Spatial variability of cadmium pollution around three zinc - ore smelrers in north - east Belgium

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Abstract :

This paper deals with the pollution by a heavy metal, cadmium (Cd), of the topsoil over a relatively large area in the north-east of Belgium. It consists of a typical environmental soil hazard originating from three zinc-ore smelters, complicated by the uneven distribution of the transporting medium, which is wind. The main goal of this paper is to map cadmium concentration in the soils of the study area. The topsoil Cd has a relatively high mean value, 3.86 mg Kg^{-1}, and the data ranged over more than 70 mg Kg^{-1}. A regional trend was identified, which showed a relationship between the Cd concentration and the distance to the nearest factory. Additionally, an anisotropic variability found, which was related to the dominant wind directions. To estimate the Cd concentration, results of modelling and the regional trend were used. The Cd concentration of this area is predicted to be almost everywhere above the background value of 1 mg Kg^{-1}. Moreover, close to the factories, large amounts of Cd can be expected. Finally, important areas around the factories, with extensions towards the north-east, have an enrichment of cadmium well beyond the limit of 5 mg Kg^{-1}.

Key Words :

Cadmium, Spatial variability, Pollution, Trend analysis.

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INTRODUCTION:

About one century ago, around 1880, three zinc factories were built in the communities of Balen (Vieille Montagene), Lommel (Maatheide), and Overpelt (Metallurgite). They are located within some 10 km from each other. Maatheide was dismantled several years ago, but the two other factories are still in operation. During the extraction of zinc, using the older roasting process, relatively large amounts of cadmium (Cd) were liberated. Because cadmium was not considered to be of any economical value, it was freely released through the chimneys until the fifties. However, since the seventies the production process has been completely changed. A conversion to a less environmental hazardous procedure, hydro-metallurgy, allowed to recuperate most of the cadmium both for economic and environmental reasons. So the presence of Cd in the area can be considered to be mainly of an historical nature.

Cadmium has no known biological function, but it is readily accumulated in the human body, preferentially in the soft tissues like the liver and kidney. Long term exposure to cadmium may lead to kidney damage, disturbance of the metabolism of the metallic elements in the body including iron, zinc, and copper, development of hypertension and cancer of prostate (2,10,12). Humans are exposed to the cadmium in soils mainly through the food chain. It is adsorbad by man, mainly through food and in a lesser extent by drinking. From the human health point of view, Cd in fresh vegetables (especially leaf vegetables) is considered as a risk element (6). Therefore, growing vegetable gardens can be considered as the most risky land use. It has been proposed a maximum tolerable limit of Cd in vegetable crops of 0.1 mg kg^{-1} fresh weight (8).

The purpose of this paper is to investigate the cadmium pollution in the study area using an exploratory data analysis. During this analysis the distribution properties of the cadmium data set and their spatial arrangement including the detection of regional trends are considered.

MATERIALS AND METHODS:

In the beginning of eighties, research was started to investigate the extent of the cadmium pollution in the study area. This resulted in an intensive sampling of vegetable gardens, ordered by the Flemish Executive. The home gardens were selected as a sampling target, since it is the most risky land use with respect to Cd pollution. This data set contains 1960 sample points measured for topsoil total Cd concentration.

In order to investigate possible relationship between the spatial variability of Cd and climatic characteristics, information on wind direction was also used.

RESULTS AND DISCUSSION:

Distributional properties:

Summary statistics of the topsoil Cd content is given in table 1, while figure 1 shows the corresponding sample histogram. The strong positive skewness evident in the histogram is confirmed by the summary statistics: the mean is greater than the median, and large values for both skewness and kurtosis are found.

Table 1. Descriptive statistics of the Cd (mg kg^{-1}) content.

Number of samples	**1690**
Mean	3.86
Median	3.00
Variance	16.20
Minimum	0.01
Maximum	70.50
Skewness	5.90
Kurtosis	61.60
Coefficient of Variation	104

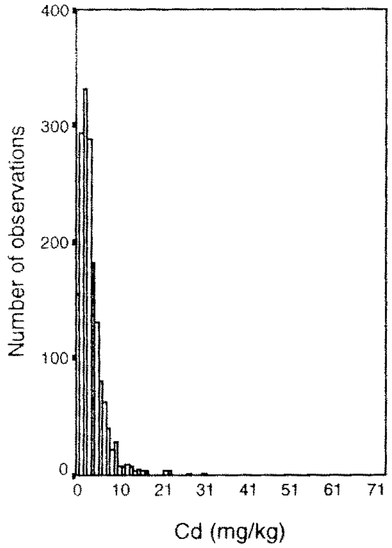

Figure 1. Sample histogram of Cd data.

The Cd data values span several orders of magnitude, from 0.01 to 70.5 mg kg^{-1}. This maximum value is almost 18 times larger than the mean value. There are several other extreme values in the cadmium data set. The histogram of Cd data gives a good impression of the overall shape of the distribution. However, to get more information on the distribution of data values, a frequency table with class intervals of 2 mg kg^{-1} is given in table 2. This table indicates that nearly a third of cadmium values falls within 0-2 mg kg^{-1} and again a third of the data are in the 2-4 mg kg^{-1} class. So, there is still a third of the remaining Cd data which are above 4 mg kg^{-1}.

Table 2. Frequency table of Cd data set, using a class width of 2 mg kg^{-1}.

Class width	**Number**	**Percentage**	**Class**	**width**	**Number**
Percentage					
0-2	517	30.59	36.38	0	0
2-4	646	38.37	38.40	0	0
4-6	275	16.27	40.42	0	0
6-8	121	7.16	42-44	0	0
8-10	57	3.37	44.46	0	0
10-12	23	1.36	46.48	1	0.059
12-14	13	0.769	48.50	0	0
14-16	12	0.710	50.52	0	0
16-18	6	0.355	52.54	0	0
18-20	0	0	0.414	44.56	0
20-22		7	0.178	56.58	0
22-24		3	0.059	58.60	0
24-26	1	0.178	60.62	0	
26-28	3	0	0.178	62.64	0
28-30		0	0	64.66	0
30-32	3	0.178	66.68	0	
32-34	1	0.059	68.70	0	
34-36	0	0	70+	0.059	

Since we are interested in the distribuiton of Cd values with respect to the threshold values, a smaller class width of 1 mg kg^{-1} was used. Results indicate that at 88% of the sites, the soil contained more than 1 mg kg^{-1}, which can be considered to be a general background value. 70% of the samples have values of more than 2 mg kg^{-1}, which corresponds approximately to the sanitation level for vegetable gardens (13). Hence, the results indicate the prevalence of soils enriched severely with Cd in this region.

Spatial properties:

Since our cadmium data are georeferenced, therefore, exploring the spatial features of the data such as the location of extreme values and the general trends in the Cd values is of considerable interest. The spatial distribution of heavy metal might be more complex than the distribution of other soil variables since their spatial variabiltiy may be influenced by both internal and external factors (1,3,4,9).

Figure 2 shows the location of all cadmium data. As a reference, the locations of the three zinc factories were also marked on the map. This figure gives a good image of the general pattern of the spatial distribution of data. It shows that most samples are clustered inside the study area. This is due to the fact that the sample population (home gardens) is clustered over the study area. Such a preferential sampling may influence the representativity of the summary statistics listed in table1.

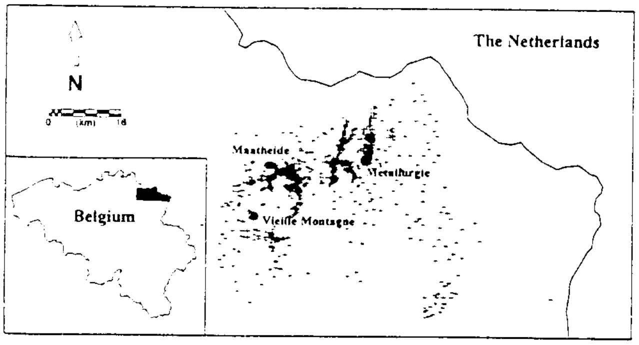

Figure 2. Localisation of the study area inside Belgium, the Cd data points (+) and the three zinc factories (grey dots).

We also draw a map of cadmium data which is classified into four classes. (see figure 3), This map reveals that most of the high cadmium values occure close to the factories, while the lowest values become dominant as one gets farther away from the factories. This map also reveals an interesting feature of the spatial distribution of the cadmium data. One can see that there are bands of high Cd values along some directions. The most prominent of these is the one that is oriented along north-east direction from the factories. This suggests that Cd values were more diffused in this direction than in other.

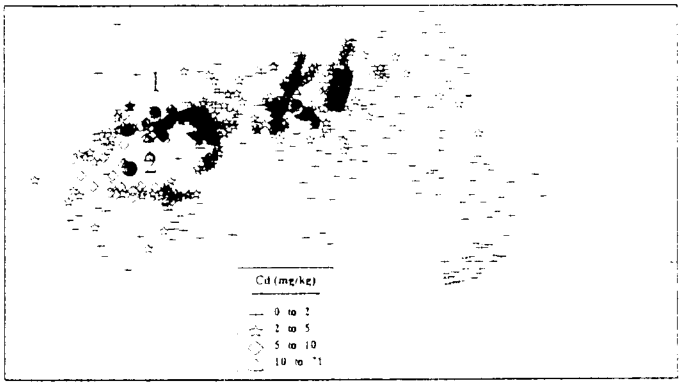

Figure 3. Classified map of Cd data values. Factories: 1 = Maatheide, 2 = Vieille Montagne, 3 = Metallurgie.

Trend analysis:

Results obtained from the studying the spatial features of the Cd data indicate that according to the available information, there are two main factors that contributed to the spatial distribution of Cd data, being (i) distance to the nearest factory, (ii) predominant wind directions.

In order to explore the effects of these two factors on the spatial features of the Cd data, we separated the Cd data according to the distance to the nearest factory, and four wind classes. We used the information on wind frequencies in the study area, and linked them to the Cd data. Figure 4 shows the year-average frequencies of wind blowing to one of the sixteen major wind directions. From this figure it could be concluded that there are two main wind orientation: the angle window north-east and the direction west. These sixteen directions were grouped into four classes of direction angles and then split the Cd data according to these four wind classes.

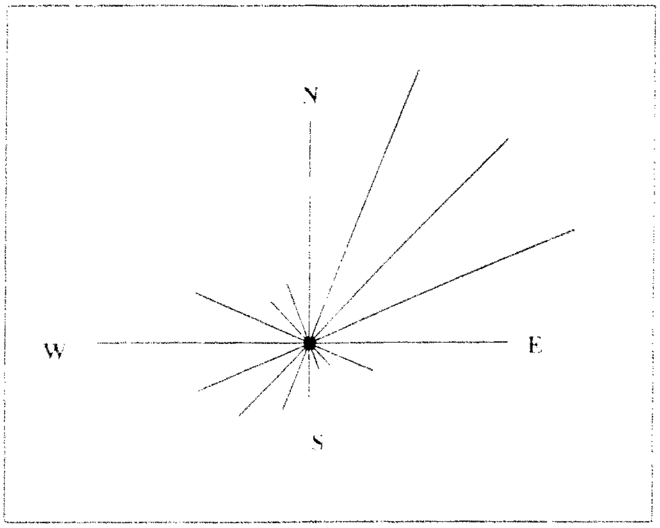

Figure 4. Yearly averaged frequencies of wind blowing to a particular direction, the longest line (NNE) corresponds to 12.5% of the days of a year.

A power function was found to model best the regional trend of the Cd data for all four wind classes as following:

$$Y = aX^b \qquad [1]$$

Where Y represents the Cd concentration (mgkg^{-1}), as a function of the distance to the nearest factory X, and a and b are both coefficients of the power function. The coefficients of the fitted models for different wind classes are given in table 3. The curves are shown in figure 5. As a result, the general shape of the trend is the same for all wind directions. Cd data values show a clear relationship witn the distance to the nearest source. It implies that the point emission of cadmium produced a regional trend in the Cd data values. Moreover, the existence of a regional trend in Cd data values shows an anisotropic behaviour: its magnitude depends not only on the distance to the nearest factory, but also on the direction. The main cause for such an anisotopic variabiltiy is the preferential wind distributions in the study area.

Table 3. The coefficients and correlation coefficients of the power funtion to the Cd values as a function of the distance to the nearest factory for four wind classes (see text for definition of model parameters):

wind classes	**a**	**b**	**r**
High frequency (336°-79°)	99.48	-0.439	-0.294**
Medium freq.(169°-281°)	1978.31	-0.822	-0.630***
Relat. Low freq.(79°-169°)	2724.39	-0.899	-0.745***
Low freq..(281°-336°)	6374.11	-0.996	-0.640***

** : significant at P=0.01

*** : significant at P=0.001

Results of the trend analysis indicate that there are differences between the model parameters for different wind classes. The differences between the slope b of the power function for the class of high wind

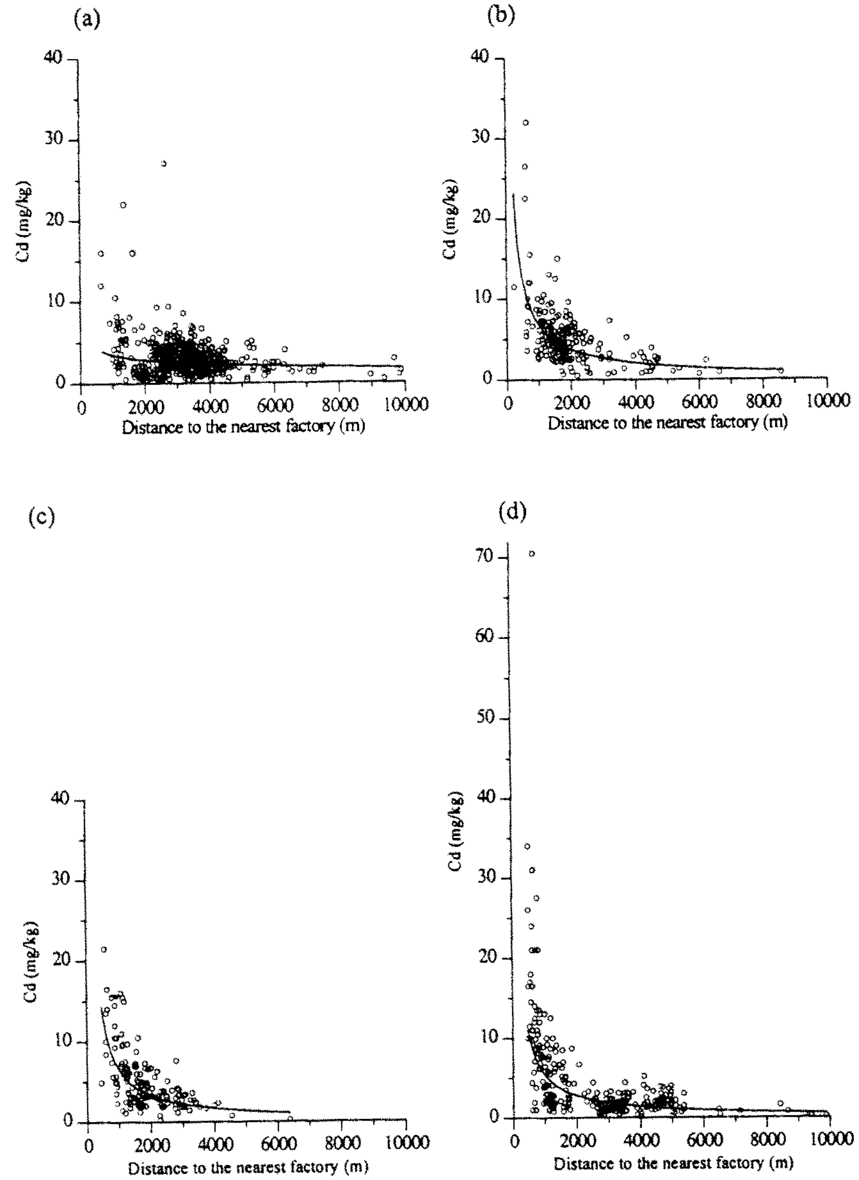

Figure 5. Fitted regional trend models for Cd data located in four wind direction classes as a function of the distance to the nearest factory: (a) wind class of high frequencies, (b) wind class of medium frequencies, (c) wind class of relatively low frequencies, (d) wind class of low frequencies.

frequencies and other classes is even prominent. The power function for the class of low wind frequencies decreases very rapidly, while for wind class of high frequencies the curve decreases very gently. Therefore, we have to expect the spatial variability of Cd data to be more continuous in the directions with high wind frequencies, while for directions with low wind frequencies some discontinuity in Cd data should be expected. Moreover, the maximum Cd value is found in the direction angle of low wind frequencies. It is apparent that the release of Cd from the chimney becomes much more diffused under dominant wind. High wind frequencies blowing to a certain angle make Cd particles to be more dispersed and diffused. While, in the direction of low wind frequencies there is a smaller chance for larger Cd particles to be transported over a long distance (5,7).

Mapping Cd concentration:

The spatial variation of Cd concentration can be mapped over the entire area using results obtained from trend analysis. Those models were used to estimate Cd concentration at unsampled locations. Figure 6 shows the map of topsoil Cd content in the study area. This map gives us a good idea of how the Cd values are spatially distributed in the study area. We can observe that Cd concentration increases strongly near the zinc factories. The extension of areas enriched in cadmium depends on the major wind directions. The estimated Cd concentrations are higher with extensions towards the north-east and also somewhat west. The Cd concentration is estimated almost everywhere above 1 mg kg^{-1}. Therefore, from practical point of view we can conclude that almost the entire area is enriched by Cd, i.e. its Cd content is above the background threshold. However, there are major areas around the factories where the Cd concentration strongly increases. Dominant wind directions enlarged these according to the main wind directions in the study area.

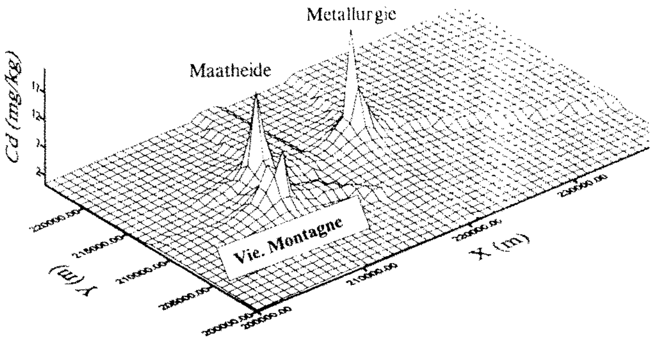

Figure 6. Estimated regional trends of Cd concentration (mg kg^{-1}).

From a methodological point of view, the spatial estimation of the Cd pollution can be done by using a deterministic model. However, the results reveal that a part of the Cd variability remained unexplained. This refers to the local variability, which should be characterized by other statistical tools. We suggest using geostatistical tools in order to characterize the structure of spatial variability of Cd data and to map it over the study area (11).

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تغییرات مکانی آلودگی کادمیوم در اطراف سه کارخانه استخراج روی واقع در شمال شرقی کشور بلژیک

دکتر جهانگرد محمدی

کلمات کلیدی:

- کادمیوم، تغییرات مکانی، آلودگی، تجزیه وتحلیل روند.

چکیده:

در این مقاله مسئله آلودگی خاک توسط عنصر کادمیوم در مناطق وسیعی واقع در شمال شرقی کشور بلژیک مورد بررسی قرار گرفته است. آلودگی مزبور ناشی از فعالیت سه کارخانه استخراج روی بوده و تحت تأثیر عوامل محیطی همچون باد واقع شده است. هدف اصلی از این بررسی عبارت از تهیه نقشه پراکنش کادمیوم در منطقه مطالعاتی می‌باشد. میانگین کادمیوم‌اندازه‌گیری شده درخاک نسبتاً زیاد، mg/kg ۳/۸۶، و دارای دامنه وسیعی بین ۰/۰۱ و mg/kg ۷۰/۵ است. در بررسی داده‌های کادمیوم یک روند منطقه‌ای مشاهده گردیده که بیانگر رابطه بین غلظت کادمیوم درخاک و فاصله تا نزدیکترین کارخانه می‌باشد. علاوه بر آن تغییرات مکانی کادمیوم دارای ناهمسانگردی می‌باشد که ناشی از اثرات باد در منطقه است. به منظور تهیه نقشه پراکنش کادمیوم، مدل‌های روند منطقه‌ای برازش داده شده بر داده‌های کادمیوم مورد استفاده قرار گرفت. نتایج بدست آمده حاکی از غلظت زیاد کادمیوم در منطقه مورد مطالعه است. از سوی دیگر غلظت کادمیوم در خاک با نزدیک شدن به منبع رهاسازی آن افزایش می‌یابد. این در حالی است که مناطق غنی از کادمیوم بخوبی اثرات وزش باد غالب در منطقه را نمایش می‌دهد.

اصل مقاله به زبان انگلیسی در انتهای Abstract است (سردبیر) .

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