

DOI 10.7764/rcia.v45i2.1887

RESEARCH PAPER

Impact of environmental variables on PCDD/F and dl-PCB levels in dairy milk of the farming region of Chile

Nicolás Pizarro-Aránguiz¹, Diego García-Mendoza², Rubén Muñoz³, Betty San Martín³, and Rodrigo Morales¹

¹Instituto de Investigaciones Agropecuarias, INIA Remehue. Ruta 5 Norte km 8. P.O.Box 24-0, Osorno, Chile.

²Wageningen University, Division of Toxicology. 6708 PB Wageningen, Netherlands

³Universidad de Chile, Faculty of Veterinary Science, Laboratory of Veterinary Pharmacology. Av. Sta. Rosa 11735, 8820808 Santiago, Chile

Abstract

N. Pizarro-Aránguiz, D. García-Mendoza, R. Muñoz, B. San Martín, and R. Morales. Impact of environmental variables on PCDD/F and dl-PCB levels in dairy milk of the farming region of Chile. 2018. Cien. Inv. Agr. 45(2): 109-119. According to a One Health perspective, the importance of persistent organic pollutants (POPs) must be assessed because of their impact on the environment, food chain and human health. However, information on these toxic compounds is limited in Latin America and the Caribbean region. Chile is no exception; therefore, this work aimed to explain previously reported dioxin levels in cow-milk samples by utilizing regression with meteorological/geographical data that were collected over a three-year survey. To accomplish this aim, a stepwise general multiple regression analysis was carried out for polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (dl-PCBs). The best statistical adjustments were achieved only for highly present congeners. Regarding PCDD/F congeners, the most relevant and significant ($P < 0.05$) factors were the year (mostly a negative coefficient), the season, and the hectares affected by forest fires. In the case of dl-PCB congeners, there was a clear, positive relationship with the geographic parameter (UTM), and this result was consistent with previous findings that dl-PCB congeners show a trend with latitude. In contrast, wind speed was a significant negative coefficient for dl-PCBs. Despite existing knowledge on pollutant levels in milk, this study is relevant to better understand these findings in the Latin America and Caribbean regions.

Keywords: Dioxin-like polychlorinated biphenyls, environmental variables, food contamination, persistent organic pollutants, polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans.

Introduction

Polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and

dioxin-like polychlorinated biphenyls (dl-PCBs) are considered to be persistent organic pollutants (POPs) due to long-range atmospheric transport, environmental persistence, and a capacity for bioaccumulation in animal fat tissue (Abad *et al.*, 2002). PCDD/Fs are chemical by-products of several industrial and combustion processes. While

large quantities of dl-PCBs were produced over the last century for industrial uses, production of these chemicals have been recently banned. These pollutants are toxic even in small concentrations and are considered carcinogenic compounds (Erickson *et al.*, 2011). Public programs for measuring environmental pollutants, as established under the Stockholm Convention, are particularly concerned about food safety, since consumption of contaminated food is the major pathway of exposure for humans. These programs aim to prevent high pollutant levels in animal feed and animal origin products for human consumption (Kotz, 2014). Many previous studies have assessed dioxin and dioxin-like compound levels in animal products for human consumption, feed, and feed additives (Esposito *et al.*, 2009, Pizarro-Aranguiz *et al.*, 2015, Pemberthy *et al.*, 2016, San Martín *et al.*, 2016) could be differential pollutant levels among animal products, including cow milk and dairy products, that are related to human exposure and represent a considerable proportion of total dietary exposure.

For instance, dairy and beef cattle researchers have shown that POPs initially enter the food chain by atmospheric deposition from local emission sources that reach soils and pasture, which are in turn consumed by livestock (Rychen *et al.*, 2008). The other alternative is by ingestion of contaminated feed supplements (i.e., risk source) and the production cycle (Lake *et al.*, 2013, Shunthirasingham *et al.*, 2013). Bioaccumulation in animal tissue from feed sources depends on the congener patterns and matrix of PCDD/Fs and dl-PCBs; in fact, there is a wide range of transfer factors for each congener (Kotz, 2014). Additionally, environmental conditions at animal farms, such as meteorological and geographical factors, can cause variations in atmospheric deposition and thus bioaccumulation in milk (Schulz *et al.*, 2005, Shunthirasingham *et al.*, 2013). It is well known that PCDD/Fs and dl-PCBs are present in the atmosphere in the form of gas and particles, and the partition process is governed by the equilibrium between these two phases, which depends on the

various environmental factors that affect the final deposition, such as air temperature or vapor pressure (Aristizábal *et al.*, 2011, Shunthirasingham *et al.*, 2013). Furthermore, a correlation has been recognized between air quality and meteorological variables, which directly affects the accumulation and diffusion of pollution (Halfon *et al.*, 2009).

Animal farming in Chile occurs between the latitudes of 32 and 41°S, in the central valley, ranging from the metropolitan to the Los Lagos regions, which includes the most populated cities in the country. These regions have an increasing latitudinal trend in precipitation, a decreasing trend in mean temperature and have the following seasons: summer (December to February), autumn (March to May), winter (June to August), and spring (September to November). Bovine production (dairy and beef cattle) is concentrated in this area, and at least 70% of the total production is concentrated in the southern area of the valley. The main production system used in this area corresponds to a free-range system based on pasture and strategic use of supplementary feeds during the winter and summer seasons, such as forage and concentrates. Confined production systems, which use conserved forage and concentrates (grains, agricultural subproducts, etc.), are typical in the northern area of the valley (i.e., Metropolitan and Valparaíso regions) because of the lack of space for extensive pasture areas.

Furthermore, there are other conditions of the central valley that should be investigated and warrant environmental pollution studies and include the following: (1) anthropogenic factors such as population size, particularly as more than half of Chile's population lives within the central valley, which leads to massive emissions from concentrated vehicle usage, especially in the northern area of the valley (Industries, such as cellulose and paper pulp industry, are common in the mid part of the valley. In the southern area, the wood combustion (for house heating) is also a serious environmental threat. There are other common activities for the entire central valley, e.g., refin-

eries, mining, cement kilns, waste incinerators, and energy generation); (2) natural factors, such as uncontrollable forest fires in the northern area of the valley during the summer and volcanic activity in the southern area of the valley.

There is a lack of data on the atmospheric deposition for dioxins in Chile, although some studies have revealed the presence of these pollutants in specific areas of the country (Pozo *et al.*, 2004, 2015, Loyola-Sepúlveda *et al.*, 2018).

In a previous study, Pizarro-Aránguiz *et al.* (2015) reported results of a three-year survey of these pollutants in milk samples from seven different regions in the central valley of Chile, with clear spatial differences shown between samples that originated near the most and least populated cities. Considering this prior evidence and despite scarce atmospheric and animal food-feed data concerning dioxin levels within the cow-milk cycle in Chile, the aim of this work is to assess possible relationships between raw cow milk pollutant levels, specifically the seventeen polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) congeners, and twelve dioxin-like polychlorinated biphenyls congeners (dl-PCBs) with various meteorological and geographical factors within Chile's central valley regions.

Materials and methods

PCDD/Fs and dl-PCBs data

Previously reported data, which are summarized in Table 1 and Supplementary Tables S1 and S2, were used in the present analyses (Pizarro-Aránguiz *et al.*, 2015). In summary, milk samples were taken between August 2011 and December 2013 for the national monitoring program that was carried out by the Agricultural and Livestock Service of Chile (SAG). The numbers of samples for each year were 32 samples for 2011, 37 samples for 2012 and 33 samples for 2013 and were obtained from various

producers in the seven different regions of Chile's central valley (32–41°S, from Metropolitan to the Los Lagos Region, See Figure 1A). Samples were liquid-liquid extracted, purified with a multilayer silica gel column, and fractionated with a carbon column. Finally, the extract was injected for High-Resolution Gas Chromatography/High-Resolution Mass Spectrometry Analysis. Full methodology and quality assurance protocols have been reported (Pizarro-Aránguiz *et al.*, 2015).

Table 1. Mean concentrations of PCDD/F and dl-PCB residues (standard deviations) on raw cow milk samples of each of the regions and years studied. Regional symbols: V (Valparaíso); RM (Metropolitana); VII (Del Maule); VIII (Del BIOBIO); IX (De la Araucanía); X (De los Lagos); XIV (De los Ríos).

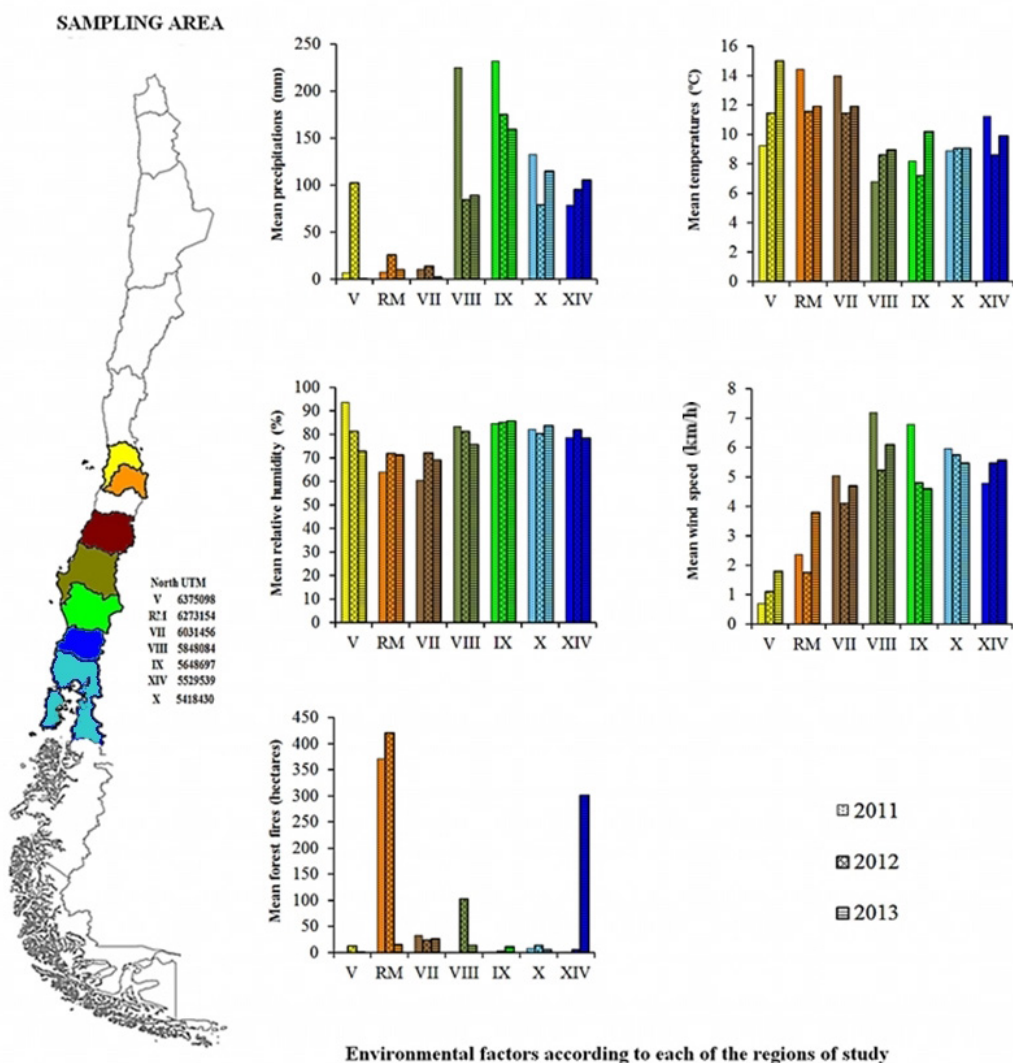
Region	Year	PCDD/Fs (pg g ⁻¹ fat) Mean (SD)	dl-PCBs (pg g ⁻¹ fat) Mean (SD)
V	2011	4.580*	139.580*
	2012	4.10 (2.2)	174.01 (2.55)
	2013	0.720*	168.410*
RM	2011	3.49 (1.49)	315.38 (223.45)
	2012	2.66 (1.97)	185.52 (57.97)
	2013	0.92 (0.62)	85.19 (14.77)
VII	2011	2.73 (1.49)	984.23 (24.42)
	2012	1.94 (0.84)	81.72 (10.34)
	2013	0.450*	67.070*
VIII	2011	2.160*	23.880*
	2012	1.35 (0.57)	81.72 (38.26)
	2013	0.900*	133.320*
IX	2011	2.80 (0.41)	38.38 (4.87)
	2012	1.60 (0.14)	55.16(20.37)
	2013	0.76 (0.44)	48.77 (12.1)
X	2011	2.26 (0.97)	38.42 (15.69)
	2012	1.30 (0.61)	35.89 (10.92)
	2013	0.41 (0.95)	62.00 (30.03)
XIV	2011	5.71 (4.04)	110.91 (84.54)
	2012	2.60 (2.11)	48.59 (23.05)
	2013	0.70 (0.27)	66.11 (25.09)

*Means that is only one sample for that period. Reference (Pizarro-Aránguiz *et al.*, 2015)

Meteorological and geographical data

According to the Köppen system, the central valley of Chile is represented by two different climate regimes: a Mediterranean climate (dry-wet) in the northern area of the valley and a temperate oceanic climate in the southern area (Luebert and Pliscoff, 2006). Daily Meteorological data including temperature (°C), precipitation (mm), relative humidity (%), wind velocity (km h⁻¹) and pressure (mmHg) were obtained from the online AGROMET database of the Chilean Institute

of Agricultural and Livestock Research (INIA, n.d.), which has a network of meteorological stations throughout Chile that are located near milk production areas. Mean values for each of the parameters were calculated for the previous and current month according to the sample period date (for each milk sample) with the understanding that the levels of POPs in animal tissue result from a bioaccumulation process and that meteorological factors can have a delayed influence on those levels. Historical data on hectares affected by wildfires were obtained from the National



Environmental factors according to each of the regions of study

Figure 1. Graphical representation of mean values of the environmental variables recorded for each of the regions assessed in the three-year survey

Forestry Corporation (CONAF, n.d.). Sampling locations were recorded using the Universal Transverse Mercator (UTM) coordinate system. A graphical representation with the mean values of these variables is presented in Figure 1, and more details are provided in Table S3 of the supplementary material. Additionally, the year, season (hot or cold) and regional origin of the sample were included as categorical predictors.

Statistical Analysis

The relationship between the meteorological and geographical data with the pollutant concentration in milk samples was assessed by performing a stepwise linear multiple regression analysis as follows:

$$\gamma = \alpha + \beta_1 \cdot \chi_1 + \beta_2 \cdot \chi_2 + \dots + \beta_n \cdot \chi_n + \mu \quad (1)$$

where γ is the mean value of the pollutant concentration in milk as a dependent variable; α is the regression intercept; $\beta_1 \dots \beta_n$ are the regression coefficients for the independent variables; $X_1 \dots X_n$ are the value of each independent variable (year, region, season, temperature, precipitation, wind speed, relative humidity, hectares of forest fires and latitude), and μ is the residual. The data of each pollutant concentration from the samples were paired with meteorological and geographical data on a Microsoft Excel sheet data matrix (according to the date of the sampling period). All analyses were conducted using the software MINITAB (Minitab Inc., Pennsylvania, USA). An explanatory data analysis was carried out, and the normal distribution was evaluated, while a log transformation was carried out where dioxin data were not normally distributed. Additionally, all congener compounds that were detected in less than 40% of the samples were excluded from the analysis; in the case of PCDD/Fs, the eliminated congeners were as follows: 123789-HxCDF, 1234789-HpCDF, 2378-TCDD and 123478-HxCDD. For dl-PCBs, the only congener with less than 80% of detection was PCB-81, and thus it was excluded from the

analysis. For detailed concentrations of PCDD/Fs and dl-PCBs congeners see Tables S1 and S2.

Results & discussion

The current study describes the relationships between PCDD/Fs and dl-PCBs congeners levels in raw milk samples from seven different regions of the central valley of Chile with various climate and geographic factors during the 2011-2013 sample period. Overall, stepwise linear multiple regression models showed a better statistical fit with those congeners having the highest contributions to the total sum expressed toxic equivalency (TEQ) values (pg g^{-1} fat). In summary, the variability percentage explained by the model for each of the congeners with a better statistical fit is as follows: 23478-PeCDF (55%), 1234678-HpCDF (52%), OCDF (66.9%), OCDD (68.2%); PCB 77 (52.8%), 118 (54.3%), 105 (64.3%), 167 (66%) and 156 (67%). For more details, see Tables 2 and 3.

According to the model results, the following meteorological and geographical factors have a positive influence on POPs levels; PCDD/Fs congeners correspond to:

-Year: Year was negatively related in general, but positively related to some of the less chlorinated congeners. This negative effect of year (higher levels in 2011 than 2013) is consistent with a previous report of a decreasing temporal trend in the total sum of these pollutants (Pizarro-Aranguiz *et al.*, 2015). Interestingly, some of the most toxic congeners (less chlorinated) could be occurring in these kinds of samples, which could support the need for an environmental forensic study to trace the emissions back to the source.

-Season: The hot season was significant for all the congeners (except for 123478-HxCDF); however, the air T, as a single covariate, was did not present statistical significance in the congener models with a good statistical adjustment. This means that there are other variables present in the hot

Table 2. Results of the model. Covariates and corresponding statistical significance (p-value), coefficients, and the R2 for each of the PCDD/F congeners.

Covariates†	HA													R2
	Y	R	HS	CS	P PM	P CM	T PM	T CM	UTM	H A F PM	F	W	RH	
	P V	Y(C)	P V	P V	P V	C/ P V	C/ P V	C/ P V	C/ P V	C/ P V	C/ P V	C/ P V	C/ P V	
2378-TCDF	0.000	12(2.361)	-	0.02	-	-	-	-	-	-	-	-	-	36.49%
12378-PeCDF	0.00	12(1.45); 13(2.87)	-	0.02	-	-	-	-	-	-	-0.006/0.1	-	-	46.09%
23478-PeCDF	0.00	12(-0.3); 13(- 0.9)	-	0	-	0.001/0.08	-	-	-	-	-0.002/0.02	-	0.06/0.02	55.13%
123478-HxCDF	0.01	-	-	-	-	-	0.11/0.03	-	0.000001/0.04	-	-	-	0.06/0.01	22.30%
123678-HxCDF	0.002	13 (-0.9)	-	0.001	-	-0.005/0.0	-	-	-	-	-	-0.1/0.1	-	40.45%
234678-HxCDF	-	-	-	0.02	-	-	-	-	0.000001/0.02	-	-	-	-	21.90%
1234678-HpCDF	0.00	12(-1.1); 13(-2.9)	-	0	0.06	-	-	-	-	-	-0.007/0.02	-	-	51.31%
OCDF	0.00	12(-3.9); 13(-5.2)	-	0.01	-	0.01/0.02	-0.01/0.0	-	-	-	-	-	-	66.93%
12378-PeCDD	-	-	-	0.006	0.04	-	-	-	-	-	0.007/0.1	-	-	24.89%
123678-HxCDD	-	-	0.02	0.001	-	-	-	-	-	-	-	-	-	34.72%
123789-HxCDD	0.01	12 (2.37)	-	0.02	-	-	-	-	-	-	-	-	-	30.67%
1234678-HpCDD	0.00	13 (-0.9)	0.006	0	-	-	-	-0.18/0.06	-	-	-	-	-	46.67%
OCDD	0.00	12 (-1.9); 13 (-4.4)	-	0	-	-	-	-	-	-	0.009/0.005	-	-	68.18%

†Covariates meanings: Y (Year); R (Region); HS (Hot Season); CS (Cold Season); P PM (Precipitation previous-month); P CM (Precipitation current-month); T PM(Temperature previous-month); T CM (Temperature current-month); UTM (latitude); HAF PM (Hectares of forest affected by fires previous-month); HAF CM (Hectares of forest affected by fires current-month); W (wind); RH (Relative humidity); R² (Coefficient of determination). PV (*p-value*); C (regression coefficient) for each covariate. Only significant covariates are presented; statistically are significant in bold. –Empty space means that for that congener the covariate was not statistically significant or was eliminated from the model.

season that are related to PCDD/Fs levels in milk samples that are not covered by these models. One hypothesis is that agricultural waste burning that occurs during this season and produces deposition of these POPs in the surrounding environment, which has been confirmed by the findings of other researchers (Dwyer and Themelis, 2012). Another interesting hypothesis is that other climatic parameters could be affecting the concentration of pollutants in the hot season, such soil temperature which Tremolada *et al.* (2009) showed reflects the climatic conditions of the recent past (one or two months) and which could be a critical parameter for understanding the higher deposition rates of these POPs during this season in the mountain region. This leads to higher POPs volatilization rates from the environmental reservoir.

-Forest hectares affected by fires in the previous month: This was a significant factor that negatively affected the model for 23478-PeCDF, 1234678-HpCDF, and OCDD. This result was not expected because, as other researchers have determined (Loyola-Sepúlveda *et al.*, 2018), forest fires are a well-known source of these POPs, which suggests that the bioaccumulation process and the transfer from air to animal is not as fast as from air to soil and water.

-Precipitation: Precipitation was not a significant factor in our model; in contrast, Correa *et al.* (2006) investigated a model of ambient PCDD/Fs concentrations, and they routinely measured air pollutants and meteorological parameters with moderate success and showed that wet deposition

Table 3. Results of the model. Covariates and corresponding statistical significance (*p-value*), coefficients, and the R² for each of the dl-PCBs congeners.

Covariates [†]	Y	R	HS	CS	PPM	PCM	TPM	TCM	UTM	HAFPM	HAFCM	W	RH	R ²	
	PV	Y(C)	PV	PV	PV	C/PV	C/PV	C/PV	C/PV	C/PV	C/PV	C/PV	C/PV		
PCB 77	0.000	13(2.5)	-	-	-	-	-	0.16/ 0.03	-	-	0.005/ 0.08	-	-	52,81%	
PCB 123	-	-	-	-	-	-	-	-	0.000006/ 0.003	-	-	-	-	8,79%	
PCB 118	-	-	-	-	-	-0.002/ 0.005	-	-	-	0.000004/ 0.00	-	-	-0.09/ 0.006	54,27%	
PCB 114	-	-	-	-	-0.4/ 0.05	-	-	-	-	0.000047/0.003	-	-	-0.3 / 0.00	38,62%	
PCB 105	-	-	-	-	-0.2/ 0.07	-	-	-	-0.15 /0.02	0.00002 / 0.01	-	-	-0.1/ 0.001	-0.05/ 0.007	64,31%
PCB 126	0.01	13 (-0.6)	-	-	-	-	-	0.17/ 0.06	-	0.00005 / 0.000	-	-	-0.24 / 0.001	44,66%	
PCB 167	-	-	-	-	-0.2/ 0.06	-	-	-	-	0.00002/ 0.004	-	-0.0001/ 0.04	-0.1 / 0.001	65,57%	
PCB 156	-	-	-	-	-	-	-	-	-	0.000019/ 0.009	-	-0.0001/ 0.043	-0.11/ 0.002	66,84%	
PCB 157	0.004	13 (-0.6)	-	-	-	-0.002/ 0.06	-	0.12/ 0.03	-0.16/ 0.003	0.000006 / 0.00	-	-	-	41,44%	
PCB 169	0.01	12 (-1.3)	-	-	-	-	-	-	-	0.00006 / 0.007	-	-	-	44,01%	
PCB 189	0.002	13 (-1.6)	-	0.01	-	-	-	-	-	0.00005 /0.03	-	-	-0.29/ 0.03	41,20%	

†Covariates meanings: Y (Year); R (Region); HS (Hot Season); CS (Cold Season); P PM (Precipitation previous-month); P CM (Precipitation current-month); TPM (Temperature previous-month); T CM (Temperature current-month); UTM (latitude); HAF PM (Hectares of forest affected by fires previous-month); HAF CM (Hectares of forest affected by fires current-month); W (wind); RH (Relative humidity); R² (Coefficient of determination). PV (p-value); C (regression coefficient) for each covariate. Only significant covariates are presented; statistically significant are in bold. – Empty space means that for that congener the covariate was not statistically significant or was eliminated from the model.

is also an efficient mechanism for removing pollutants from the air. It is possible that the time frame considered in our model (previous and current month according to the sample date) does not reflect the effect of wet deposition.

For the rest of the PCDD/Fs congeners that did not have a good model adjustment ($R^2 < 50\%$), there were no significant differential trends in the coefficients (variables); however, the 123478-HxCDF and 234678-HxCDF models were positively related with UTM, which means there is spatial variability for higher levels of these congeners at higher UTM values (north area of the valley). Although the reasons are unclear, it is possible that local emission sources influence the various congener profiles and that climatic factors play an

essential role in the transfer of POPs to milk as other researchers have shown (Shunthirasingham *et al.*, 2013).

In the case of the dl-PCBs congener models that were carried out, the most relevant variables are:

-UTM coordinates: These were clearly and positively related to the dl-PCBs congeners, reaching the highest R², which is consistent with previous findings of a downward latitudinal trend. In Chile, this trend may be related to the proximity of the central region to the largest industrialized cities in the country, especially in the northern area of the valley as reported by other researchers (Kavouras *et al.*, 2001, Sanhueza *et al.*, 2009).

-Wind: Wind was negatively related with the congeners with higher R^2 . In this case, data about wind is related to wind flow velocity, but it would be interesting to incorporate the wind direction in further research because it could be possible that particle deposition occurs in other places due to different wind directions. However, particle deposition depends on aerosol size distribution and atmospheric turbulence, which is influenced by wind speed (Castro-Jiménez *et al.*, 2012).

-Year: Generally, year was negatively related with dl-PCBs pollutant levels, and this indicates the tendency that was previously described of decreased POP levels with higher mean values in 2011 and lower mean values in 2013.

In general, the statistical significance of other variables did not follow consistent trends between the various pollutant models that were carried out. The hectares of forest affected by fires did not show a statistically significant relationship with the majority of the analyzed congeners (except for dl-PCB 167 and 156). This result suggests that the emissions of congeners from this kind of source is more related to PCDD/Fs as other studies indicate (Salamanca *et al.*, 2016).

Subsequent bivariate analyses between the dependent variables (pollutant levels expressed as the total sum) and some representative covariates (Figure 1S a and b) were performed to corroborate the results of the multiple regression analysis and to isolate the different relationships. No substantial differences were found in covariates with high R^2 values. This step was performed while other multiple linear regression models were tested, using a backward selection of terms, which resulted in the masking or removal of some covariates that were known a priori to have a strong relationship. In other cases, covariates had a lower R^2 for the different congeners than the present final model, and for this reason, these covariates were maintained in the final model.

Our study has several limitations. First, we performed a retrospective study with a limited set

of data (milk sample pollution data) from seven regions with only a few sampling spots within each region, but the optimal study design would have more representative sample spots within each region and would consider a balance between the contrasting production systems used for cow milk (free-range vs. confined); a second option would be to focus on one production system and the seasonal variation of pollutant levels in the production cycle associated with the land use for pastures, forage, feed or mixes (Lake *et al.*, 2013). Furthermore, there are multiple factors possibly influencing the pollutant levels in the milk samples, such as agricultural waste burning and agrochemicals used in the pastures, that are related to local emission sources but were not covered by this model because the aim of this work was to find relationships with some of the critical variables that could affect milk samples, instead of an environmental forensic fingerprint study.

On the other hand, some relationships were demonstrated with moderate success by our use of a multifactor process that integrated several conditions and took into account the lack of regional and national scientific data about POPs in environmental, agricultural and livestock studies.

In summary, our results are useful in better understanding our previous findings, which also provided insight into how these pollutants behave in the environment and how they are influenced by different geographical-meteorological variables, similar to other studies that incorporate these kinds of variables (Castro-Jiménez *et al.*, 2012, Correa *et al.*, 2006, Aristizábal *et al.*, 2011). Future research should aim to regularly monitor PCDD/Fs and dl-PCBs pollution levels in the Chilean environment from a One Health perspective. Such information would be aid in preventing the entry of these contaminants into the environment and in creating predictive models that examine the food chain and thus protect food safety and consumer health in coming year by taking in to account the possible regional

difficulties in the animal production cycle in a climate change scenario.

Funding

This work was supported by the “Comisión Nacional de Investigación Científica y Tecnológica” (CONICYT; Doctoral Grant No. 21110904) in collaboration with the FARMAVET Laboratory, Universidad de Chile.

Acknowledgments

To the “Programa de Atracción e Inserción de Capital Humano Avanzado” (PAI-CONICYT; Project N°I7816020005). Also, the authors want to thank Dra. Natalie Urrutia C. for her help with language and grammatical editing. To Dr. Cristóbal Galbán and Dr. Cristian Estades for their help in the theory and modeling, respectively, in this article.

Resumen

N. Pizarro-Aránquiz, D. García-Mendoza, R. Muñoz, B. San Martín, y R. Morales. Impact of environmental variables on PCDD/F and dl-PCB levels in dairy milk of the farming region of Chile. 2018. Cien. Inv. Agr. 45(2): 109-119. Desde la perspectiva de “Una Salud” la importancia de los contaminantes orgánicos persistentes está dada por su impacto sobre el medio ambiente, los alimentos y finalmente las personas. A pesar de esto, la información sobre estos compuestos tóxicos es limitada en América Latina y la región del Caribe. Chile no es la excepción. El objetivo de este trabajo fue encontrar alguna explicación a los niveles de dioxinas en muestras de leche, previamente publicados, utilizando un modelo con datos de factores meteorológicos/geográficos. Para esto, se realizó un análisis de regresión múltiple general por pasos para los residuos de dibenzo-p-dioxinas policloradas (PCDD), dibenzofuranos policlorados (PCDF) y bifenilos policlorados similares a las dioxinas (dl-PCBs). Se observaron los mejores ajustes estadísticos solo para los congéneres mayoritarios. Con respecto a los congéneres de PCDD/Fs, los factores más relevantes y significativos ($P < 0.05$) fueron: Año (principalmente un coeficiente negativo), la temporada, y las hectáreas afectadas por los incendios forestales. En el caso de los congéneres de dl-PCBs, hubo una relación clara y positiva con UTM, resultado consistente con la tendencia latitudinal decreciente publicada con anterioridad. A su vez, la velocidad del viento fue un coeficiente negativo significativo para dl-PCBs. A pesar del conocimiento existente sobre los niveles de contaminantes en la leche en Chile, la información obtenida en este estudio es relevante para ayudar a entender estos hallazgos, además en la región de América Latina y el Caribe.

Palabras clave: Bifenilos policlorados similares a las dioxinas, contaminación de alimentos, contaminantes orgánicos persistentes, dibenzo-p-dioxinas policloradas, dibenzofuranos policlorados, variables ambientales.

References

- Abad, E., J.J. Llerena, J. Sauló, J. Caixach, and J. Rivera. 2002. Study on PCDDs/PCDFs and co-PCBs content in food samples from Catalonia (Spain). *Chemosphere* 46:1435–1441.
- Aristizábal, B.H., C.M. Gonzalez, L. Morales, M. Abalos, and E. Abad. 2011. Polychlorinated dibenzo-p-dioxin and dibenzofuran in urban air of an Andean city. *Chemosphere* 85:170–178.
- Castro-Jiménez, J., J. Eisenreich, G. Mariani, H. Skejjo, and G. Umlauf. 2012. Monitoring atmospheric levels and deposition of dioxin-like pollutants in sub-alpine Northern Italy. *Atmospheric Environment*, 52:194–202.
- CONAF, n.d. Datos históricos de incendios forestales en Chile. Corporación Nac. For. URL <http://www.conaf.cl/incendios-forestales/incendios-forestales-en-chile/estadisticas-historicas/> (accessed Aug 05, 2015).
- Correa, E., L. Raun, H. Rifai, M. Suarez, T. Holsen, and L. Koenig. 2006. Depositional flux of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans in an urban setting. *Chemosphere* 64:1550–1561.
- Dwyer, H., and N.J. Themelis. 2012. Inventory of U.S. dioxin emissions to atmosphere. *Waste Manag.*;46:242–246.
- Esposito M., Cavallo S., Serpe F.P., D'Ambrosio R., Gallo P., Colarusso G., Pellicanò R., Baldi L., Guarino A., and Serpe L. 2009. Levels and congener profiles of polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans and dioxin-like polychlorinated biphenyls in cow's milk collected in Campania, Italy. *Chemosphere* 77(9):1212–1216.
- Erickson, M., and R. Kalley II. 2011. Applications of polychlorinated biphenyls. *Environ Sci pollut res.* 18:135–151.
- Halfon, N., Z. Levin, and P. Alpert. 2009. Temporal rainfall fluctuations in Israel and their possible link to urban and air pollution effects. *Environ. Res. Lett.* 4:25001.
- INIA, n.d. Red de estaciones meteorológicas AGROMET. Inst. Investig. Agropecu. Chile. URL <http://agromet.inia.cl/> (accessed Aug 04, 2015).
- Kavouras, I.G., P. Koutrakis, F. Cereceda-Balic, and P. Oyola. 2001. Source Apportionment of PM10 and PM25 in Five Chilean Cities Using Factor Analysis. *J. Air Waste Manage. Assoc.* 51:451–464.
- Kotz, A. 2014. Dioxins and PCBs in feed and food — Review from European perspective. *Sci. Total Environ.* 491:2–10.
- Lake, I.R., C.D. Foxall, A. Fernandes, M. Lewis, M. Rose, O. White, and A. Dowding, 2013. Seasonal variations in the levels of PCDD/Fs, PCBs and PBDEs in cows' milk. *Chemosphere* 90:72–79.
- Loyola-Sepúlveda, R., M. Salamanca, F., Guitierrez-Baeza, C. Figueroa, C. Chandia, C. Bravo-Linares, and S. Mudge. 2018. Contributions of dioxins and furans to the urban sediment signature: The role of atmospheric particles. *Science of the total Environment*, 615:751–760.
- Luebert, F., P. and Plischoff, P., 2006. Sinopsis bioclimática y vegetalacional de Chile. Editorial Universitaria.
- Pemberthy, D., A. Quintero, M.G. Martrat, J. Parera, M. Ábalos, E. Abad, and A.L. Villa. 2016. Polychlorinated dibenzo-p-dioxins, dibenzofurans and dioxin-like PCBs in commercialized food products from Colombia. *Sci. Total Environ.* 568:1185–1191.
- Pizarro-Aranguiz, N., C.J. Galbán-Malagón, P. Ruiz-Rudolph, C. Araya-Jordan, A. Maddaleno, and B. San Martín. 2015. Occurrence, variability and human exposure to Polychlorinated Dibenzop-dioxins (PCDDs), Polychlorinated Dibenzofurans (PCDFs) and Dioxin-Like Polychlorinated Biphenyls (DL-PCBs) in dairy products from Chile during the 2011–2013 survey. *Chemosphere* 126:78–87.
- Pozo, K., V.H. Estellano, T. Harner, L. Diaz-Robles, F. Cereceda-Balic, P. Etcharren, K. Pozo, K., V. Vidal, F. Guerrero, and A. Vergara-Fernández. 2015. Assessing Polycyclic Aromatic Hydrocarbons (PAHs) using passive air sampling in the atmosphere of one of the most wood-smoke-polluted cities in Chile: The case study of Temuco. *Chemosphere* 134:475–481.
- Pozo, K., T. Harner, M. Shoeib, R. Urrutia, R. Barra, O. Parra, and S. Focardi. 2004. Passive-Sampler Derived Air Concentrations of Persistent Organ-

- ic Pollutants on a North–South Transect in Chile. *Environ. Sci. Technol.* 38:6529–6537.
- Rychen, G., S. Jurjanz, H. Toussaint, and C. Feidt. 2008. Dairy ruminant exposure to persistent organic pollutants and excretion to milk. *animal* 2:312–323.
- Salamanca, M., C. Chandía, and A. Hernández. 2016. Impact of forest fires on the concentrations of polychlorinated dibenzo-p-dioxin and dibenzofurans in coastal waters of central Chile. *Science of the total Environment*, 573:1397–1405.
- San Martín, B.V., N. Pizarro-Aranguiz, D. García-Mendoza, C. Araya-Jordan, A. Maddaleno, E. Abad, and C.J. Galbán-Malagón. 2016. A four-year survey in the farming region of Chile, occurrence and human exposure to polychlorinated dibenzo-p-dioxins and dibenzofurans, and dioxin-like polychlorinated biphenyls in different raw meats. *Sci. Total Environ.* 573:1278–1286.
- Sanhueza, P.A., M.A. Torreblanca, L. Diaz-Robles, L. Schiappacasse, M. Silva, and T. Astete. 2009. Particulate Air Pollution and Health Effects for Cardiovascular and Respiratory Causes in Temuco, Chile: A Wood-Smoke-Polluted Urban Area. *J. Air Waste Manage. Assoc.* 59:1481–1488.
- Shunthirasingham, C., F. Wania, M. MacLeod, Y.D. Lei, C.L. Quinn, X. Zhang, M. Scheringer, F. Wegmann, K. Hungerbühler, S. Ivemeyer, F. Heil, P. Klocke, G. Pacepavicius, and M. Alaee. 2013. Mountain Cold-Trapping Increases Transfer of Persistent Organic Pollutants from Atmosphere to Cows' Milk. *Environ. Sci. Technol.* 47:9175–9181.