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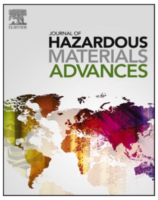

Contamination assessment and risk evaluation of organophosphorus pesticides in groundwater: A study on contamination patterns and implications

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ABSTRACT

Groundwater contamination by organophosphorus pesticides (OPPs) poses a serious environmental and public health threat, particularly in regions with intensive agricultural activity. While previous studies have focused on pesticide contamination in surface water, there is a lack of comprehensive data on OPP concentrations in groundwater and their associated health risks. This study addresses this gap by analyzing the occurrence, spatial distribution, and health risks of ten OPPs in 1168 groundwater samples from the Campania Plain, Southern Italy. The results revealed contamination hotspots in Caserta, Naples, and Salerno, with mean total OPP concentrations of 26.4, 26.0, and 24.9 ng/L, respectively. Dimethoate (13.5 %), chlorpyrifos (17.8 %), and parathion (13.0 %) were the most frequently detected pesticides, with chlorpyrifos persisting despite its EU ban in 2020. A human health risk assessment indicated that while non-carcinogenic risks (Hazard Index = 0.634) were below safety thresholds, carcinogenic risks exceeded acceptable limits for children, particularly for dichlorvos (4.26×10^{-3}) and dimethoate (5.89×10^{-3}). Statistical analysis, including Principal Component Analysis (PCA), revealed significant correlations between pesticide distribution and land use, emphasizing the role of intensive agriculture in groundwater contamination. This study highlights the urgent need for stricter regulations, improved groundwater monitoring, and sustainable pesticide management practices to mitigate health risks and protect water resources.

1. Introduction

In recent years, groundwater quality has been significantly impacted by anthropogenic pollution, making its protection a critical priority (Mallick et al., 2021). Population growth, intensive farming, waste management, and industrial activity are key contributors to groundwater contamination (Muhammad et al., 2015). Nearly 50 % of the global population relies on groundwater for drinking and other needs (Carrard et al., 2019), and its contamination poses serious health risks (FAO, 2018). Evaluating aquifer vulnerability is essential for monitoring and preventing pollution (Elzain et al., 2022; Chukwuma et al., 2023). Recent advancements in pesticide monitoring and groundwater quality assessment have led to the development of more sensitive and selective analytical techniques, such as high-resolution mass spectrometry

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(HRMS) and portable biosensors, enabling real-time detection of pesticide residues at ultra-trace levels. Additionally, spatial modeling approaches and machine learning algorithms have improved the prediction of contamination hotspots, facilitating targeted mitigation strategies. The integration of these technologies with long-term monitoring programs enhances the ability to track pesticide fate, degradation, and potential risks to human health. However, challenges remain in harmonizing regulatory standards and ensuring the continuous update of monitoring frameworks to include emerging contaminants.

Approximately 20 tonnes of pesticides are used annually worldwide, with Europe accounting for 4 5 % and the U.S. for 2 4 % (Rani et al., 2020). Organophosphate pesticides (OPPs), widely employed in agriculture and public health, have been in use for over 60 years (Derbalah et al., 2019). Synthesized from phosphoric acid and alcohols, they serve as fungicides, herbicides, and insecticides due to their cost-effectiveness and broad-spectrum activity. OPPs have largely replaced organochlorine pesticides (OCPs) due to their shorter environmental persistence, yet they still represent over 40 % of the global pesticide market (Montuori et al., 2022). However, their extensive use, coupled with runoff and improper disposal, has led to contamination of groundwater, surface water, and soil (Wang et al., 2021).

OPPs pose health risks due to their inhibition of acetylcholinesterase (AChE), leading to respiratory and neurological disorders (Wang et al., 2022). They also impact immune systems and aquatic life, affecting tissue histology and endocrine function (Sunanda et al., 2016; Amenyogbe et al., 2021). In addition to their well-known AChE inhibition, OPPs can exert multiple toxic effects on biological systems, compromising cellular homeostasis through mechanisms beyond neurotoxicity. One of the most worrying impacts is mitochondrial dysfunction, which alters oxidative phosphorylation, leading to reduced ATP synthesis and excessive production of reactive oxygen species (ROS). This oxidative stress triggers a cascade of harmful effects, including lipid peroxidation, protein oxidation and DNA damage, ultimately contributing to cellular apoptosis and tissue degeneration (Ray and Shaju, 2023; Gonçalves et al., 2021). Moreover, prolonged exposure to OPPs has been associated with endocrine dysfunction, as some compounds interfere with hormone biosynthesis, receptor binding and transduction signals, with potential reproductive toxicity effects, metabolic disorders and developmental abnormalities (ur Rahman et al., 2021). Immunosuppression effects have also been reported, with studies indicating alterations in the production of cytokines and a reduction in both innate and adaptive immune responses, thus increasing susceptibility to infections and inflammatory diseases. These systemic effects, often underestimated in relation to their primary neurotoxic action, highlight the environmental and public health risks associated with OPPs.Fatal human intoxications, mostly related to suicides, have been documented and provide valuable forensic data (Takayasu et al., 2012). Furthermore, OPPs degrade through volatilization, adsorption, oxidation, biodegradation, and hydrolysis, sometimes forming more toxic and persistent byproducts with endocrine-disrupting potential (Morin-Crini et al., 2022). Recent studies have increasingly highlighted the widespread presence of organophosphorus pesticides (OPPs) in groundwater, highlighting their persistence and potential health risks. Research conducted in several regions has reported detectable levels of OPPs in water sources, often above safety thresholds, especially in areas with high agricultural intensity (Wang et al., 2022; Pan et al., 2019; Liu et al. 2025). Advances in analytical techniques have improved the ability to detect OPPs residues, revealing their ability to infiltrate deep aquifers and persist despite regulatory restrictions. In addition, emerging studies have focused on the degradation pathways of OPPs in groundwater, showing that some transformation products may present a higher toxicity than the original compounds (Mitra et al., 2024).

This study evaluates the concentrations of ten OPPs: diazinon, dimethoate, malathion, dichlorvos, pirimiphos-methyl, fenitrothion, methidathion, tolclofos-methyl, parathion, and chlorpyrifos in groundwater from the Campania Plain (CP). This region was chosen as the study area because it has an intense agricultural activity, a high population density and historical concerns related to environmental pollution. This region is one of the most intensively cultivated areas in Italy, with a widespread use of pesticides that increases the risk of groundwater contamination (Zuzolo et al., 2020; Cafiero et al., 2019). In addition, the presence of different hydrogeological formations, including alluvial and volcanic aquifers, makes it a relevant case study to understand the mobility and persistence of organophosphorus pesticides (OPPs) in different groundwater systems. Furthermore, to its local relevance, the contamination patterns observed in the Campania plain reflect global challenges related to pesticide use in intensive agriculture

The study aims to: (i) assess OPP concentrations in CP groundwater, (ii) evaluate potential health risks, and (iii) examine pollutant distribution. The findings will contribute to understanding the environmental behavior of OPPs and underscore the need for improved management strategies to mitigate contamination and exposure risks.

2. Materials and methods

2.1. Study area: campania plain

The Campania Plain (CP), covering 13,600 km^2, is an administrative region along the Tyrrhenian margin of southern Italy. Its geology is dominated by sediments, carbonate rocks, and volcanic lithotypes, shaped by complex geological processes. The Volturno River Plain and the Sarno River Basin form its structural perimeter, including the southernmost CP, the eastern Naples Metropolitan Area, and southern Caserta Province (Fig. 1).

The area hosts extensive urban, industrial, and agricultural zones, with the northern part being the most industrialized. The main industries include vegetable canning, textiles, and apparel manufacturing (Albanese et al., 2007). Agriculture plays a crucial role in the regional GDP (Qu et al., 2022). The Mediterranean climate is characterized by hot, dry summers (25–31 °C) and mild, rainy winters (11–13 °C), with precipitation mainly between October and May (Catani et al., 2020).

The CP features diverse hydrostratigraphic systems, including alluvial, pyroclastic, karst, and silico-clastic aquifers. Groundwater flows from east to west (Ducci et al., 2017; Fusco et al., 2017). Alluvial aquifers, both coastal and internal, are key groundwater sources due to their high permeability. Volcanic aquifers include those in Ischia, Roccamonfina, Somma-Vesuvius, and Phlegraean Fields, valued for their thermal and mineral resources. Minor aquifers consist of Miocene–-Pliocene terrigenous formations and Cretaceous-Paleogene deposits. Water samples were collected from coastal plains (GAR, VCP, VES, SAR, SEL), volcanic districts (PHLE, VES), and carbonate massifs (MAS, LAT) (Montuori et al., 2023).

2.2. Sampling design

In recent years, extensive groundwater sampling has been carried out across the CP. These investigations encompassed all five provinces: Naples, Caserta, Salerno, Avellino, and Benevento. The analysis employed a Municipal Environmental Pressure Index (MIEP), developed through an advanced mathematical model. This model offers a valuable framework for geostratification in human biomonitoring studies and can be effectively utilized to guide remediation strategies and public health interventions (Pizzolante et al., 2021). The MEPI was increasingly recognized as a valuable tool for assessing environmental stressors at the municipal level. It integrates various environmental parameters to provide a comprehensive assessment of pollution levels and their potential impact on human health and ecosystems. It was specifically developed to assess environmental stressors in the Campania Region, providing a comprehensive evaluation of pollution levels and their potential impact on human health and ecosystems. This index integrates multiple environmental parameters, allowing for a detailed assessment of municipal-level environmental pressures. While initially designed for

Fig. 1. Maps of study area, Campania Region, Southern Italy.

Campania Region, its methodological framework can be adapted to different environmental contexts, making it a valuable tool for environmental monitoring, land-use planning, and policy-making.

Ten representative OPPs (diazinon, dimethoate, malathion, dichlorvos, pirimiphos-methyl, fenitrothion, methidathion, tolclofos-methyl, parathion and clorpirifos) were the principal targets and their specific chemical properties are indicated in Table S1 in Supplementary materials. Approximately 1200 samples were collected from the studied wells (with two aliquots collected for each sample) using pre-washed 2 L clear glass bottles. The bottles were then sealed immediately after sampling and transported to the laboratory at a stable temperature of 4 °C. Subsequently, all the samples were filtered using 0.45-μm glass fiber filters to eliminate coarse materials. On-site, physical parameters such as pH, temperature, and electrical conductivity (EC) were quantified using XS PC 70 Vio sensors and results are reported in Table S2.

2.3. *Analytical approaches*

2.3.1. *Sample treatment and evaluation*

Groundwater samples were extracted following the methodology proposed by Montuori et al. (Montuori et al., 2022). Briefly, groundwater samples (500 mL) were preconcentrated using SPE Oasis HLB cartridges (6 mL, 500 mg; Waters, Milford, MA, USA). The extracts were eluted and analyzed by GC–MS. A mixture of Semi-volatile organic compound (SVOC) surrogate standards (2-fluorobiphenyl, nitrobenzene-d5, p-terphenyl-d14, 2-fluorophenol, phenol-d5, 2,4,6-tribromophenol) was introduced into the samples before extraction to assess the effectiveness of the analytical method. However, SVOC internal standard (acenaphtene-d10, crysene-d12, 1,4-dichlorobenzene-d4, naphtalened8, perylene-d12, phenantrene-d10) mixtures were used as internal standards and they were added to each sample immediately before injection to facilitate the quantification of target analytes through the internal standard method.

2.3.2. *Instrumental analysis*

A Gas Chromatograph TRACE™1310 coupled to a ISQTM7000 Single Quadrupole Mass Spectrometer (GC–MS, Thermo Scientific, Waltham, MA, USA) was used for qualitative and quantitative analysis of OPPs in groundwater samples,. The GC–MS had a TG-5MS capillary column (30 $m \times$ 0.25 mm i.d. $\times$ 0.25 m film thickness), and used high purity helium as the carrier gas at constant flow rate of 1 mL/min. The injection port and detector temperature were set as 250 and 280 °C, respectively. The oven temperature was set according to the following program: 35 °C for 3 min, increasing to 100 °C at 25 °C min^{-1} (kept for 4 min), to 280 °C at 30 °C min^{-1} (kept for 4 min), and finally, to 320 °C at 10 °C min^{-1} (kept for 1 min). Compound identification was achieved by comparing retention times with reference standards and verifying the characteristic ions and their ratios for each target analyte. For samples with higher concentrations, identification was further confirmed in full-scan mode. The concentrations of OPPs were determined using calibration curves.

2.3.3. *Quality assurance and control (QA/QC)*

Prior to sampling, each bottle was rinsed three times with the water sample to be collected. The analysis was conducted multiple times, and blank samples were analyzed to verify the precision of the extraction process. The limits of detection (LODs) and quantification (LOQs) were estimated as three and ten times, respectively, according to criterion of the signal /noise (S/N) level for individual analyte. In particular LODs values were in the range of 0.0030–0.0063 μg L^{-1} and LOQ were in the range of 0.0100–0.0210 μg L^{-1}. OPPs quantification was evaluated using a five-point calibration curve (10–50–100–500–1000 ng/L) for ten OPPs ($r^2 > 0.995$). The quantification of individual compounds was performed by comparing the peak areas with those of the recovery standards (between 70 and 130 %). Standard reference solutions were employed to monitor the performance of the instruments, which were calibrated daily. The relative percent differences between the five-point calibration and the daily calibrations were kept below 20 % for all target analytes.

2.4. *Human health risk estimation in groundwater samples*

The health risk assessment for OPPs in groundwater was conducted to evaluate potential adverse effects on human health. This analysis involved calculating both non-carcinogenic, cumulative risks and carcinogenic risks assessed for adults, children, and infants. The risk evaluation followed the guidelines established by the United States Environmental Protection Agency (EPA) (USEPA, 2017).

The use of these models for OPPs risk assessment in this study represents a suitable approach to the complex environmental and health challenges of the Campania Region. Traditional risk assessment methods are often based on direct concentration thresholds, which may not fully capture the cumulative and synergistic effects of multiple contaminants. Using these models, this study integrates spatial distribution models, human exposure pathways and toxicological data to provide a more comprehensive and predictive risk assessment. These models enable the identification of high-risk areas, facilitating targeted interventions and improved regulatory strategies. Application of these models in this study improves the accuracy of health risk estimates, providing a valuable tool for environmental monitoring and policy making in regions with extensive agricultural and industrial activities.

The non-carcinogenic risk for each OPP was quantified using the Hazard Quotient (HQ), calculated using Eq (1):

$$HQ = ADD/RfD \tag{1}$$

where ADD is the average daily dose of the pesticide in groundwater (mg/L), and RfD is the reference dose for the pesticide (mg/kg/day). RfD values for OPPs were procured from the Integrated Risk Information System (USEPA, 2017). HQ>1 showed that the exposed individual is adversely affected.

For ADD calculation the average daily intake (D_{IP}) was used and Eq. (2) was employed:

$$D_{ip} = C \times IR \tag{2}$$

where C is the concentration of OPPs in groundwater (mg/L) and IR (L/day) is the ingestion rate of water (2, 1 and 0.75 for adults, children and infants, respectively).

The D_{IP} was converted to an average daily dose (ADD) by dividing the average body weight, given below, where BW body weight (kg) for infants, children and adults is 5, 10 and 60 kg, respectively (WHO, 2008; Ali et al., 2018). Eq. (3) was used:

$$ADD = D_{ip}/BW \tag{3}$$

For multiple OPPs detected in the same samples, the cumulative risk was assessed using the Hazard Index (HI), which is the sum of the individual HQ values as shown in Eq. (4):

$$HI = \sum HQ_i \tag{4}$$

where HQ_i represents the Hazard Quotient for the i th pesticide. An HI value greater than 1 indicates potential health risks from cumulative exposure.

To calculate the Carcinogenic risk (CR) of groundwater exposure, Eq. (5) was used:

$$CR = CDI \times SF \times ADAF \tag{5}$$

the Chronic daily intake (CDI) is calculated for different exposure pathways, such as ingestion, inhalation, or dermal contact. However for this study, the oral exposure pathway was considered, as it is regarded as the primary exposure route. For ingestion the Eq. (6) was:

$$CDI = C \times IR \times EF \times ED/BW \times AT \tag{6}$$

where C is the concentration of OPPs in groundwater (mg/L); IR (L/day) is the ingestion rate of water (2, 1 and 0.75 for adults, children and infants, respectively); EF = exposure frequency (days/year); ED exposure duration for infants, children and adults is 2, 6 and 70 years, respectively; BW body weight (kg) for infants, children and adults is 5, 10 and 60 kg, respectively (WHO, 2008) and AT (lifetime for non-carcinogenic effects) is day. The default parameters for IR, EF, ED, and BW were chosen based on the EPA's recommendations (USEPA, 2017; Ali et al., 2020; Wang et al., 2022).

The slope factor (SF) was a quantitative value used in risk assessment to estimate the risk of cancer associated with exposure to a carcinogenic substance and ADAF were based on scientific evidence indicating that exposure to carcinogens during critical growth periods can result in greater cancer risks compared to similar exposures in adults. The EPA agency recommends the following ADAF values for age-specific adjustments 1, 3 and 10 for adult, children and infant, respectively.

The acceptable risk of CR from OPPs for a regulatory purpose is between 10^{-6} and 10^{-4}. (USEPA, 2017). All the parameters used for the calculation of non-carcinogenic and carcinogenic risk are reported in Table S3 of the supplementary materials.

2.5. *Statistical analysis and spatial distribution*

The statistical analysis was conducted through multiple methodological steps to assess the distribution of pesticides across the analyzed provinces and identify significant patterns in their presence. The descriptive analysis reports, the distribution of the variables, the median [q1 = first quartile; q3 = third quartile] for continuous variables and the percentage for categorical variables. The Kruskal–Walli's test was used to assess whether statistically significant differences existed between provinces (AV=Avellino, BN=Benevento, CE=Caserta, NA=Napoli, SA=Salerno) for the continuous variables under study, and if a $p \leq 0.05$ was found, Dunn's post hoc test was applied. This provided an initial overview of pesticide concentration distributions and highlighted potential regional differences. To reduce dataset dimensionality and identify latent patterns in pesticide distribution, Principal Component Analysis (PCA) was applied. This technique transformed the original variables, corresponding to the analyzed pesticides, into a reduced number of principal components, each representing a linear combination of the original variables. The scree plot analysis guided the selection of the optimal number of components, indicating that the first three principal components explained 69.8 % of the total variance, allowing for a meaningful representation of the dataset while minimizing information loss. The weights of each pesticide in the principal components were determined using orthogonal rotation, which aimed to improve the interpretability of the components by maximizing the contrast between the contributions of the original variables to each component. This process facilitated a clear distinction between the dominant pesticides in each component and enhanced the interpretation of distribution patterns. Finally, to visualize the territorial distribution of the principal components, geographic maps were generated, showing the average component scores for each province (PC1, PC2, and PC3). These maps allowed for the spatial analysis of pesticide presence, variability and helped explore potential associations with local agricultural practices and phytosanitary management strategies. Statistical analysis was performed using the R environment for statistical computing and graphics version 4.3 (R Foundation for Statistical Computing, Vienna, Austria).

3. Results and discussions

3.1. *OPPs concentrations in groundwater samples*

In Table 1, the results of OPPs analyzed in this study are presented. A total of 1168 groundwater samples were collected across the Campania Region, and the data were grouped according to the five provinces: Naples, Caserta, Salerno, Avellino, and Benevento. Total OPPs concentrations for all locations varied from N.D to 47.5 ng/L, with a mean value of 26.6 ng/L (Fig. 2). Based on the division of the study area, the results revealed that groundwater from the Caserta, Napoli and Salerno provinces had the greatest concentrations of OPPs, with a mean value of 26.4, 26.0 and 24.9 ng/L (mean values), respectively. Avellino and Benevento's OPPs concentrations were reported to be 23.0 and 21.0 ng/L (mean values), respectively. Comparing the concentrations found in this study with those reported in recent groundwater research, we can conclude that the concentrations of pesticides in the Campania Region aquifers are significantly lower than those in other study areas. Wang

Table 1
Minimum, maximum, mean, and total values related to OPP concentrations in groundwater samples from Campania Plain.

OPPs (μg L^{-1})	Min	Max	Mean	Total
Diazinon	0.010	0.086	0.039	21.4
Dimethoate	0.010	0.280	0.060	36.0
Malathion	0.013	0.099	0.041	22.7
Dichlorvos	0.011	0.870	0.049	28.4
Pirimiphos-methyl	0.011	0.098	0.042	23.9
Fenitrothion	0.011	0.096	0.047	26.1
Methidathion	n.d	n.d	n.d	n.d
Tolclofos-methyl	0.010	0.090	0.042	25.5
Parathion	0.011	0.230	0.052	34.6
Clorpirifos	0.011	1.100	0.042	47.5

n.d: not detected.

et al. (2022) observed particularly high concentrations for the pesticides Dimethoate (321,79 ng/L in summer and 268,8 ng/L in winter), Dichlorvos (306,79 ng/L in summer and 252,74 ng/L in winter) and Malathion (174,40 ng/L in summer and 152,27 ng/L in winter). Similarly, Huang et al. (2019) in China found OPPs at all sampled sites ranging from 128 to 3973 ng/L, showing a very high percentage of Methyl Parathion (96.7 %) and Diazinon (80 %). Pan et al. (2019) also reported extremely high concentrations, with a range between 1738.8 and 2194.3 μg/L. In particular, its study recorded elevated mean values for Chlorpyrifos (329.5 ng/L) and for Parathion (364.2 ng/L).

The higher concentrations of OPPs in the provinces of Caserta, Salerno, and Naples compared to Benevento and Avellino can be attributed to several factors. First, these provinces were characterized by more intensive agricultural activities, with a higher prevalence of crops that are particularly susceptible to pest infestations, such as fruits and vegetables. This leads to a greater use of chemical pesticides, including OPPs (Campania Region, 2015). The region is characterised by extensive cultivation of vegetables, fruits and cereals, which require significant pesticide application. In particular, areas such as Caserta, Naples and Salerno have a high density of farms where pesticides can infiltrate the soil and reach the aquifers through infiltration and percolation. In addition, surface water irrigation, commonly used in the region, can introduce additional pesticide residues into aquifers, especially in areas near rivers such as the Volturno and the Sarno. Another important source of contamination is industrial activity, particularly in areas with problems of agrochemical production and waste disposal. Illegal dumping and inadequate waste management in parts of Campania aggravates the problem, as improperly disposed pesticides and agricultural runoff contribute to groundwater pollution. In addition, urban runoff from parks and residential areas can further introduce pesticide residues into surface and underground water systems.

Second, the region’s proximity to major urban areas and agricultural

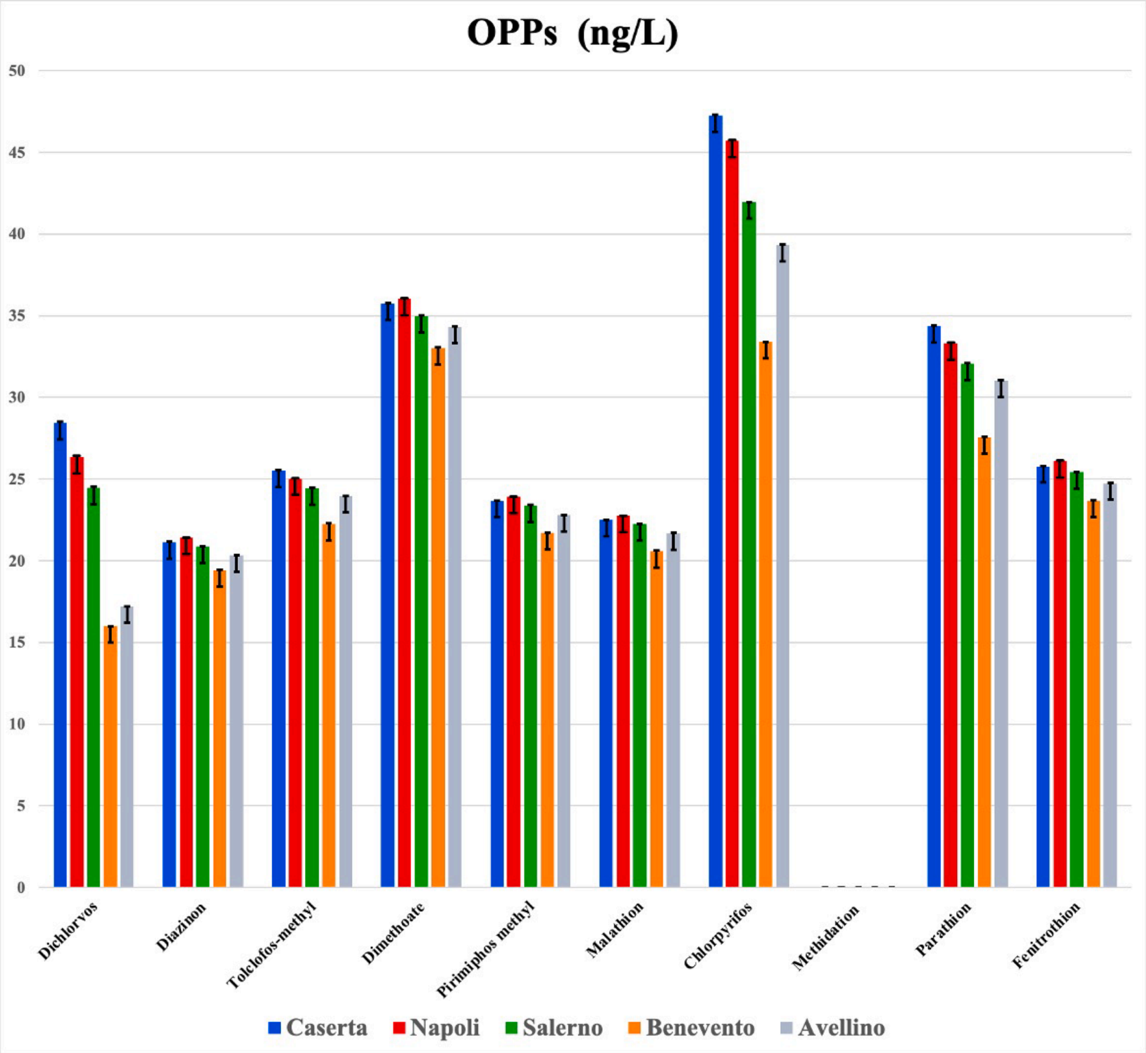

Fig. 2. Total values ± standard deviation (SD) of OPPs in groundwater samples. The results were divided based on the five provinces in Campania Region. For each point, $n = 3$ replicates were performed.

markets may increase pesticide application rates due to the demand for high yields and the competitive nature of agricultural production. Additionally, the differences in land use, with Caserta, Salerno, and Naples having more expansive and commercialized agricultural fields, may result in higher pesticide usage compared to the more rural and less intensively cultivated areas of Benevento and Avellino. Furthermore, the environmental conditions, including irrigation practices and climatic factors, can influence the persistence and dispersion of OPPs in the environment, contributing to higher concentrations in specific areas. Lastly, differences in agricultural practices, such as the use of more advanced or less regulated pesticide application techniques, may also play a significant role in the observed variations (Campania Region, 2015).

Among all the compounds analyzed, the highest concentrations were detected for dimethoate, chlorpyrifos, and parathion, with percentages of 13.5 %, 17.8 %, and 13.0 %, respectively, relative to the total concentrations detected for all sampling sites. In fact, these compounds were the most ubiquitous as they were detected in almost all groundwater samples. In addition, dichlorvos, diazinon, tolclofos-methyl, fenitrothion, malathion and pirimiphos-methyl were found in medium/low concentrations, contributing 10.6, 8.0, 9.0, 8.9, 8.5 and 9.0 %, respectively, to the total amount of OPPs. Only methidathion compound, among the ten compounds analyzed, that was consistently absent in all groundwater samples distributed throughout the study area. Among the compounds detected in groundwater, chlorpyrifos was one of the main contaminants. This pesticide was banned by the European Union as of April 1, 2020, due to its severe effects on DNA and its negative impact on the development of the nervous system. In 2019, the EFSA published a detailed assessment concluding that it was impossible to establish a safe exposure level for chlorpyrifos, highlighting significant risks of genotoxicity and neurotoxicity, particularly in children (EFSA, 2019). These findings are supported by numerous studies, including those conducted by Rauh et al. 2011 and Mit et al., 2021 which documented the effects of low-dose chlorpyrifos exposure and its association with reduced cognitive development in children exposed during the prenatal phase. The elevated concentrations of dimethoate and parathion in groundwater can be explained by a combination of factors, primarily related to their physicochemical properties. Specifically, dimethoate, due to its high solubility (25,900 mg L^{-1}), readily moves towards groundwater through the infiltration process. Additionally, its hydrophilic nature ($\mathrm{LogK_{ow}} = 0.78$) reduces soil adsorption, increasing its mobility (Montuori et al., 2022; Narayanan et al., 2022). Although parathion is less soluble than dimethoate, it is moderately persistent and can be transported through the soil, especially in areas with low retention capacity. While both compounds have been banned or used under restrictions in the European Union, they were widely used in maize cultivation and olive oil production in the past. Furthermore, in many non-European countries, these compounds are still used in certain agricultural applications, prolonging their environmental persistence, particularly in aquatic environments (Ore et al., 2023).

The results obtained are consistent with the aforementioned findings and with the existing literature. This is further confirmed by previous studies, which indicate that the pesticides with the highest concentrations are not only those most commonly sold in the study area but also those with the highest levels detected in the surface waters analyzed within the same regions of interest.

The results of the study conducted by Triassi et al. 2019 on the surface waters of the Volturno River indicated that, in the province of Caserta, the highest concentrations of the analyzed OPPs were those of chlorpyrifos and dimethoate, with mean values of 3.71 and 1.85 ng/L, respectively. Similarly, the study on the surface waters of the Sele River in the Salerno area confirmed comparable results. Montuori et al. 2022 report that in surface waters, particularly near the river mouth, higher concentrations of OPPs were detected, with chlorpyrifos, dimethoate, and parathion showing mean values of 0.64, 1.50, and 0.62 ng/L, respectively. These data provide further evidence of the environmental contamination caused by these chemical compounds in some areas of the Campania Region, where the pollution appears to be widely distributed. This comparison can confirm factors that influence the distribution of OPPs, highlighting a strong correlation between groundwater and surface water in these areas.

The observed discrepancy between OPPs concentrations in groundwater and surface water highlights critical environmental and hydrological processes influencing contaminant distribution (Wendell et al., 2024). While surface water samples in the study area reported a maximum Chlorpyrifos concentration of 3.71 ng/L, groundwater samples exhibited a significantly higher mean concentration of 42 ng/L, with peak values reaching 1100 ng/L. This more than tenfold increase suggests the influence of multiple factors, including groundwater recharge dynamics, pesticide transport mechanisms, and differences in degradation rates between surface and subsurface environments.

One key factor contributing to this discrepancy is the infiltration and percolation of pesticides through soil layers, which is particularly pronounced in agricultural areas where irrigation is extensive. Surface water, often enriched with pesticide residues due to runoff from treated fields, can seep into underlying aquifers, especially in regions with high soil permeability and fractured geological formations (Wang et al., 2022). Additionally, the use of contaminated surface water for irrigation may introduce OPPs directly into the soil, where they can subsequently leach into groundwater systems. The physicochemical properties of specific pesticides further influence their environmental behavior; for instance, dimethoate's high solubility and low soil adsorption capacity facilitate its transport through the soil profile, increasing its likelihood of reaching groundwater. In contrast, compounds with lower solubility and higher adsorption potential may be retained in surface environments for longer periods, leading to their lower concentrations in groundwater (Araya et al., 2024).

Another crucial aspect to consider is the persistence of OPPs in groundwater compared to surface water. In surface water bodies, pesticides are more exposed to photodegradation, microbial activity, and volatilization, leading to relatively faster degradation rates. However, in groundwater, where conditions are often anaerobic and microbial activity may be limited, pesticide breakdown can be significantly slower. This prolonged persistence means that even pesticides that have been banned or restricted for several years may still be detected in groundwater at concerning concentrations (Wang et al., 2021).

In summary, the study highlights significant OPPs contamination in groundwater across the Campania Region, with marked variations between provinces, driven by the physicochemical properties of specific pesticides and their historical use in agriculture; moreover, the correlation between groundwater and surface water contamination underscores the need for comprehensive management strategies to mitigate environmental and public health risks. Such a correlation could be the focus of attention and further investigation in future studies. Moreover, an important point to highlight is that the persistence and bioaccumulation of OPPs have implications not only in environmental science but also in forensic medicine, contributing to toxicological analyses to determine chronic exposure, poisoning cases, or correlations with environmentally induced diseases.

3.2. Risk assessment of OPPs for human health

The results of the assessment for non-carcinogenic and carcinogenic risks were based on average concentration values expressed in mg/L, with data primarily referring to oral exposure. Among the ten OPPs analyzed, all were evaluated for non-carcinogenic risk, while only Dichlorvos, Diazinon, Dimethoate, Malathion and Parathion were assessed for carcinogenic risk, as these compounds have been classified by the IARC as possibly or probably carcinogenic to humans. In non-carcinogenic risk assessment, all pesticides studied reported an HQ value lower than one for both adults and children, including infants (Fig. 3a). These results, presented in Table 2 suggest that it is unlikely that these contaminant compounds would have negative health effects through oral exposure. Indeed, in accordance with the updated US EPA guidelines, a value higher than one indicates that water consumption is not recommended due to potential adverse health effects. The HI results was 6.34×10^{-1}, indicating a low potential health risk from cumulative exposure. It is essential to remember that children and infants are the most vulnerable age groups in the population, as even minimal exposure to these contaminants can impair their health and growth, as reported in the literature (Perera-Rios et al., 2022; Rincón-Rubio et al., 2024).

According to IARC classification, in carcinogenic risk evaluation, only pesticides classified as possible or probable carcinogens were assessed, including Dichlorvos, Diazinon, Dimethoate, Malathion, and Parathion. The results obtained were deemed acceptable for both adults and children, including infants (Fig. 3b). The risks associated with the exposure of children exceeded the parametric values of 4.26×10^{-3} and 5.89×10^{-3} for Dichlorvos and Dimethoate, respectively recommended by USEPA for carcinogens compounds (Table 2).

It is simple to reconstruct the lesivity associated with acute intoxication in both postmortem and emergency medical care situations (Dharmani et al., 2005). Chronic effects of OPP intoxication that involve central nervous system have been decribed in literature for years. In Forensic Pathology it is easy to correlate OPP lesivity and acute intoxication when a suspect is consistent. Nevertheless, it is difficult to determine a casual link in forensic field between OPP exposure and carcinogenesis due to the fact that in Italy, from a judicial point of view, in penalistic field the limit of "beyond reasonable doubt" has to be achieved, whereas in civilistic judgement a consistent "probability" is sufficient. In order to establish a causal link between exposure to

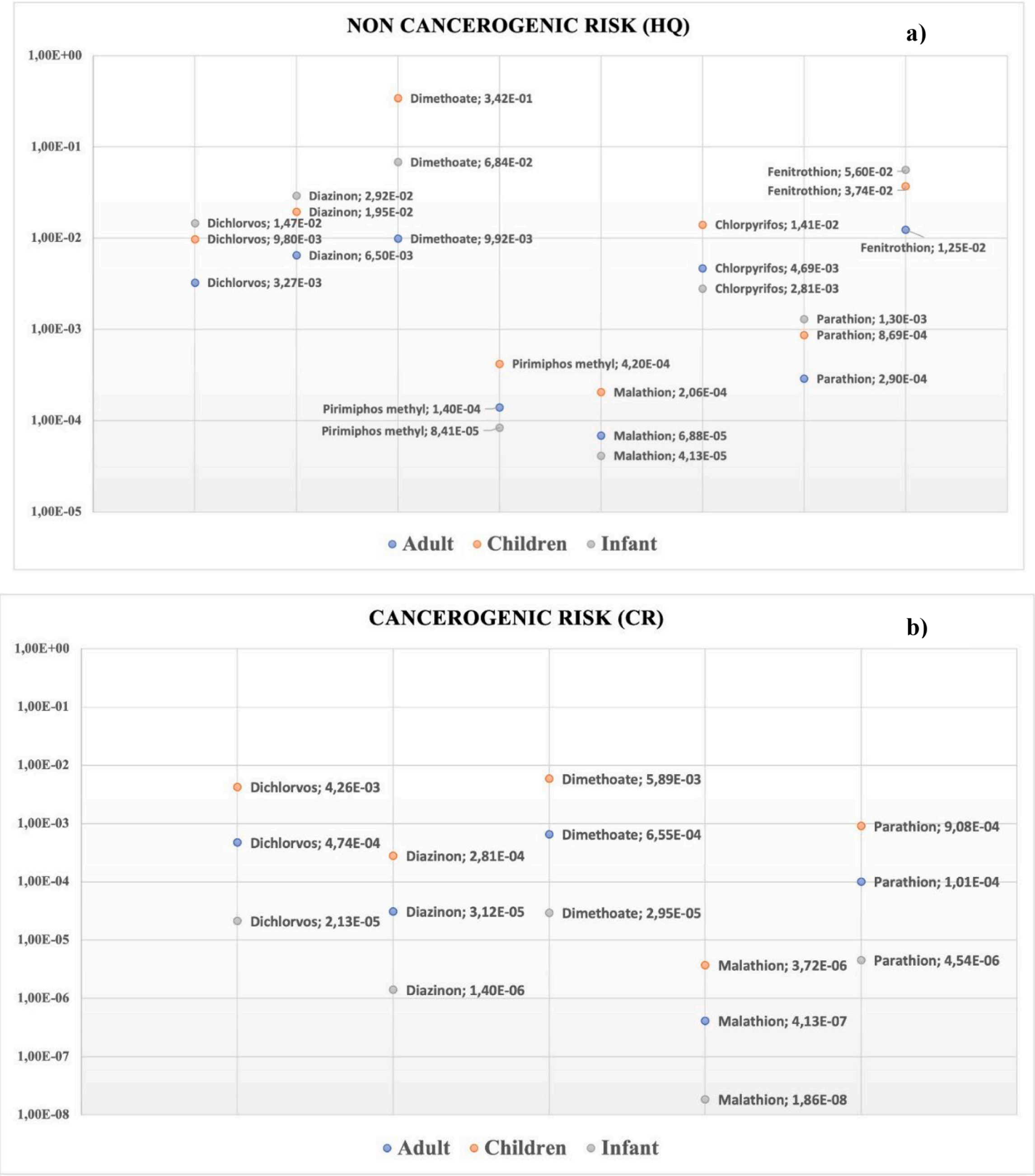

Fig. 3. a and 3b Non Cancerogenic Risk (HQ) and Carcinogenic Risk (CR) of the detected OPPs in groundwater samples of the Campania Region. All of pesticides studied reported an HQ value lower than one for both adults and children, including infants, instead Dichlorvos and Dimethoate, highlighted in black, exhibit values exceeding the acceptable range for children in CR.

chemicals and neoplasms developement the medico-legal physician uses the classical methodology and medico- legal criteriology.

Table 2
Non-Carcinogenic and Carcinogenic Risk for adult, children and infant conducted on detected OPPs pesticides in groundwater of Campania Region.

Compounds	Non-Carcinogenic risk			Carcinogenic risk		
	Adult	Children	Infant	Adult	Children	Infant
Diazinon	3.27×10^{-3}	1.95×10^{-2}	2.92×10^{-2}	3.12×10^{-5}	2.81×10^{-4}	1.40×10^{-6}
Dimethoate	9.92×10^{-3}	3.42×10^{-1}	6.84×10^{-2}	6.55×10^{-4}	5.89×10^{-3}	2.95×10^{-5}
Malathion	6.88×10^{-5}	2.06×10^{-4}	4.13×10^{-5}	4.13×10^{-7}	3.72×10^{-6}	1.86×10^{-8}
Dichlorvos	3.27×10^{-3}	9.80×10^{-3}	1.47×10^{-2}	4.74×10^{-4}	4.26×10^{-3}	2.13×10^{-5}
Pirimiphos-methyl	1.40×10^{-4}	4.20×10^{-4}	8.41×10^{-5}	n.d	n.d	n.d
Fenitrothion	1.25×10^{-3}	3.74×10^{-2}	5.60×10^{-2}	n.d	n.d	n.d
Methidathion	n.d	n.d	n.d	n.d	n.d	n.d
Tolclofos-methyl	n.d	n.d	n.d	n.d	n.d	n.d
Parathion	2.90×10^{-4}	8.69×10^{-4}	1.30×10^{-3}	1.01×10^{-4}	9.08×10^{-4}	4.54×10^{-6}
Clorpirifos	4.69×10^{-3}	1.41×10^{-2}	2.81×10^{-3}	n.d	n.d	n.d

3.3. statistical analysis and spatial distribution

Table S4 presents the median and interquartile range for each pesticide in the analyzed provinces (AV, BN, CE, NA, SA), revealing significant differences in pesticide concentrations. Chlorpyrifos stands out for its consistently high values across all provinces, indicating widespread and generalized use. In contrast, pesticides such as Dimethoate and Tolclofos-methyl exhibit greater interquartile variability, suggesting a more targeted and localized application. These differences highlight the necessity of further analysis to uncover patterns in pesticide distribution and usage across different territories.

The bar plot in Fig. S1 shows the percentage of samples in which each pesticide was quantified in each province. Chlorpyrifos is the most widely detected pesticide, with quantification rates exceeding 90 % in all provinces, reflecting its broad agricultural use. Dimethoate shows particularly high values in Naples and Salerno, exceeding 50 %, which suggests its targeted application for specific crops. Tolclofos-methyl, although generally less frequent, displays a relatively high presence in Salerno (36.2 %), indicating a more selective local use. Fenitrothion is more evenly distributed among the provinces, with peaks in Naples and Benevento, suggesting differentiated pest management strategies. These variations in pesticide quantification imply regional differences in agricultural practices, likely influenced by predominant crops and local phytosanitary policies.

A principal component analysis (PCA) was conducted to synthesize the complexity of the pesticide dataset and identify underlying patterns.

The loadings of the pesticides on the principal components are reported in Table S5, showing how each pesticide contributes to the extracted components. The first three principal components account for 69.8 % of the total variance, effectively summarizing the dataset's structure (Fig. S2). The geographic maps in Fig. 4, representing the mean component scores for each province, provide insights into the spatial distribution of pesticide combinations.

In Table S4 the first principal component (PC1), explaining 44.7 % of the variance, is primarily driven by pesticides such as Chlorpyrifos (−0.631), Diazinon (−0.464), and Malathion (−0.454). Provinces with negative PC1 scores tend to have high concentrations of these pesticides, whereas positive scores indicate lower presence. Geographically,

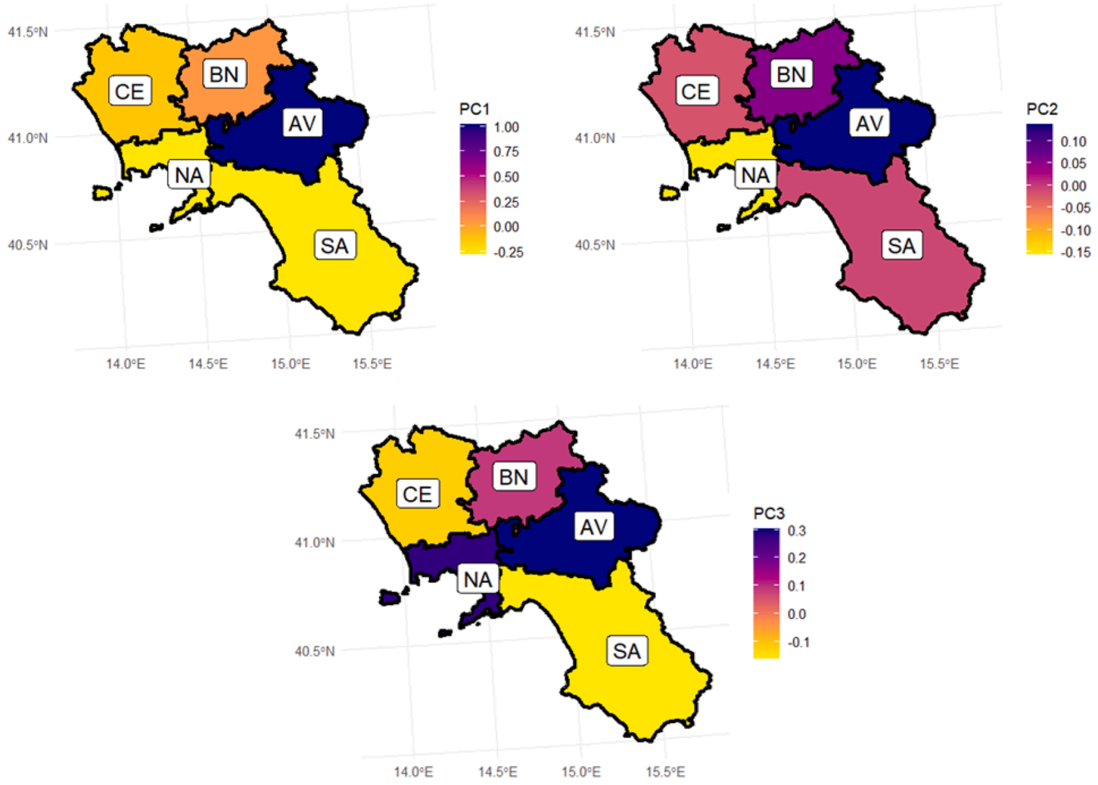

Fig. 4. Geographic distribution of the first three principal components (PC1, PC2, PC3) across provinces. PC1 differentiates regions based on the concentration of widely used pesticides like Chlorpyrifos and Diazinon, while PC2 captures variability in Dimethoate and Parathion distribution. PC3 highlights the localized presence of Tolclofos-methyl in Naples and the prevalence of Dichlorvos and Chlorpyrifos in Salerno and Caserta. AV=Avellino, BN=Benevento, CE=Caserta, NA=Napoli, SA=Salerno.

Salerno and Caserta exhibit strongly negative values, suggesting intensive pesticide use, likely associated with large-scale agricultural production. Conversely, Avellino has a positive score, indicating a lower concentration of these pesticides, possibly due to different agricultural practices or less pesticide-dependent crop management.

The second principal component (PC2), accounting for 13. 5 % of the variance, is mainly influenced by Dimethoate (0.594) and Parathion (0.670). This component differentiates provinces with high concentrations of these pesticides from those where they are less present. Benevento shows a high positive score in PC2, indicating a substantial use of Dimethoate and Parathion, which may be related to specific crop protection strategies. In contrast, Naples and Avellino have negative scores, suggesting a lower presence of these pesticides, potentially reflecting a different crop composition or alternative pest management approach.

The third principal component (PC3), which explains 11. 6 % of the variance, distinguishes provinces based on the presence of Dichlorvos (−0.671) and Chlorpyrifos (−0.632), both with strong negative loadings, against Tolclofos-methyl (0.372), which has a positive loading. Provinces with high PC3 scores, such as Naples, exhibit a greater presence of Tolclofos-methyl, suggesting a selective use, possibly for addressing specific phytosanitary challenges. In contrast, Salerno and Caserta show negative PC3 scores, indicating a higher concentration of Dichlorvos and Chlorpyrifos, pesticides commonly associated with large-scale agricultural applications.

The geographic maps in Fig. 4 confirm the spatial distribution of these components. The PC1 map indicates that Salerno and Caserta are characterized by high levels of widely used pesticides such as Chlorpyrifos and Diazinon, while Avellino stands out with lower levels. The PC2 map highlights Benevento as the province with the highest score, suggesting an increased use of Dimethoate and Parathion, whereas Naples and Avellino show lower values. Finally, the PC3 map reveals a significant concentration of Tolclofos-methyl in Naples, while Salerno and Caserta are dominated by Dichlorvos and Chlorpyrifos.

4. Conclusions

This study provides a comprehensive evaluation of OPPs contamination in groundwater across the Campania Plain, a region with intense agricultural and industrial activities. The highest concentrations were found in Caserta, Naples, and Salerno, with dimethoate, chlorpyrifos, and parathion as the most prevalent pollutants due to their current and historical use and mobility in aquifers.

The health risk assessment shows that while non-carcinogenic risks remain within safe limits, carcinogenic risks from dichlorvos and dimethoate exceed safety thresholds for children, posing long-term health concerns.

Statistical analyses (PCA) revealed correlations between pesticide distribution and land use, highlighting agricultural intensity as a key factor. The link between surface and groundwater contamination further underscores the need for integrated management strategies.

These findings call for enhanced monitoring, stricter regulations, and sustainable agricultural practices. Future research should analyze contamination trends, explore remediation strategies, and consider other emerging contaminants to improve groundwater quality assessment.

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CRediT authorship contribution statement

Elvira De Rosa: Writing – original draft, Formal analysis. **Pellegrino Cerino:** Resources, Project administration, Funding acquisition. **Maria Triassi:** Visualization, Supervision. **Fabiana Di Duca:** Formal analysis, Data curation. **Annamaria Porreca:** Methodology, Formal analysis. **Immacolata Russo:** Formal analysis. **Stefano Scippa:** Formal analysis. **Alessandro Venuta:** Formal analysis. **Annachiara Coppola:** Formal analysis. **Antonio Pizzolante:** Formal analysis. **Federico Nicodemo:** Investigation. **Ugo Trama:** Supervision. **Fabio Policino:** Formal analysis. **Paolo Montuori:** Validation, Supervision.

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.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.hazadv.2025.100701.

Data availability

Data will be made available on request.

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