Small Ruminant Research 106S (2012) S49–S53

Contents lists available at SciVerse ScienceDirect

Small Ruminant Research

journal homepage: www.elsevier.com/locate/smallrumres

Sheep milk as a potential indicator of environmental exposure to dioxin-like polychlorinated biphenyls (dl-PCBs)☆

E.G. Herrera Nuñez[a], M. Perugini[a,*], M. Esposito[b], L. Baldi[c], M. Amorena[a]

[a] Dipartimento di Scienze degli Alimenti, University of Teramo, Via Crispi 212, 64100 Teramo, Italy
[b] IZS del Mezzogiorno, via Salute 2, 80055 Portici (Na), Italy
[c] ORSA Osservatorio Regionale Sicurezza Alimentare, via Salute 2, 80055 Portici (Na), Italy

ARTICLE INFO

Keywords:
Dioxin-like PCBs
Sheep milk
GIS

ABSTRACT

Dioxin-like polychlorinated biphenyls are lipophilic persistent organic pollutants which accumulate in the fat tissue of animals. Moreover, in mammals, these contaminants are secreted into milk during lactation periods. The contamination level of milk is closely related to the animal dl-PCBs exposure through diet, inhalation and dermal contact. The study of the relationship between the environment in which the animals live and the milk contamination level is a key component in assessing the presence of pollutants in dairy products. In this paper, soil was selected as a reliable matrix of environmental pollution with dl-PCBs as it reflects as well the possible contamination of vegetation and air in the areas where sheep graze and live. However, dl-PCBs analysis could be problematic due to the high cost. To overcome this problem, the approach of this study was focused on using GIS and geostatistics to integrate data from different research institutions, aiming to assess the relationship between milk and soil contaminated with dl-PCBs. Therefore, samples of soil and sheep milk collected in Campania (Italy) during 2008 monitoring plans were introduced in a GIS. The relationship between the two variables showed heteroscedasticity, and the global regression technique OLS indicated a R^2 of 0.24. To improve the OLS results, the altitude was integrated in the GIS considering that this variable can influence PCBs contamination. The results showed that altitude is strongly related with soil contamination but was not strongly related with milk contamination. Thus, analyzing the relationship between milk and soil contamination dividing data in three different groups of altitude ranges, the R^2 values were higher. Furthermore, this correlation changes with the altitude, becoming weaker with higher altitude. These results show the possibility to use sheep milk as indicator of PCBs contamination at least from sheep bred under 300 m of altitude.

1. Introduction

The dioxin-like PCBs (dl-PCBs) are a group of 12 polychlorinated biphenyls, showing chemical and toxicological properties similar to those of dioxins. They are persistent organic pollutants (POPs) (Colles et al., 2008; Liu and Liu, 2009) and, due to their carcinogenic and mutagenic effects and their endocrine disrupting action (Salihoglu and Tasdemir, 2009; Vives et al., 2008) represent a threat to human health. Since they are widespread environmental contaminants, they are found in environmental samples, in food for human consumption, in animal feed and in grassland (De Mul et al., 2008). In the environment, these compounds decompose very slowly and due to their liposolubility they accumulate in the food chain and

☆ This paper is part of a supplement titled "SIPAOC Congress 2010" guest edited by Antonello Carta, Maria Teresa Manfredi, Giuseppe Moniello and Massimo Trabalza-Marinucci.

* Corresponding author. Tel.: +39 0861 266988; fax: +39 0861266989.
E-mail address: mperugini@unite.it (M. Perugini).

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http://dx.doi.org/10.1016/j.smallrumres.2012.04.033

represent a serious hazard for human and animal health. Due to atmospheric deposition, these compounds accumulate in the leafy vegetation (such as grass) and in the soil, that acts as a natural sink. Once settled into soils, dl-PCBs concentration is directly related with the environmental pollution (Zhang et al., 2009; Vives et al., 2008). Grazing animals as sheep, ingest contaminated grass and these contaminants are rapidly absorbed from the gastrointestinal tract (Faye and Sinyavskiy, 2008; Esposito et al., 2010). Dl-PCBs once absorbed by animals, due to their lipophilic nature, accumulate in the liver and adipose tissues and during lactation periods, milk represent the main excretion pathway for these contaminants (Schmid et al., 2003; Esposito et al., 2009).

Campania (Italy) is strongly inclined toward agriculture, and it is a region where sheep extensive breeding is largely used. Until the 1990s the "shepherd system" was the most important way of breeding: it was based on pastures located in places far from each other; now the situation has changed, with the "permanent" breeding. Sheep introduce dl-PCBs both eating forage which has been contaminated by atmospheric deposition and ingesting direct contaminated soil from pasture. In assessing the impact of soil contamination on plants, it appears that for PCBs there is an increasing concentration with decreasing soil particle size (Smith and Jones, 2000). Considering that the sheep breeding system is permanently in the open air, milk could be an ideal matrix for monitoring the PCBs presence in the environment providing exposure information through a non-invasive collection method. Since 2007, the Campania Region Health Authority launched a dioxin surveillance plan in order to control the pollution degree and examine contamination extent and sources. This plan was focused on milk (sheep, cow and buffalo) samples collection to determine the PCDD/F and dl-PCB levels. At the same time the Agenzia Regionale Protezione Ambientale della Campania (ARPAC) monitored the levels of PCBs-dioxins in the soil.

In this context, Geographic Information Systems (GIS) rise as a powerful tool for data analysis. GIS is a computer-based technology and methodology for collecting, managing, analyzing, modeling and presenting geographic data for a wide range of applications (Davis, 2001). Features on maps have spatial relationships which can be very important in many applications, and GIS is an excellent tool for determining such characteristics (Davis, 2001). One of the chief virtues of GIS is that it allows the visualization of spatial data as well as providing a means of utilizing fuzzy data. While the quantitative science prefers clear and precise facts, GIS provides a way to include data that is not so pristine (Schuurman, 2004). GIS offer a variety of geo-processing features, all dealing with the management of data (e.g. dissolving, clipping, merging and intersecting) allowing to aggregate information from one layer to another. Transformation, conversion or the translation of data from one unit or level to another is the next step of coupling. Most important for the process of integration is the interpolation of single data-points to area-wide information. Interpolation creates a surface, a regionalization, from point-data by geostatistical operations (Jopp et al., 2011).

Table 1
dl-PCBs concentrations in sheep milk and soil samples (WHO-TEQ-ng/kg).

Number of observations	dl-PCBs concentrations in milk WHO-TEQ ng/kg	dl-PCBs concentrations in soil WHO-TEQ ng/kg
1	0.84	0.03
2	1.885	0.26
3	0.567	0.29
4	0.71	0.08
5	0.66	0.03
6	1.163	0.38
7	0.582	0.17
8	0.605	0.25
9	0.934	0.21
10	0.08	0.14
11	0.599	0.07
12	0.41	0.22
13	0.623	0.09
14	1.4	0.12
15	2.65	0.31
16	0.859	0.10
17	0.597	0.08
18	0.578	0.16
19	6.57	0.45
20	0.58	0.05
21	1.31	0.43
22	0.768	0.37
23	4.8	0.38
24	4.74	0.37
25	1.21	0.14
26	4.75	0.26

This paper aims to study the relationship between sheep's milk and soil contaminated with dl-PCBs as a first stage of further studies to assess a valid bioindicator of environmental pollution. Data from different research institutes were integrated in a GIS as it allows to manage different information layers and perform spatial analyses.

2. Materials and methods

Sheep milk samples were collected from 26 farm located in Campania. The sheep breeding was extensive livestock farming and no feed were given to the animals.

Total dl-PCBs concentration in sheep's milk samples is expressed as the sum of 12 congeners in WHO-TEQ pg/g fat in agreement with the Regulation EC 1881/2006 (Table 1). All data derive from the EU monitoring plan, Residues National Plan (PNR) and Surveillance Regional Plan for PCBs and dioxins carried out in 2008 by Campania Region. These data were obtained in collaboration with the Istituto Zooprofilattico Sperimentale del Mezzogiorno-Portici, Italy, where dl-PCBs analyses in sheep's milk were carried out, according to the EPA method 1613 (USEPA, 1994).

Total dl-PCB concentrations in soil samples are expressed as the sum of 12 congeners (WHO-TEQ ng/kg) and all data derived from the ARPA Campania web site (http://www.arpacampania.itat_cont_area2.asp?id=10).

2.1. Geostatistical analyses

Data was analyzed using ESRI ArcGis software (Environmental Systems Research Institute, Redlands, CA) version 9.3. The Ordinary Kriging method was used to interpolate soil contamination values. The parameters of lag size, number of neighbors and shape of semivariogram were combined to obtain the best result. The different maps obtained were compared using cross-validation and the best combination was the one with the less root mean square error. The interpolation of soil contamination values was performed to overcome the problem of different sampling locations between the variables (milk and soil), obtaining a continuous map. This permitted to estimate soil contamination values for each point where milk was sampled. The relationship between soil and milk contaminated with dl-PCBs was analyzed using Ordinary Least Squares (OLS). Furthermore, Koenker and Global Moran's Index statistical tests were applied. The Koenker test was performed to determine