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A Fuzzy Model of Heavy Metal Loadings in Marine Environment

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Abstract

We design a fuzzy model of the loadings of heavy metals for two coastal areas of the Irish Sea (Liverpool Bay and Morecambe Bay). Each metal concentration is associated with a fuzzy set "contaminated", defined over a set of sampling sites (70 in Liverpool Bay and 203 in Morecambe Bay). The higher the concentration, the higher the degree of membership of the site. Six overall loading indices are calculated using aggregation connectives between fuzzy sets. The loading indices are then interpolated and plotted on a map. A visual inspection shows that: (i) product aggregation is most indicative for the locations of the disposal grounds; (ii) mean aggregation reflects well the sediment movement in the bay; (iii) maximum aggregation indicates all highly contaminated sites. The proposed fuzzy model is easy to implement and the results are directly interpretable.
Keywords: Fuzzy aggregation connectives, environmental modelling, Liverpool Bay, Morecambe Bay, heavy metal concentrations, index of spatial distribution, spatial data analysis.

1 Introduction

Near shore waters have been used as a resource for the disposal of waste material over a prolonged period extending back to the later part of the industrial revolution. In the case of the Irish Sea disposal of sewage material from the heavily populated industrial region of south Lancashire has been carried out since the late 1800's. For most of this time the attitude to disposal may be summarised as 'out of sight out of mind'. With the development of reliable and sensitive methods for measuring trace contaminants, questions have been raised as to the environmental impact of long-term disposal in coastal areas.

Assessing the impact of such long-term activities in areas such as Liverpool Bay and Morecambe Bay (Figure 1) is fundamental to environmental risk management in coastal zones of the Irish Sea.

There are several strands to impact assessment.

1. Assessing the degree of contamination above normal, or background, levels.

2. Defining the pattern, or the geographical distribution, of contamination.

3. Determining the likely changes in contamination with time.

This present work addresses the first two components of risk assessment and management by analysis of data sets on metals in the surface sediments of two distinct coastal environments of the Irish Sea.

The dynamic nature of coastal waters presents a severe challenge to environmental assessment of disposal activities in near shore waters. Surface sediments lying in shallow water act as a primary sink for pollutants delivered through rivers and groundwater flow and are most often studied in an attempt to identify and assess contaminant sources. However, sediments are subjected to strong oscillating tidal forces resulting in bed load transport; storms may result in mixing of contaminated sediment with material from a different source and organisms in sediments may re-work deeper consolidated material to the surface layer. The overall effect of these uncontrollable environmental variables is to obscure spatial data.

The basic problem centers on combining metal data in a way which produces a meaningful distribution pattern given that the metal have different concentration scales. Large data sets are being collected and stored, awaiting processing and analysis. The current study falls into the general (and controversial) category of *statistical spatial data analysis* as defined in (Bailey, 1994). This type of analysis has been recognized as an important research line, gaining speed in the past 10 years and trying to establish its identity. The importance lies with the demand on the part of *Geographical Information Systems* (GIS) for "systems that 'do something' other than display and organize data" (Fotheringham and Rogerson, 1994). In this study we have multiattribute data, i.e., for each location a set of metal concentrations has been measured. The problem is to combine this information for each location and to devise a contamination distribution picture of the whole area of interest. Typical choices for processing such data are principal component analysis (PCA) or cluster analysis, possibly because these techniques are available in most statistical software packages. The results of both methods are difficult to interpret, unless the data has favorable structure and characteristics. Fuzzy set modeling is a straightforward option for this kind of problems.

We study the loadings of 10 heavy metals in Liverpool Bay and 7 in Morecambe Bay, and design *loading indices* to represent the overall metal concentration. Section 2 describes the environmental problem and the data set. In

Figure 1: Liverpool Bay and Morecambe Bay location

Section 3 we briefly introduce fuzzy sets and give the fuzzy aggregation operators used to design the loading indices. The results are shown and discussed in Section 4 along with PCA and cluster analysis results.

2 Liverpool Bay and Morecambe Bay data

The situation in Liverpool Bay is complicated by multiple sources of contaminants. The bay receives heavy metals from continuous sources (Mersey and Dee Estuaries), point sources (offshore disposal ground) and a higher than normal input of some heavy metals (notably arsenic) through erosion of natural mineral sources (Camacho-Ibar, 1992). Although water circulation in the bay depends on tides, winds, freshwater inputs, etc., it has been found that there is a well established estuarine-like circulation induced by the low-density freshwater inputs from the rivers (the Mersey in particular) and by higher density sea water from the Irish Sea (Camacho-Ibar, 1992). Low-density water moves offshore through the surface and high-density water moves inshore near the bottom. This density-driven inshore movement of bottom water, coupled with tidal asymmetry producing stronger flood than ebb tides, induces a net sediment transport directed east and south-east toward Mersey. The combined effect of environmental factors and multiple sources of contamination is to generate complex and changing patterns in the distribution of metal contaminants in surface sediments. The problem from the point of view of environmental management is to develop patterns from metal data which reflect the current status of sediments. The impact of metal contamination on the biota in sediments is complex; exposure of organisms to high levels of more than one metal introduces further environmental stress. It is therefore appropriate that an approach to developing patterns of contaminant distribution should include all metals.

Environmental management in Liverpool Bay is based on yearly analysis of heavy metals in surface sediments from a sampling grid approximately 20 km^2. Figure 2 (lefthand side plot) shows the Liverpool Bay area, the disposal ground and the sampling sites (stations). Interpretation of the sampled data is limited by the factors described above and there is a need to develop a rational protocol whereby a realistic picture may be produced from spatial data.

The data set in this study consists of concentrations of 10 heavy metals relative to aluminium content, measured between 14th and 16th September 1988 at the 70 sites on the sampling grid. The metals are: mercury (Hg), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), arsenic (As), manganese (Mn), and iron (Fe).

The same stations have been sampled every year. A database has been collected over the period 1986-1993. Our aim in this pilot study was to develop a mathematical tool for analysing metal distributions using one data set. We chose the 1988 data set because a thorough analysis of the processes in Liverpool Bay for year 1988 can be found in (Camacho-Ibar, 1991, 1992).

Morecambe Bay is geographically contiguous with the northern sector of Liverpool Bay (Figure 1) and this area has not been used routinely for disposal of

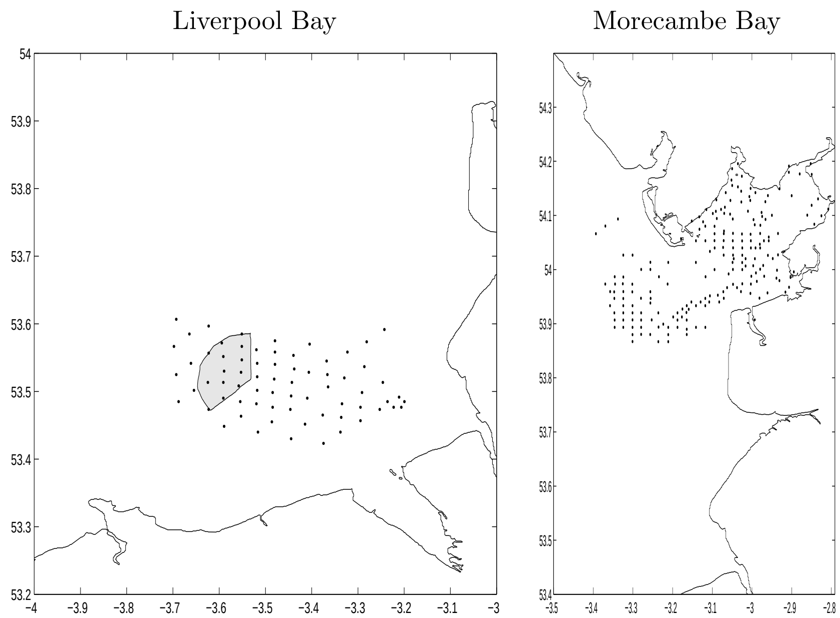

Figure 2: Liverpool Bay, the disposal ground (light gray area), Morecambe Bay, and the sampling sites (70 in Liverpool Bay and 203 in Morecambe Bay).

waste organic material. Morecambe Bay sampling stations are shown in Figure 2. A large data set for the late 1980's has been used for the generation of patterns of metals in surface sediments for purposes of comparison with Liverpool Bay.

In this study, Liverpool Bay contamination is the central environmental theme, and Morecambe Bay data is only used as a counter example. The loading indices for Liverpool Bay should be able to verify the low level of contamination of Morecambe Bay.

3 Loading Indices design by fuzzy aggregation

3.1 Fuzzy sets

Lotfi Zadeh introduced the simple and intuitive concept of a fuzzy set in his seminal paper in 1965 (Zadeh, 1965). Since then fuzzy sets have been applied to a vast number of areas including environmental sciences: soil, forest and air pollution, meteorology, water resources, etc. (Bezdek, 1999).

Let U be an ordinary set with elements $u_1, \ldots, u_m$. A *fuzzy set* A on U is defined by assigning a *degree of membership* between 0 and 1 to each $u_i \in U$, usually with regard to a linguistic term. For example, let U be the set of integers from 1 to 100 denoting the age of a person, and let A be "middle aged". We can define a (subjective!) function that assigns to each u_i a degree of membership $\mu_A(u_i) \in [0,1]$. Degree 0 denotes non-membership, degree 1 denotes full membership, and any other value is partial membership. A plausible model of "middle aged" will be obtained using a function (*membership function*) that yields high values between, say, 40 and 55 and gradually decreases towards the two edges of the scale. Thus, the degree of membership of 37, $\mu_A(37)$, can be 0.75, and of 82, $\mu_A(82) = 0.1$. A fuzzy set is determined by its membership function, so the two notions will be used interchangeably.

Let $S = \{s_1, \ldots, s_{70}\}$ be the set of 70 sites in Liverpool Bay. Let $A_1, \ldots, A_{10}$ be fuzzy sets on S, one for each metal, with membership functions

$$\mu_{A_i} : S \to [0,1], \quad i = 1, \ldots, 10.$$

The higher the ith metal concentration at site s_j, the higher the degree of membership $\mu_{A_i}(s_j)$. We chose the simple rescaling to devise the 10 membership functions from data: Let $x_i(s_j)$ be the concentration of the ith metal measured at site s_j, and let

$$LB^i_{min} = \min_{k=1}^{70}\{x_i(s_k)\},$$

and

$$LB^i_{max} = \max_{k=1}^{70}\{x_i(s_k)\}.$$

Then

$$\mu_{A_i}(s_j) = \frac{x_i(s_j) - LB^i_{min}}{LB^i_{max} - LB^i_{min}} \tag{1}$$

Figure 3 plots the membership function of mercury over the 2-dimensional space (sampling ground) spanned by the 70 sites in Liverpool Bay. Referring the plot to the original geographical problem, we observe a high concentration of mercury in the area of river estuaries. The contamination with mercury in that area is higher than that at the disposal ground (the second highest peak).

Another way of representing the membership functions (adopted here) is to use color or contour plot and overlay the scatterplot of the sampling sites.

Clearly, the concentration pattern would be the same if we did not scale the concentration between 0 and 1. Although individual metal distribution is an interesting topic on its own, it was argued above that an index of overall loading (contamination) is needed.

3.2 Fuzzy aggregation connectives

Dubois and Prade (Dubois, 1997) point out that although fuzzy membership functions have numerous possible interpretations, fuzzy mathematics has gone a long way disregarding fuzzy sets semantics: "The risk is to leave the user with no guidelines about how to apply fuzzy set theory..." They distill three main semantics: similarity, preference and uncertainty. The interpretation of the fuzzy sets used here fits best in the second category: preference (in a broad sense) because the membership functions do not measure a similarity to some

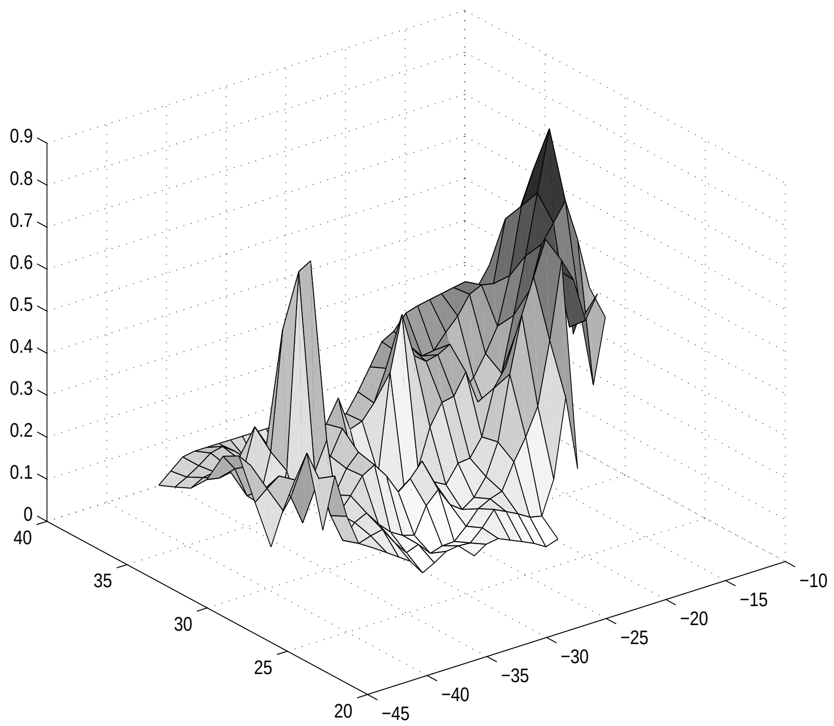

Figure 3: Hg membership function for the fuzzy set "contaminated". Th horizontal axis with the negative numbers is longitude between -4.00 and -3.00 degrees (west of Greenwich) and the one with the positive numbers is 53 degrees and decimal minutes north. The vertical axis is the degree of membership (between 0 and 1).

"ideal" prototypes nor do they express any type of uncertainty. For example, we *prefer* (to call "contaminated") a site with degree of membership 0.7 to a site with degree 0.4. Taking this interpretation, fuzzy decision-analysis approach seems reasonable. Fuzzy aggregation connectives (aggregation operators) will be used to define overall loading indices.

An m-place *aggregation operator* $\mathcal{A}$ is defined as

$$\mathcal{A} : [0,1]^m \quad \rightarrow \quad [0,1],$$

satisfying the following properties:

1. *Limit conditions*:

$$\mathcal{A}(0,0,\ldots,0) = 0, \;\; \mathcal{A}(1,1,\ldots,1) = 1.$$

2. *Commutativity*:

$$\mathcal{A}(a_1, a_2, \ldots, a_m) = \mathcal{A}(b_1, b_2, \ldots, b_m),$$

where $b_1, \ldots, b_m$ is any permutation of $a_1, a_2, \ldots, a_m$, $a_i \in [0, 1]$, $i = 1, \ldots, m$.

3. *Monotonicity*:

$$\mathcal{A}(a_1, a_2, \ldots, a_m) \geq \mathcal{A}(b_1, b_2, \ldots, b_m),$$

for any $a_1, a_2, \ldots, a_m, b_1, \ldots, b_m \in [0, 1]$, such that $a_i \geq b_i, \forall i = 1, \ldots, m$.

There are a great variety of fuzzy connectives and aggregation operators (Bloch, 1996, Dubois, 1985, Grabisch, 1995, Yager, 1994). Since this is a pilot study, here we use perhaps the simplest 6 aggregation operators:

1. *Pessimistic-type aggregation*

   - Minimum

$$\mathcal{A}_1(a_1, a_2, \ldots, a_m) = \min\{a_1, a_2, \ldots, a_m\},$$

   - Product

$$\mathcal{A}_2(a_1, a_2, \ldots, a_m) = a_1 \cdot a_2 \cdot \ldots \cdot a_m.$$

   - Geometric mean

$$\mathcal{A}_3(a_1, a_2, \ldots, a_m) = (a_1 \cdot a_2 \cdot \ldots \cdot a_m)^{1/m}.$$

2. *Optimistic-type aggregation*

   - Maximum

$$\mathcal{A}_4(a_1, a_2, \ldots, a_m) = \max\{a_1, a_2, \ldots, a_m\},$$

3. *Indifferent-type aggregation*

   - Arithmetic mean

$$\mathcal{A}_5(a_1, a_2, \ldots, a_m) = \frac{1}{m}\left(a_1 + \ldots + a_m\right).$$

   - "Competition jury". This is an operator, where we discard the highest and the lowest values from the set $a_1, a_2, \ldots, a_m$, and average the remaining $m - 2$ values.

$$\mathcal{A}_6(a_1, a_2, \ldots, a_m) =$$

$$\frac{1}{m-2}\left(a_1 + \ldots + a_m - \max_k a_k - \min_k a_k\right).$$

Replacing a_i with $\mu_{A_i}(s_j)$, each of these 6 aggregation operators defines a **Loading Index** as a fuzzy set on S. The indices are denoted respectively $\mathrm{LI}_1, \ldots \mathrm{LI}_6$.

4 Results

Table 1 shows the correlation coefficients ($\times$ 100) between the metal concentrations in Liverpool Bay.

Table 1: Correlations between the metal concentrations ($\times$ 100)

	Cd	Cr	Cu	Ni	Pb	Zn	As	Mn	Fe
Hg	0	-16	-23	-34	-31	-23	-27	-31	-27
Cd		55	54	63	71	70	51	55	48
Cr			77	83	76	85	70	64	87
Cu				70	77	80	60	57	78
Ni					78	84	65	73	79
Pb						95	81	82	78
Zn							88	88	88
As								90	87
Mn									76

Cluster analysis is often used in environmental studies to find spatial areas corresponding to values “low”, “medium”, and “high” of a certain parameter indicative of the concentration (Markus, 1996). When this parameter is based on more than one variable, e.g., contamination with heavy metals, clustering might not be appropriate. Figure 4 shows the difference between “favourable data” (where the clusters correspond to “low” and “high” contamination) and “unfavourable data” (where such correspondence does not exist). Shown on the left plot are two clusters found by the hard c-means (HCM) clustering procedure for zinc (Zn) and lead (Pb) from the Liverpool Bay data. Their concentrations correlate well (0.95), so the two cluster centers can be labeled “low” and “high” as shown. The plot on the right shows the results from the same procedure applied to mercury (Hg) and lead (Pb). The metal concentrations have a negative correlation coefficient (-0.31), so the cluster centers (circled) do not correspond to “low” and “high” in any order.

When more than two components are involved such inconsistencies are difficult to resolve, likely to be obscured, and the result can be misinterpreted.

As Table 1 shows, most of the correlations are positive (due to high concentrations around the waste disposal ground and low elsewhere) except for Hg, and therefore cluster analysis can be expected to produce sensible results. For three suspected clusters the hard c-means algorithm on the scaled data (eqn. (1)) found the centers shown in Table 2.

All center components but the first one (Hg) are ordered so that the three clusters correspond to “high”, “low”, and “medium”, respectively. Figure 5 shows the result as the spatial distribution of loading on the geographical sampling grid. Dark regions correspond to high metal loading, and bright regions, to low loading. Hard c-means clustering uses random initialization, hence different sets of centers can be obtained. The two plots show the results from clustering

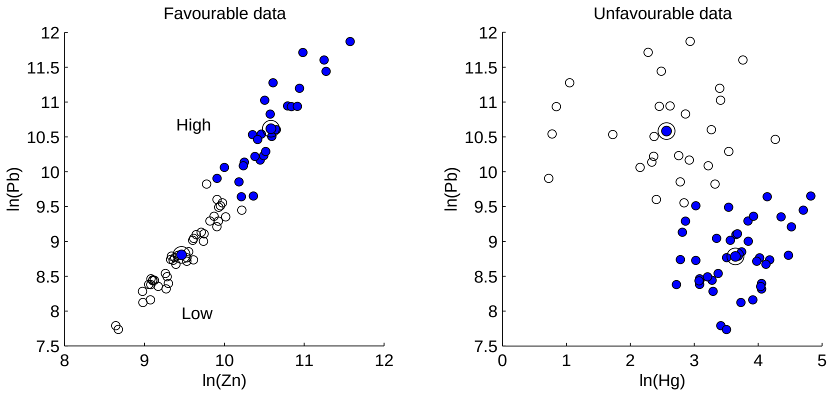

Figure 4: Scatterplot of “favourable” and “unfavourable” data

Table 2: Three cluster centers from one HCM run

	Cluster 1	Cluster 2	Cluster 3
Hg	0.1259	0.3011	0.2448
Cd	0.4159	0.0810	0.1679
Cr	0.7228	0.1739	0.5275
Cu	0.6875	0.0663	0.3147
Ni	0.7823	0.1846	0.4810
Pb	0.5663	0.0322	0.1836
Zn	0.5707	0.0674	0.2413
As	0.4846	0.0252	0.1312
Mn	0.4184	0.0092	0.0863
Fe	0.7141	0.1339	0.3602

the original metal concentrations (left) and the scaled data (right). Clustering results are influenced by data transformation (Duda and Hart, 1973). This effect is demonstrated by the differences in the two plots and clearly raises the question of whether the data should be scaled. If not, the metals with concentrations which are by orders of magnitude higher than the others will dominate and determine the clustering result. On the other hand, if we decide to scale the data, we need to choose a scaling method (e.g., taking the logarithm, z-normalisation, scaling as in eqn (1), etc.). Each of these methods might lead to a different clustering result.

Fuzzy c-means has also been used for clustering purposes in spatial data analysis (Markus, 1996). It is debatable, however, what the added value of using fuzzy c-means is over that of the hard c-means.

Principle Component Analysis (PCA) gives results that are not easily interpretable in the general case. Here, the variance of most metals is along the

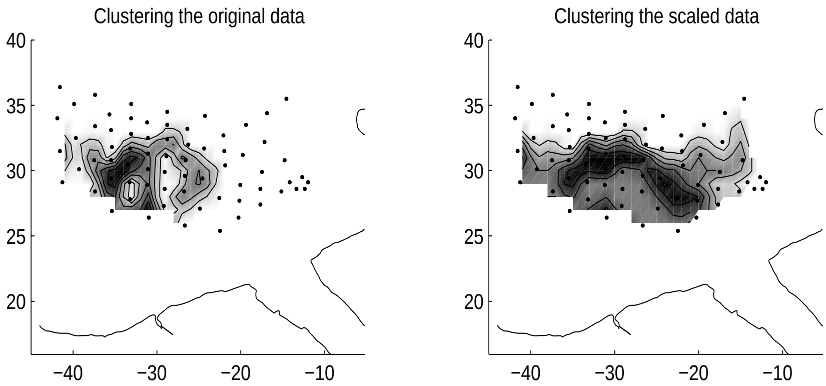

Figure 5: Overall distribution of the 10 heavy metals in Liverpool bay calculated by hard c-means clustering

same axis: high values at the disposal site, which are an order of magnitude higher than the values elsewhere. In this case the first principal component should follow the pattern found by the HCM clustering. Figure 6 shows the distribution defined by the first and the second principal components. While the first component might correspond to metal loading, the second component is not easy to name. The top two plots show the results with the original data, and the bottom two plots, with the scaled data. As the figure shows, similarly to clustering, PCA is sensitive to the type of data scaling. The difference is especially clear in the second principal component which makes its interpretation even more obscure.

Contour maps of the loading indices $\mathrm{LI}_1, \ldots \mathrm{LI}_6$ for the 10 fuzzy sets $A_1, \ldots, A_{10}$ are plotted in Figure 7.

The LI results identify clearly a number of important patterns in the spatial data. The feature common to all 6 methods is the area of low metal contamination in the north west (top left) section of the sampling grid. The product method of calculation efficiently resolves the disposal ground whilst most of the other methods identify a residual south easterly movement of material from the disposal ground towards the estuary mouth. The smaller high area in the extreme eastern sector (seen most clearly in the Maximum plot) identifies a widening area at the estuary mouth where there is net deposition of suspended material delivered from industrial sources higher up the River Mersey.

We calculated the six indices with the Morecambe bey data. To make the results comparable, we used formula (1) with the limits LB^i_{min} and LB^i_{max} of metal i as in Liverpool Bay. All negative values were set to 0 and all values greater than 1 were set to 1. Figure 8 gives an example of the scaling of Morecambe data using the liverpool Bay limits.

Because many of the concentrations in Morecambe Bay were lower than LB^i_{min}, a significant number of degrees of membership were set to 0. Thus the product and the minimum loading indices produced entirely flat distributions

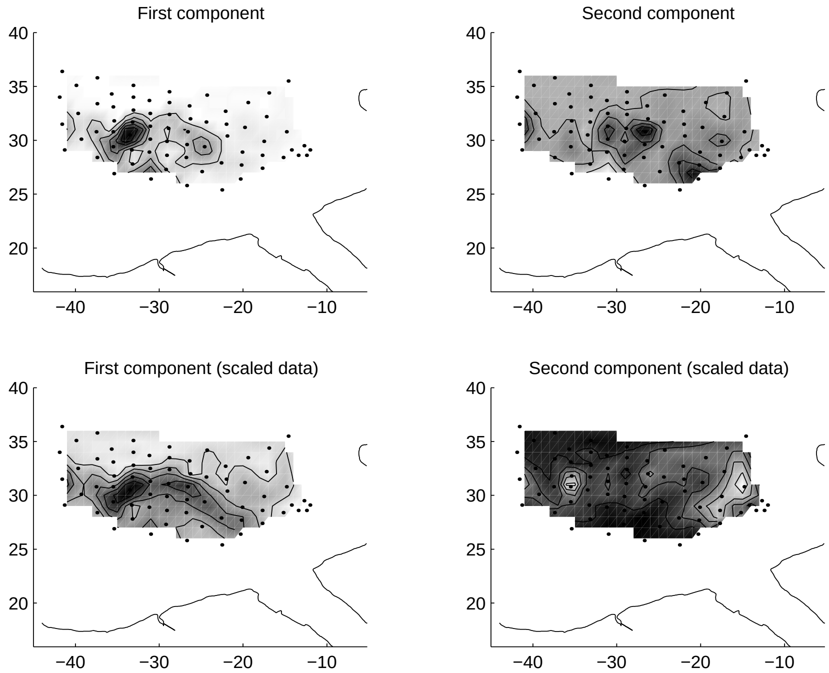

Figure 6: Overall distribution of the 10 heavy metals in Liverpool Bay by the first and second principal components of PCA

indicating no significant contamination of Morecambe bay. The maximum loading index is the most sensitive of all six and would label a site as contaminated if even one metal has high concentrations, regardless of the concentrations of the other metals. Figure 9 shows the distributions of maximum and product loading indices for Morecambe Bay. As with Liverpool Bay, the product loading index generates a pattern which is easily interpreted. The featureless distribution in Morecambe Bay indicates, in direct contrast with Liverpool Bay, no point source for heavy metals. This observation is consistent with the fact that specific areas of Morecambe Bay have not been used for routine disposal.

Initial plots of the maximum loading index showed almost blanket contamination of Morecambe Bay. Stepwise removal of individual metal data sets revealed that this effect was due entirely to lead. The question of lead in Morecambe Bay will be addressed in a future paper. For the purposes of this present work, the maximum loading index plot in Figure 9 has been generated without the lead data. The maximum loading index is a sensitive indicator of possible contamination and the resulting pattern is more complex. There are isolated areas of apparent contamination which require further examination of the data for specific sampling stations.

The difficulty in assessing results such as those in this study comes from the

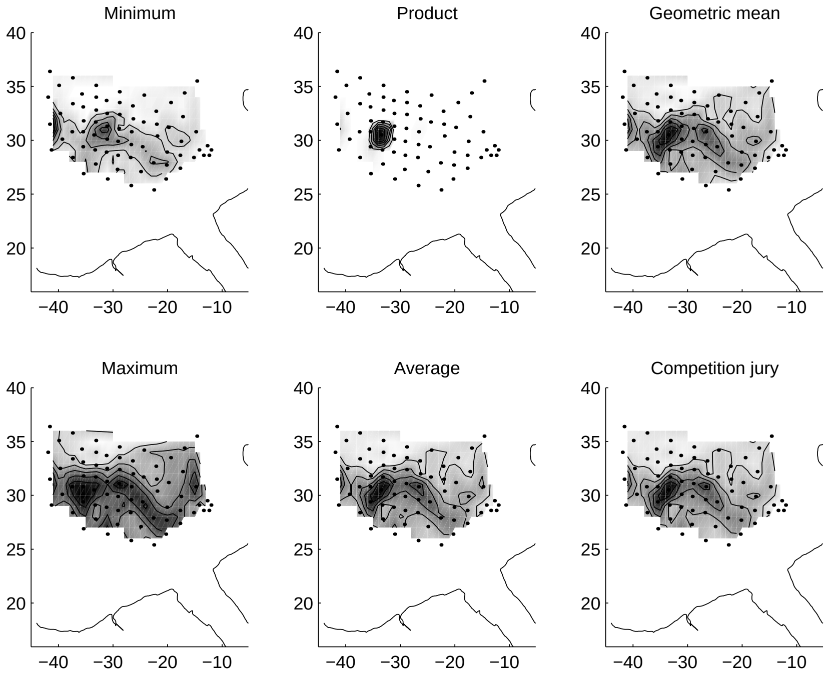

Figure 7: Overall distribution of the 10 heavy metals in Liverpool Bay calculated by the 6 Loading Indices

fact that there is no benchmark against which the new solution can be matched. The *plausibility* of the results can be judged only by eye and the interpretation will therefore be subjective. *Product aggregation* clearly indicates where the highest contamination is. This loading index may be favored by the user responsible for the waste disposal. *Maximum aggregation* shows all contaminated sites even if the contamination is due to just one of the components. This loading index may be selected by the user concerned about, say, the fish diversity in the region. The bottom line is that there is no *true* loading distribution nor is there a single one that can be "useful" from all points of view. What the proposed fuzzy approach offers is a collection of indices, each one with comprehensible interpretation, thereby giving the user a chance to make an educated choice. In this respect the fuzzy sets approach to spatial data analysis has an advantage over clustering and PCA where the interpretation is not straightforward and the results are at the mercy of the data.

An Index of Toxicity can also be designed by weighting the fuzzy sets $A_1, \ldots, A_{10}$ with respect to the toxicity of the metals and then applying a proper fuzzy aggregation, if the relative toxicities are known. There are many fuzzy set connectives that can incorporate individual weights for the fuzzy sets but the problem here is more complex. Different combinations of metals could have

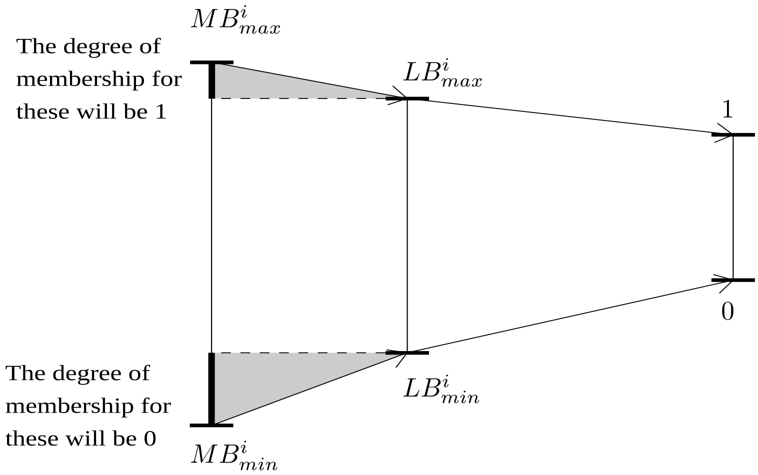

Figure 8: An example for calculating the degrees of membership of Morecambe Bay data using Liverpool Bay limits LB^i_{min} and LB^i_{max}.

different implications on the biota. Besides, these implications could be specific for different groups of species. Therefore, a more complex coefficient scheme has to be considered where each combination of metals has its own weight. For example, for Pb, Mn and Hg the aggregation rule should be able to account for seven toxicity coefficients: for (Pb), (Mn), (Hg), (Pb, Mn), (Pb, Hg), (Mn, Hg), and (Pb, Mn, Hg). An apt fuzzy model for this type of weighted aggregation is the fuzzy integral (cf. Grabisch, 1995). However, determining a set of coefficients that assess the joint effect of two or more metals on a variety of species is not a trivial task.

5 Conclusions

We show how fuzzy set theoretic aggregation operators can be used for modeling the spatial distribution of a set of variables in environmental problems, thereby providing the non-mathematical user with a simple and effective modeling tool. We applied 6 different fuzzy aggregation techniques to a set of heavy metal concentrations sampled from Liverpool Bay. For verification of our findings we used data from Morecambe Bay which is supposed to be low- or non-contaminated. The results were displayed as geographical scatterplots of metal loading and were assessed visually. Morecambe Bay data analysis supported our conclusions about the loading indices. The low contamination appearing on the maximum aggregation loading index plot was not unexpected and we offered a hypothesis about its origin. The other indices such as product and minimum, indicated no

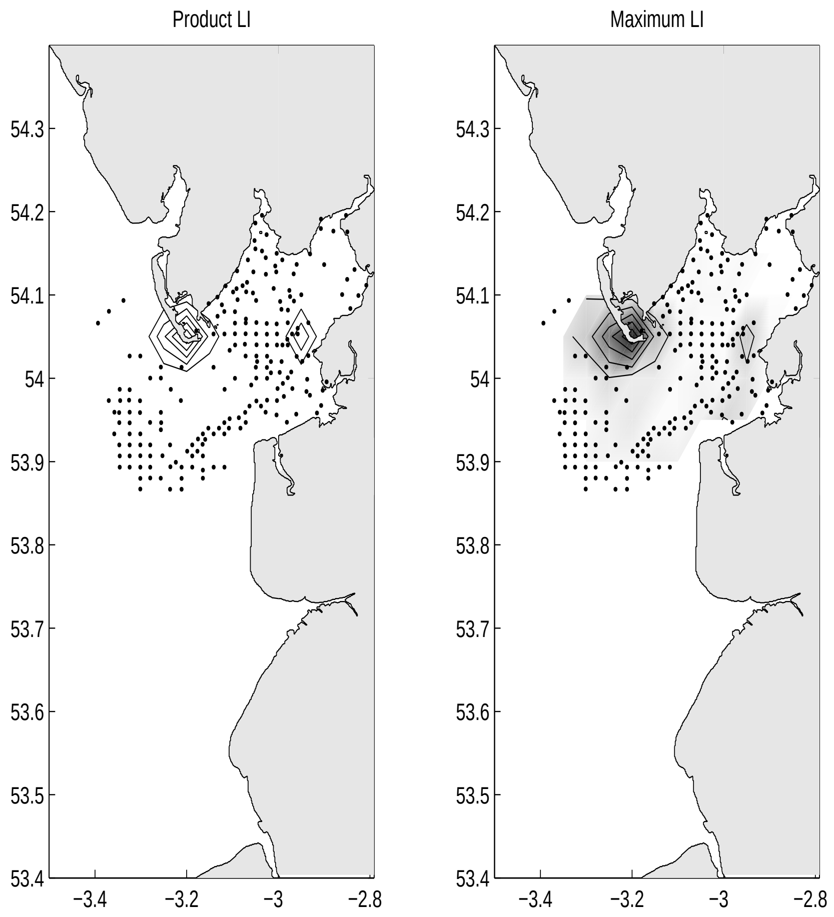

Figure 9: Product and maximum LI for Morecambe Bay

contamination across the whole bay, which confirmed the supposed difference of contamination between Liverpool Bay and Morecambe Bay. Unlike PCA and HCM, fuzzy aggregation offers a variety of plots with different information in them. On the two edges of the scale are the product aggregation, which resolves the disposal grounds clearly, and the maximum aggregation, which identifies all possible sites with suspected high contamination. The main advantage of our fuzzy model over PCA and clustering is that the results are directly interpretable in the domain context.

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