $p < 0.001$, 95% CI: 1.49 to 1.97), whereas goats showed levels 1.28 times higher than cattle ($\beta = 0.250$, $SE = 0.068$, $p = 0.0002$, 95% CI: 1.12 to 1.47).

Between 2016 and 2025, all four models showed a statistically significant annual increase in total combined contamination, with an average of 5% per year ($\beta = 4.878 \times 10^{-2}$, $SE = 0.016$, $p = 0.003$, 95% CI: 1.76% to 8.38%) (Fig. 3). A comparable annual growth rate was observed for pure dioxins, which increased by an average of 5.15% per

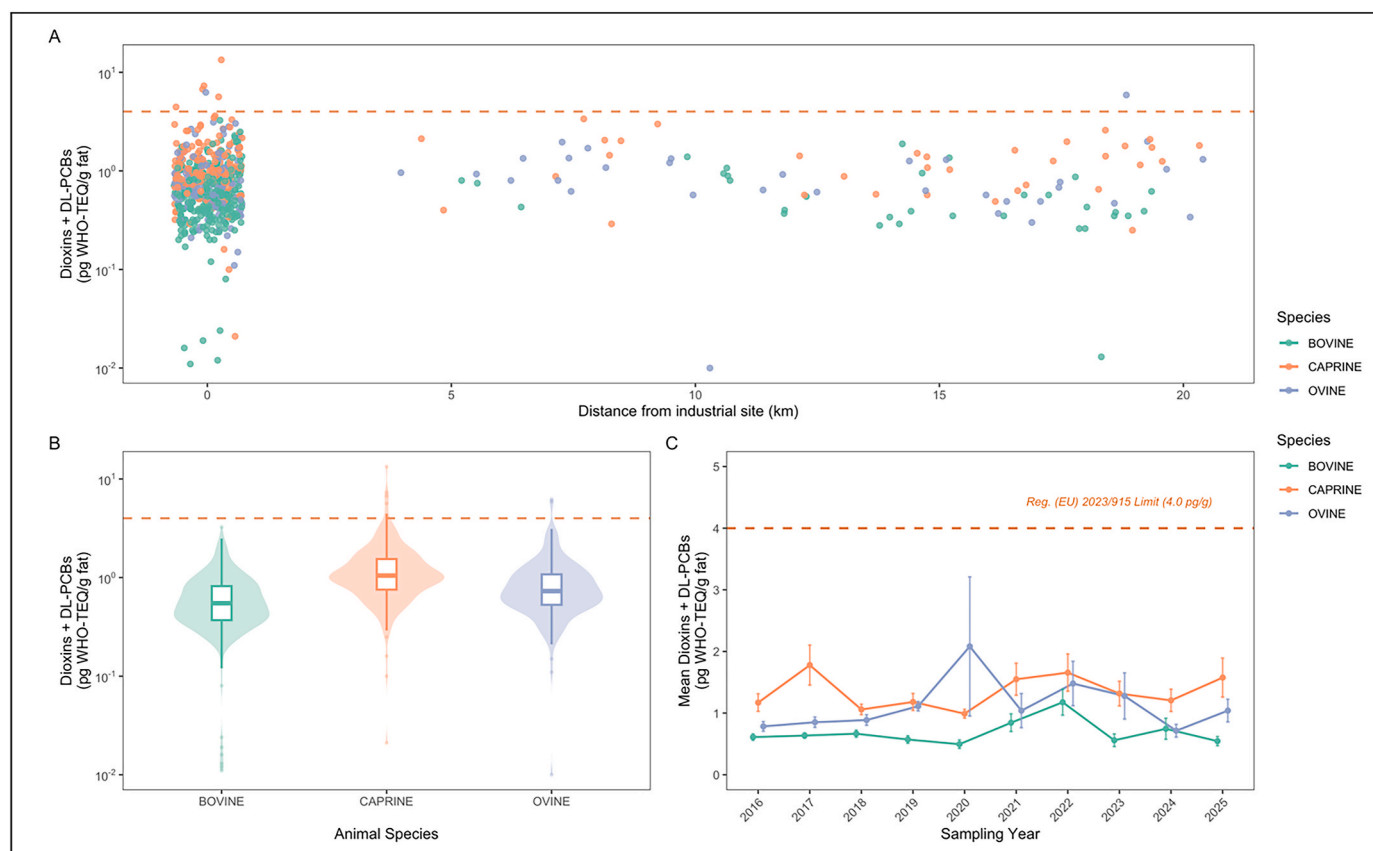


Fig. 3. Spatial, inter-species, and temporal profiles of combined dioxin and Dioxin-Like PCB contamination in milk.

year ($\beta = 5.023 \times 10^{-2}$, $SE = 1.688 \times 10^{-2}$, $p = 0.003$, 95% CI: 1.74% to 8.68%) (Fig. 6). Isolated DL-PCBs followed a similar trajectory, incrementing approximately 4.69% per year ($\beta = 4.583 \times 10^{-2}$, $SE = 1.769 \times 10^{-2}$, $p = 0.01$, 95% CI: 1.12% to 8.43%) (Fig. 4). Finally, NDL-PCB concentrations demonstrated an annual accumulation rate of approximately 5.28% per year ($\beta = 5.146 \times 10^{-2}$, $SE = 1.660 \times 10^{-2}$, $p = 0.002$, 95% CI: 1.91% to 8.80%) (Fig. 5). These parameters indicate a gradual increase in contamination across the decade within the sampled population. Because these annual coefficients were estimated from log-transformed concentrations, they describe the measured contamination level on a continuous scale and are independent of any regulatory threshold; they are therefore distinct from the classification of samples as compliant or non-compliant, which depends on the maximum levels in force and is affected by the 2023 revision of the regulatory limit discussed below.

The evaluations for all four models confirmed a satisfactory adherence to the assumption of residual normality, which validates the robust nature of the statistical tests applied to the dataset. The error distributions across the models were well-centred around zero with a symmetric, bell-shaped bulk. In all cases, the majority of the residuals fell between -1 and 1 , confirming that the models provide unbiased predictions for the core data. Negligible deviations were observed in the far-left tails of the distributions due to a small number of extreme negative residuals; these represent isolated samples that exhibited much lower contamination levels than expected, ranging between -4 and -6 . Given the large sample size of this study ($N = 671$), these slight deviations in the extreme tails are not statistically significant and do not compromise the mathematical validity and the associated p -values of the fixed effects estimates.

Figs. 3–6. Environmental contamination profiles in milk samples collected between 2016 and 2025 across bovine, caprine, and ovine species relative to the ILVA industrial site. Panel A illustrates how

individual contaminant levels change relative to the farm's distance from the industrial site. Panel B presents the overall distribution and density of contamination levels across the three animal groups, displaying the operational range, interquartile and median values through a combined violin and boxplot layout. Panel C delineates the chronological trajectory of the annual mean concentrations throughout the decade, where the vertical bars represent the standard error of the mean for each specific sampling year. The official European regulatory thresholds are indicated by horizontal dashed lines that represent the maximum limits set by Regulation (EU) 2023/915 (European Commission, 2023) for combined dioxins and dioxin-like compounds (4.0 pg WHO-TEQ/g fat), pure dioxins (2.5 pg WHO-TEQ/g fat), non-dioxin-like indicator PCBs (40 ng/g fat), alongside the EU Action Level for isolated dioxin-like PCBs (1.25 pg WHO-TEQ/g fat).

4. Discussion

This study provides one of the most temporally extensive and species-diverse longitudinal datasets on PCDD/F and PCB contamination in raw milk from a European peri-industrial agropastoral system. The linear mixed-effects framework (Bates et al., 2015) resolves four analytically distinct results whose interpretation advances both the environmental chemistry of persistent organic pollutants and the evidence base for risk-based food safety surveillance.

The significant inverse distance–concentration relationships for combined contamination ($\beta = -2.145 \times 10^{-5}$, $p = 0.010$), DL-PCBs ($\beta = -2.413 \times 10^{-5}$, $p = 0.009$), and NDL-PCBs ($\beta = -4.018 \times 10^{-5}$, $p < 0.001$) confirm that proximity to the industrial perimeter governs PCB burden in milk through the soil ingestion pathway, which accounts for 2–20% of daily dry matter intake in extensively managed ruminants (Scortichini et al., 2015; Weber et al., 2018). The steeper NDL-PCB decay (-3.94% per km) reflects the higher vapor pressure of

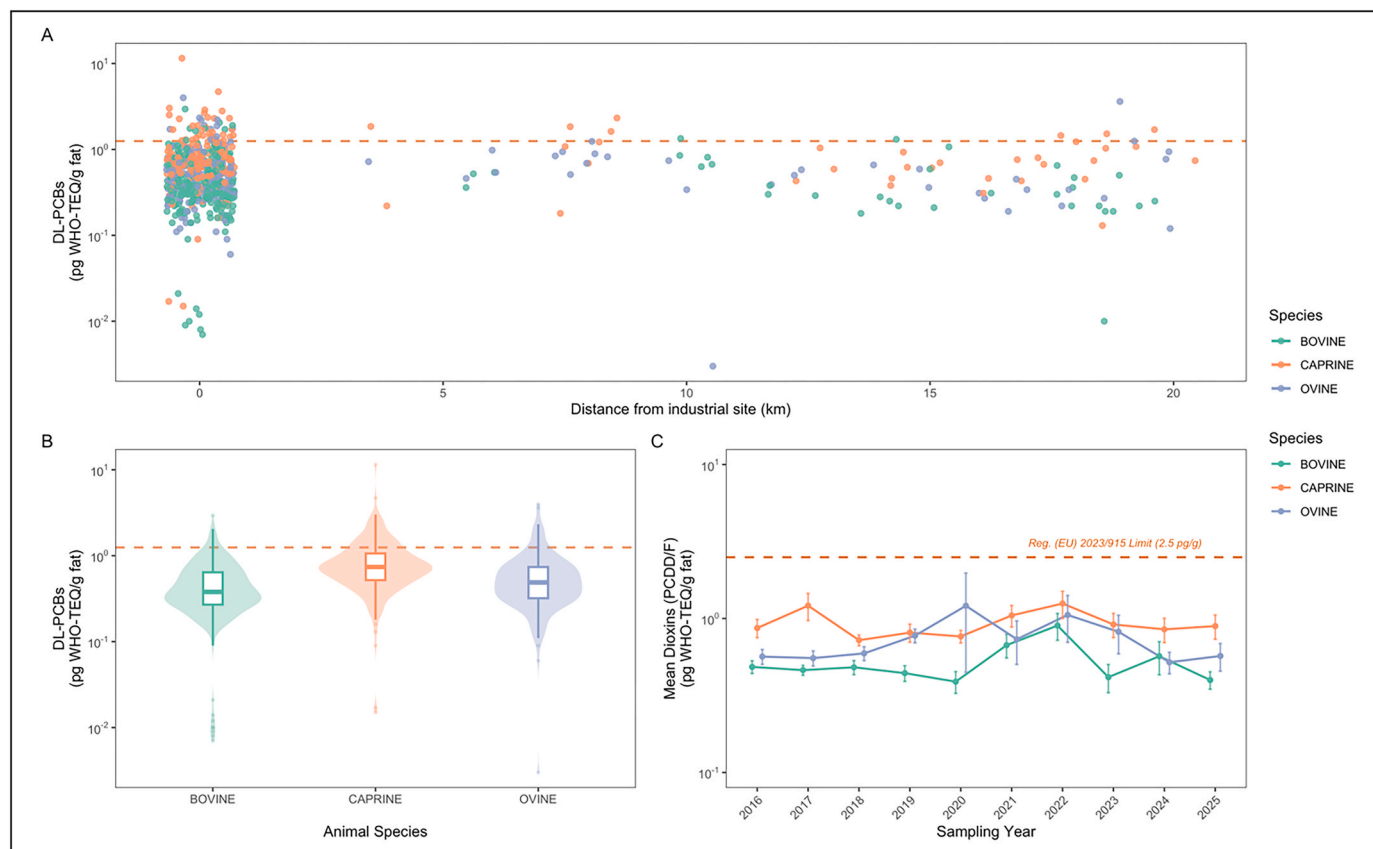


Fig. 4. Spatial, inter-species, and temporal profiles of Dioxin-Like PCBs contamination in milk.

lower-chlorinated congeners (Cousins and Mackay, 2001). Comparable gradients have been documented at the Brescia-Caffaro SIN (Brescia, Lombardy Region, northern Italy), where bovine milk reached tenfold EU limits on the most proximal farms (Turrio-Baldassarri et al., 2009), around incinerators in Tuscany Region (Battisti et al., 2022), and in Germany, where beef exceeds EU limits once soil PCB-TEQ surpasses ~ 5 ng/kg dry matter (Weber et al., 2018).

The absence of a spatial gradient for pure PCDD/Fs ($\beta = -1.222 \times 10^{-5}$, $p = 0.162$) is mechanistically expected: with log KOW values of 6.5–8.8 and soil half-lives spanning years to decades (Terzaghi et al., 2020), PCDD/Fs resist redistribution once soil-bound. Five decades of deposition, integrated through variable wind directions and soil mixing, are likely to have progressively homogenized the PCDD/F reservoir across the study radius. Although the present study analysed milk rather than repeated soil measurements, this interpretation is consistent with independent topsoil monitoring in the same territory (Liberatori et al., 2021) and with observations at comparable European legacy sites (Terzaghi et al., 2020), indicating that distance-based risk stratification alone is insufficient for PCDD/F management in legacy contaminated territories.

The attribution of this contamination to the industrial complex is supported by the characteristic congener signature of iron ore sintering. Sinter plants emit a PCDD/F profile in which polychlorinated dibenzofurans predominate over dibenzo-p-dioxins, a pattern stable enough across facilities and operating conditions to serve as a source fingerprint (Aries et al., 2006). The dioxin-like PCB congeners reported as most abundant in these emissions, PCB-118, -105, and -77, are among those quantified in the present study, and the sinter plant of the Taranto complex has been documented as the dominant local source of PCDD/F emissions (Esposito et al., 2014). The inverse distance gradient for PCBs, the inter-species hierarchy, and the analyte suite measured here are therefore consistent with atmospheric deposition from the

sintering process rather than with the refinery operating within the same industrial complex, whose organic micro-pollutant profile differs. A full sample-level source apportionment based on the complete homologue and congener distribution was beyond the scope of the routine WHO-TEQ compliance data analysed here and represents a priority for subsequent investigation.

The near-zero between-farm variance ($\sigma^2 = 0$ for PCDD/Fs, 0.0083 for DL-PCBs) confirms contamination is landscape-driven rather than farm-specific (Weber et al., 2018). Against this background, caprine milk exceeded bovine by 2.35-fold for pure PCDD/Fs ($p < 0.001$), 1.96-fold for DL-PCBs, and 2.07-fold overall; ovine milk exceeded bovine by 1.94-fold and 1.47-fold overall. Three converging mechanisms explain this hierarchy, schematically illustrated in Fig. 7: (i) close-to-ground grazing driving soil ingestion up to 20% of dry matter intake versus 2–5% in cattle (Scortichini et al., 2015); (ii) smaller body mass reducing adipose dilution (EFSA CONTAM Panel, 2018); (iii) and substantially higher feed-to-milk biotransfer, with carry-over rates in goats reaching 38.8% for TCDD and $>80\%$ for key DL-PCB congeners (Costera et al., 2005), and biotransfer factors in ewes reaching 1.24 day kg^{-1} lipids (Lerch et al., 2024). Elevated hepatic PCDD/F accumulation in sheep, up to fivefold higher than cattle, sustains a larger body burden for mammary excretion (Benedetto et al., 2016). This vulnerability is Union-wide: in the Netherlands, sheep fat showed 5.0% non-compliance versus 0% for bovine milk over a decade of national monitoring (Adamse et al., 2016); in Campania Region (southern Italy), ovine milk near industrial sites reached 3.89–12.90 pg g^{-1} fat (Esposito et al., 2010); in Lazio Region (central Italy), only ovine bulk milk exceeded regulatory limits across seven years of multi-species surveillance (Battisti et al., 2022), supporting the interpretation that the hierarchy quantified here is a recurrent feature of European peri-industrial dairy systems rather than a local particularity.

The inversion of the sheep-goat hierarchy for NDL-PCBs alone, ovine

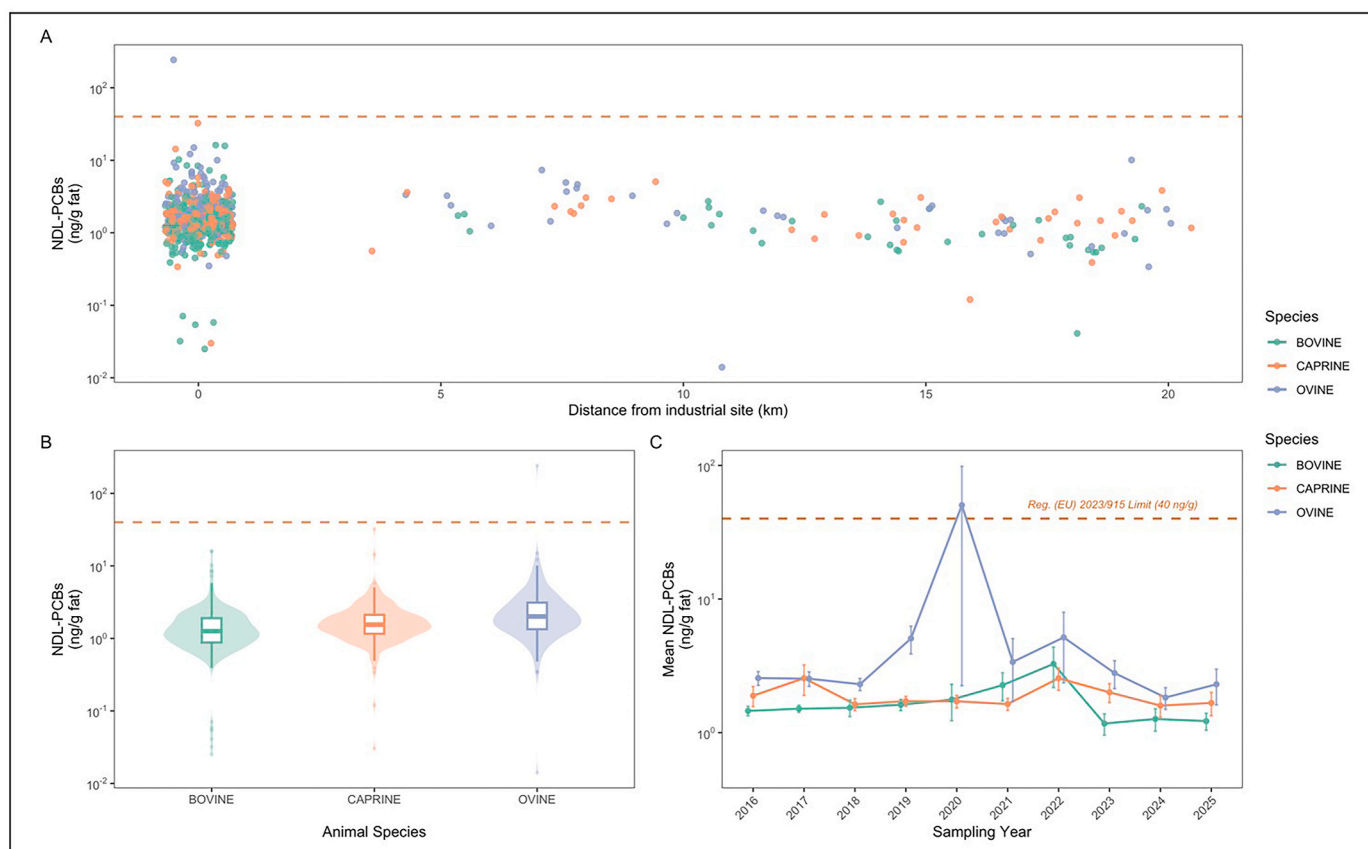


Fig. 5. Spatial, inter-species, and temporal profiles of Non-Dioxin-Like PCBs contamination in milk.

1.72-fold above bovine versus 1.28-fold for caprine, reflects the extreme adipose sequestration kinetics of PCBs 138, 153, and 180 (Cousins and Mackay, 2001; Driesen et al., 2022): a higher fat-to-lean mass ratio and lower milk-fat output in sheep paradoxically elevate fat-basis concentrations despite lower total excretion (Benedetto et al., 2016).

Compliance assessment against Regulation (EU) 2023/915 (European Commission, 2023) mirrors the spatial and inter-species patterns above: small ruminants, particularly goats, account for the largest share of non-conformities. A critical normative discontinuity must be considered when interpreting compliance trends: Regulation (EU) 2022/2002 (European Commission, 2022) reduced the maximum level for PCDD/Fs + DL-PCBs from 5.5 to 4.0 pg WHO-TEQ/g fat from 1 January 2023, a 27% reduction, reclassifying holdings as non-compliant whose actual contamination had not changed. The post-2023 apparent rise in non-conforming samples must therefore be read against this regulatory background, not attributed exclusively to environmental deterioration, a dynamic well documented in European food safety monitoring (EFSA CONTAM Panel, 2018; Malisch and Kotz, 2014).

The significant positive temporal trend across all contaminant groups, annual increases of 5.0% (combined, $p = 0.003$), 5.15% (PCDD/Fs, $p = 0.003$), 4.69% (DL-PCBs, $p = 0.010$), and 5.28% (NDL-PCBs, $p = 0.002$), reflects two intertwined factors that the surveillance design cannot fully disentangle. The first is environmental persistence: without active soil remediation, the bioavailable PCDD/F and PCB fraction cannot decline on operationally relevant timescales, as documented in Brescia (Lombardy Region, northern Italy) (Turrio-Baldassarri et al., 2009), in Germany (Weber et al., 2018), and in the Netherlands, where only minor decreases were observed over a full decade of national surveillance (Adamse et al., 2016). The second factor is the deliberate evolution of the monitoring plan towards a sentinel architecture, that is, the progressive concentration of sampling effort on a stable subset of historically high-contamination holdings monitored continuously as

early-warning indicators: systematic prioritization of historically high-contamination holdings concentrates analytical resources on the highest-risk stratum and maximizes non-compliant product detection, as recommended in Codex Alimentarius guidelines (Codex Alimentarius Commission, 2006) and EFSA monitoring guidance (EFSA CONTAM Panel, 2018). This is not a confound but a feature of a well-functioning system: the plan is increasingly detecting exactly what it is designed to detect. It must be stressed, however, that the Extraordinary Monitoring Plan is a risk-based food-safety instrument operated by the competent health authority, not an environmental cohort study: by protocol, holdings that return durable compliance are progressively de-prioritised in favour of those with persistent contamination, so that the composition of the sampled population evolves over time by design. The aggregate temporal coefficient estimated over the full dataset therefore reflects this deliberate, risk-based evolution of the sampling frame in addition to any underlying environmental change, and is best read as a food-safety surveillance signal rather than as a direct measure of environmental deterioration.

The Extraordinary Monitoring Plan at Taranto, with its decade-long continuity, three-species design, and sentinel architecture, offers a directly transferable model for European territories where industrial atmospheric deposition has shaped pastoral landscapes for decades. Three design principles emerge: proximity-based stratification should be supplemented by soil bioavailability assessment for PCDD/Fs; sampling intensity should be species-differentiated to reflect the demonstrated vulnerability of small ruminants; and sentinel farm continuity should be formally incorporated as a surveillance design element. In Italy, integrated risk-based farm categorization is already institutionalised through the ClassyFarm system, which ranks holdings by biosecurity, welfare, and antimicrobial-use indicators (Dimuccio et al., 2026); extending such frameworks to environmental contaminant exposure represents a logical development of the surveillance design principles

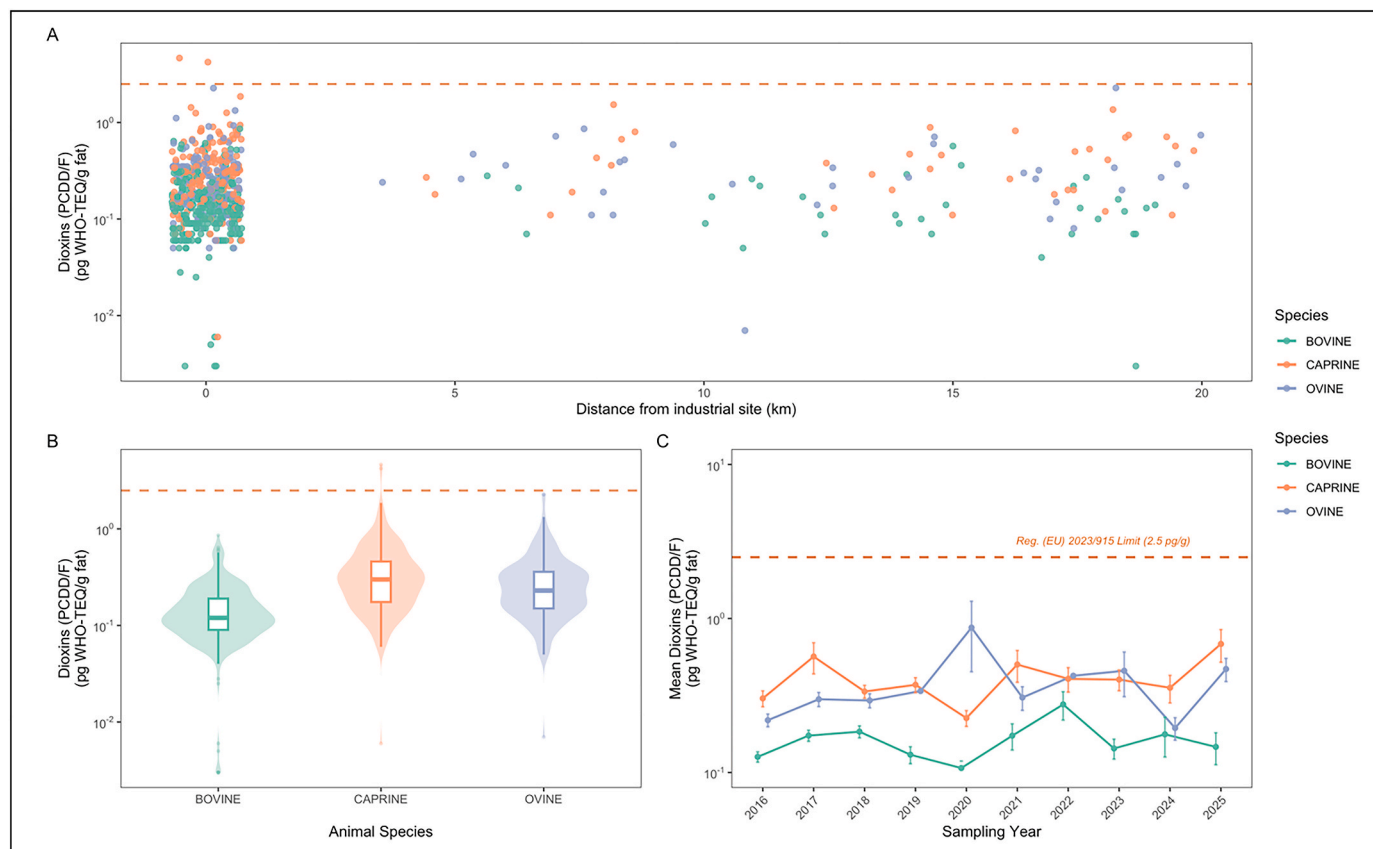


Fig. 6. Spatial, interspecies, and temporal profiles of pure dioxin contamination in milk.

proposed here.

5. Conclusions

This ten-year, three-species surveillance of PCDD/F and PCB contamination in raw milk from the Taranto peri-industrial territory provides one of the most temporally extensive datasets of its kind in the European literature, with findings of direct relevance to food safety governance beyond the local context.

The spatial analysis demonstrated a clear distance-dependent gradient for PCBs but not for PCDD/Fs, reflecting the differential environmental persistence and mobility of these compound classes. The absence of a distance gradient for PCDD/Fs is consistent with a spatially diffuse PCDD/F soil reservoir across the study area after more than five decades of continuous industrial deposition, an interpretation also supported by independent topsoil monitoring in the same territory (Liberatori et al., 2021). This indicates that distance from the emission source alone is an insufficient criterion for PCDD/F risk stratification in legacy contaminated territories.

The inter-species analysis confirmed a large and statistically robust hierarchy of contamination, with caprine and ovine milk consistently exceeding bovine levels across all contaminant classes. This pattern, supported by established toxicokinetic mechanisms and corroborated by comparable findings from the Netherlands, Campania Region (southern Italy), and Lazio Region (central Italy), indicates that small ruminants represent the most sensitive sentinel species for soil-mediated PCDD/F and PCB contamination in peri-industrial agropastoral systems.

The observed increasing temporal trend reflects the combined influence of sustained environmental contamination, in the absence of active soil remediation, and the progressive evolution of the monitoring plan towards a risk-stratified sentinel architecture. The latter, by concentrating sampling effort on holdings with historically elevated

contamination, increases the sensitivity of the surveillance system rather than indicating a deterioration in food safety performance. The reduction of the EU maximum level introduced in 2023 further contributes to the observed pattern of compliance and should be taken into account in any temporal interpretation of regulatory outcomes.

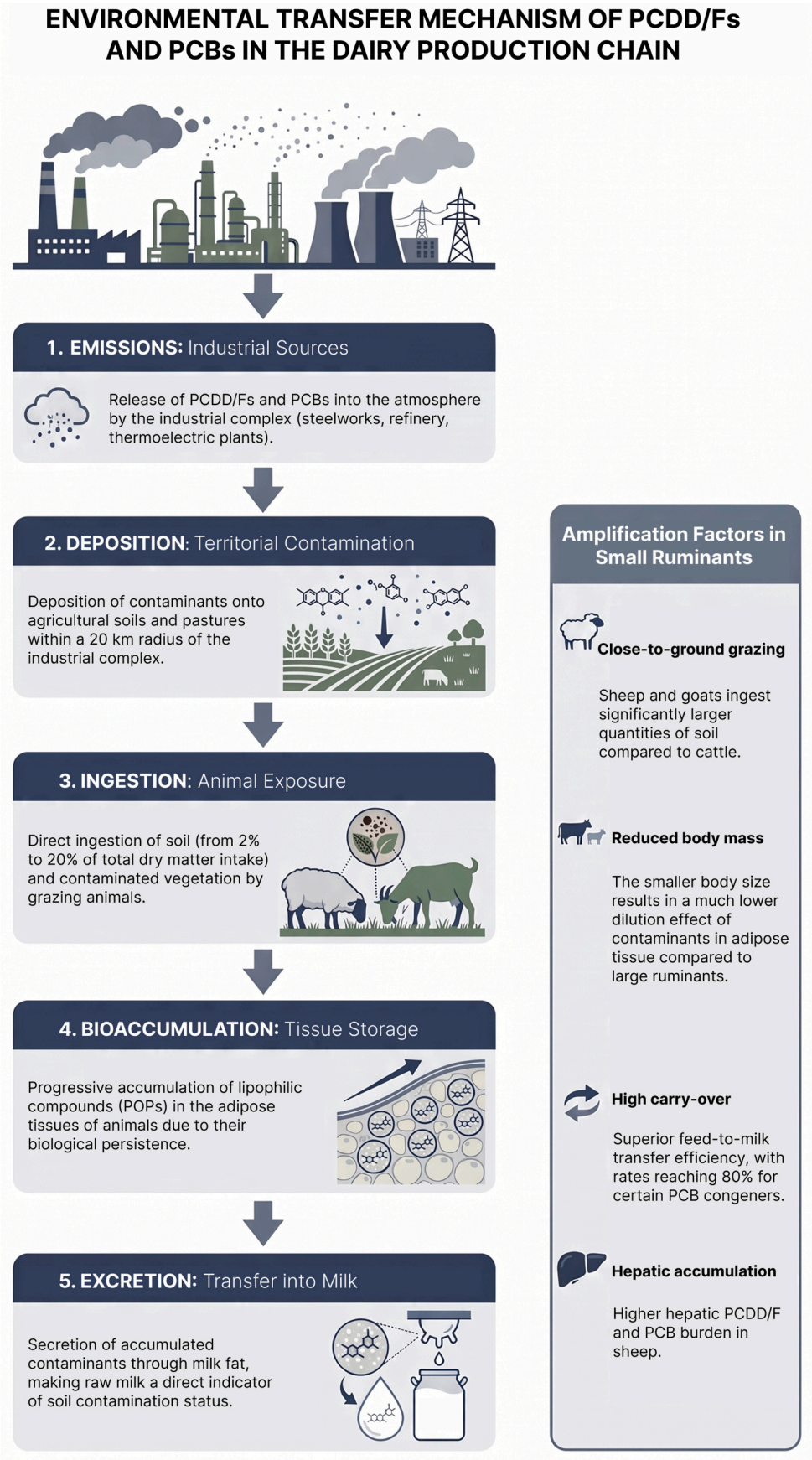
Taken together, these findings support three recommendations for the design of food safety surveillance in peri-industrial agropastoral territories across the European Union: the integration of soil bioavailability assessment alongside distance-based criteria for PCDD/F risk stratification; the adoption of species-differentiated sampling intensity to reflect the demonstrated vulnerability of small ruminants; and the formal incorporation of sentinel farm monitoring as a core element of long-term surveillance design.

CRediT authorship contribution statement

Conforti Virginia: Writing – review & editing, Writing – original draft, Visualization, Methodology, Conceptualization. **Capuozzo Flavia:** Writing – original draft, Visualization, Methodology. **Dimuccio Michela Maria:** Writing – original draft, Visualization, Methodology. **Basile Fabrizio:** Formal analysis, Data curation. **Franco Ettore:** Formal analysis, Data curation. **Pellegrino Angelo:** Formal analysis, Data curation. **Bonerba Elisabetta:** Visualization, Methodology. **Dambrosio Angela:** Writing – review & editing, Conceptualization. **Ceci Edmondo:** Visualization, Methodology. **Quaglia Cristiana Nicoletta:** Writing – review & editing, Conceptualization. **Bozzo Giancarlo:** Writing – review & editing, Conceptualization.

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Fig. 7. Schematic representation of the environmental transfer mechanism of PCDD/Fs and PCBs along the soil-to-milk pathway in extensively managed ruminants. The five sequential stages, atmospheric emissions from industrial sources, deposition onto agricultural soils and pastures, direct soil and vegetation ingestion by grazing animals, progressive bioaccumulation of lipophilic persistent organic pollutants (POPs) in adipose tissue, and mammary excretion into milk fat, constitute the primary contamination route in peri-industrial agropastoral systems. The right panel summarises the species-specific amplification factors that render small ruminants disproportionately sensitive to soil-mediated contamination relative to cattle.

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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