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The status of organochlorine pesticide contamination in the soils of the Campanian Plain, southern Italy, and correlations with soil properties and cancer risk☆

Chengkai Qu [a, b], Stefano Albanese [a], Wei Chen [b, c], Annamaria Lima [a], Angela L. Doherty [a], Alessandro Piccolo [d], Michele Arienzo [a], Shihua Qi [b, *], Benedetto De Vivo [a]

[a] Dipartimento di Scienze della Terra, dell'Ambiente e delle Risorse, Università di Napoli "Federico II", Via Mezzocannone 8, 80134 Napoli, Italy
[b] State Key Laboratory of Biogeology and Environmental Geology, School of Environmental Studies, China University of Geosciences, 430074 Wuhan, China
[c] Lancaster Environment Centre, Lancaster University, Lancaster, Lancashire LA1 4YQ, UK
[d] Dipartimento di Agraria, Università di Napoli "Federico II", Via Università 100, 80055 Portici, Italy

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ABSTRACT

The distribution, inventory, and potential risk of organochlorine pesticides (OCPs), including Hexachlorocyclohexanes (HCHs) and Dichlorodiphenyltrichloroethanes (DDTs), and their correlation with soil properties and anthropogenic factors were investigated in soils of the Campanian Plain. The total concentrations of HCHs and DDTs ranged from 0.03 to 17.3 ng/g (geometric mean: GM = 0.05 ng/g), and 0.08 −1231 ng/g (GM = 14.4 ng/g), respectively. In general, the concentration of OCPs in farmland and orchards was higher than on land used for non-agricultural purposes. There are significant differences in the concentration of OCPs in the soils across the region, more specifically, the Acerra-Marigliano conurbation (AMC) and Sarno River Basin (SRB) are recognized as severely OCP-contaminated areas. The recent application of technical HCHs and DDTs in large quantities appears unlikely in light of the ratio of α-HCH/β-HCH and *p,p′*-DDT/*p,p′*-DDE, and the prohibition of the use of these chemicals in Italy nearly forty years ago. The clear correlation between the concentration of DDTs and organic carbon suggests a typical secondary distribution pattern. The mass inventory of OCPs in soils of the Campanian Plain is estimated to have a GM of 17.3 metric tons. There is no clear evidence linking the impact of geographical distribution of OCPs on the incidence of cancer, and the 95% confidence interval of total incremental lifetime cancer risk (TILCR) data falls below the internationally accepted benchmark value of 1×10^{-5}.

1. Introduction

While the prolific use of organochlorine pesticides (OCPs) over the last few decades has greatly decreased the incidence of agricultural and vector-borne diseases, it has raised new concerns regarding the potential dangers of OCPs to human health and the environment (Yadav et al., 2015). Many treaties and laws have been enacted to eliminate or restrict the production and use of some OCPs, of which the Stockholm Convention on Persistent Organic Pollution (POPs) is one of the most famous. Two of the most representative OCPs are Dichlorodiphenyltrichloroethanes (DDTs) and Hexachlorocyclohexanes (HCHs), which were used extensively worldwide from the 1950s to the 1980s as agricultural and domestic pesticides (Tieyu et al., 2005). In the latter half of the 20th century, the total global production of HCHs and DDTs reached at 2 and 3 million metric tons, respectively, and the consumption rate of these both pesticides in Mediterranean countries reached approximately 2000 t/yr (Bacci, 1993 and references therein).

OCPs generally break down in the environment over time, however, as they are persistent and have an affinity for organic matter, a considerable quantity of these chemicals may remain stored in soils (Zhang et al., 2012). These OCPs tend to accumulate in organisms and are biomagnified through the food chain (Naso et al., 2003). The soil then becomes a reemission source as the OCPs are released into the atmosphere (Tao et al., 2008). A large

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* Corresponding author.
E-mail address: shihuaqi@cug.edu.cn (S. Qi).

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quantity of OCPs in agricultural soils can be transported into the surface water by farm drainage and surface runoff, and into groundwater via farmland sewage irrigation (Montuori et al., 2014). The environmental fate of OCPs in soils is affected by many factors, of which, the physicochemical characteristics of agrochemicals (e.g., vapor pressure, water solubility), land use type, application history and agricultural practices (e.g., flood irrigation, superphosphate application, and tilling), soil properties (e.g., organic carbon [OC], pH, texture, and moisture content), as well as meteorological variables (e.g., temperature, wind direction), are, perhaps, the most important (Yu et al., 2013; Zhang et al., 2011). As a result, soil plays an important role in the circulation of these substances in different sectors of the environment, but the behavior and fate of OCPs in soils on a regional scale is a complicated process.

Many recent studies have produced ecological risk assessments in soils contaminated with OCPs (e.g., Yu et al., 2013; Zhang et al., 2009), however, literature regarding the carcinogenic risk of human exposure to OCPs is limited. While the risk to human health from exposure to soil-borne OCPs has recently been evaluated by calculating the incremental lifetime cancer risk (Ge et al., 2013; Hu et al., 2014; Qu et al., 2015), in general, the research into environmental epidemiology of OCPs, determining possible links between the incidence of specific diseases (including various forms of cancer) and the distribution patterns of OCPs, is scarce. For examples, Albanese et al. (2013) observed a clear spatial correlation between the incidence of some types of cancer and the distribution of certain toxic metals in stream sediments, while other research has demonstrated a close relationship between human cancers of the pancreas, breast, uterus, and liver with OCPs (Botella et al., 2004). Studying the impact of geographical distribution and ultimate fate of OCPs on the incidence of cancer is especially critical to the potential mitigation and/or remediation of the contaminated area.

So far, the Stockholm Convention has not been ratified by Italy, however, several other regulatory schemes, such as a European Directive in 2000, the UNECE POPs Protocol, and the Rotterdam Convention, are actively followed (Estellano et al., 2012). Italy was one of the main consumers of DDTs in late 20th century Europe, with consumption of DDTs reaching 2178, 1570, 197, 103, and 55 t/yr in 1970, 1975, 1980, 1985, and 1990, respectively (Pacyna et al., 2003). The Campania region is a very important area for Italian agriculture where fruits and vegetables are the predominant agricultural product, and uses a greater quantity of pesticides (28.4 kg/ha in 1978) relative to other Italian regions (Berardi, 1983). Recent studies have reported the levels of OCP contamination in the water, sediment and organisms of the Campania region (e.g., Ferrante et al., 2010; Montuori et al., 2014; Naso et al., 2005), however, a regional study of the soil contamination is still lacking. The Campania region falls within the subtropical zone and enjoys a Mediterranean climate. High temperatures in tropical/subtropical regions can facilitate the volatilization and escape of OCPs from soils, sediments, and water (Yadav et al., 2015). However, the lack of sufficient data on the residual concentration of OCP in soils, limits the understanding of the health effects, environmental dynamics, and ultimate fate of these chemicals.

This paper presents the results of an investigation using a systematic grid sampling method and geostatistics to illustrate spatial variations in the concentrations of OCPs in the soils of the Campanian Plain. The main objectives of this study were: (I) to investigate the residual levels, distribution, and possible sources of OCPs, and their subsequent mass inventories in soils of the Campanian Plain, (II) to analyze the impact of soil properties on contaminant distribution, and (III) to evaluate the potential ecological and health risks of OCPs and preliminarily explore whether there is a significant correlation between epidemiological data and carcinogenic risk of OCP residues.

2. Study area

The study area covers approximately 2400 km^2, and roughly corresponds to the Campanian Plain, defined as a wide coastal belt that extends from the Volturno River Plain to the Sarno River Basin (Fig. 1A). Large urban settlements are located within the study area. These are densely populated and industrialized, and adjacent to extensive areas dedicated to agriculture. Recently, this area has become infamous for the illegal disposal of toxic/radioactive waste by local criminal organizations (Senior and Mazza, 2004). The study area consists of three idealized territorial units based on land use (Fig. 1A) and morphology (Fig. 1B): (1) the Domizio-Flegreo Littoral (DFL) and Agro Aversano (AA) areas, (2) the metropolitan area of Naples (NAP), and (3) the Sarno River Basin (SRB) and Sorrento Peninsula (SP).

2.1. Domizio-Flegreo Littoral, Agro Aversano and Acerra-Marigliano conurbation

The Domitizio-Flegreo Littoral (DFL) and Agro Aversano (AA) areas (including the Volturno River Plain, the Regi Lagni Basin, and the Phlegrean volcanic field) have been declared as Sites of National Interest by the Italian Government due to their high potential for contamination. Chemical industries, intensive agriculture and buffalo farms are the main productive activities in the Volturno River Plain and the Regi Lagni Basin, and are also considered the main sources of heavy metal and organic pollution in both stream waters and soils (Bove et al., 2011; Grezzi et al., 2011). Across the DFL and AA areas, illegal waste dumping and uncontrolled burning of agricultural and industrial wastes continue to affect the overall environmental equilibrium.

Within the Agro Aversano territory, the Acerra-Marigliano conurbation (including the municipalities of Acerra, Pomigliano d'Arco, Castelcisterna, Marigliano, Mariglianella and Nola, collectively known as the AMC) was once an important agricultural heartland, and remains the home of important chemical (Montefibre) and automotive (FIAT) industries. In the northern sector of the Acerra municipal territory, an urban waste incinerator started activity in 2009, burning, at the start of its operation, undifferentiated waste from the metropolitan area of Naples. Additionally, the AMC is known as part of the infamous "Triangle of Death", the region where Senior and Mazza (2004) found an abnormal incidence of cancer in the local population, and postulated a link to the widespread illegal dumping (and burning) of waste.

The AMC also belongs to a huge territorial meta–entity, comprising 57 municipalities, and approximately two and half million inhabitants, renamed by Italian media as the "Land of Fires", due to the almost daily unauthorized burning of industrial waste of unknown origin across these territories.

2.2. Naples metropolitan area

Naples, the third-largest municipality in Italy with a high density of both population and industry, faces a serious challenge in mitigating the increasing environmental contaminants caused by persistent toxic substances (Albanese et al., 2015a). The metropolitan area of Naples (NAP), extending from the Phlegrean Fields in the west, to the Mt. Vesuvius volcanic complex in the east, is surrounded on three sides by agriculture (Fig. 1A).

2.3. Sarno River Basin and Sorrento Peninsula

The Sarno River Basin (SRB), comprising the alluvial plain of the Sarno River and the valleys of the Solofrana and Cavaiola

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