

Ranking and Screening Hazardous Chemicals for Human Health in Southeast China

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Abstract

Copeland and comprehensive multi-index comparison methods were used to rank and screen hazardous chemicals using original and pre-treatment data sets. The results show that the Copeland method can yield similar results for the two data sets. The results of a comprehensive multi-index comparison with the pretreatment dataset also show some similarities to those obtained using Copeland method. The results of the two methods show 18 common chemicals that belong in the top 20 chemicals. Of these chemicals, six are different types of dichlorodiphenyltrichloroethane, seven are POPs, three are polycyclic aromatic hydrocarbons, and two are pesticides. These substances should be regarded as chemicals of concern, and appropriate handling should be followed. Overall, the Copeland method with the original dataset can rapidly, reasonably, and effectively rank and screen hazardous chemicals.

Keywords: Chemical risk ranking; Priority setting; Copeland method; Comprehensive multi-index comparison; Human health

Introduction

With the recent rapid development and change in technologies, the chemical industry frequently produces new materials that can pose a threat to public health and the environment if improperly handled. Approximately 46,000 chemicals have been registered in China. However, a significantly higher number of chemicals have been produced and used, with some produced in significant amounts. The chemicals have brought considerable convenience but can also have drastic effects to the environment and human health. Thus, the management of chemical hazards has become increasingly important. Screening, ranking, and scoring systems are key technologies to the determination of hazardous chemicals.

Ranking, decision-support, and scoring systems can be used to determine potential risks. In environment chemistry, ranking and scoring systems are always used to identify the hazardous chemicals as well as the project to consider. Although numerous ranking and scoring systems have been developed, a consensus on the effective ranking methods has been made. In recent years, chemical risk-ranking programs have been implemented in China, for example, to identify which chemical should be placed in the priority pollutant lists.

To date, chemical risk ranking and scoring methods have been developed in countries such as the US, Canada, EU members, Japan, and Germany. In general, ranking methods can be classified as a "scalar approach method" and "vector performance" method by Halfon and Reggiani [1]. The "scalar approach method" means that an overall rank or score is determined by its own characters. Each object can obtain a score according to the indicators used in the ranking model and the score will not change. The objects are then ranked according to these scores. "Vector performance" is based on the elements of the vector and uses mathematical analysis to obtain the scores. The vector is created by using the indicators of objects. An increase or decrease in the objects will affect the scores. The variability of the score is the main difference between the two methods. Some examples of the "scalar approach method" are CHEMS-1 by Swanson et al. [2], CHEMS-2 by Dunn [3], EURAM by Hansen et al. [4], and SCRAM by Snyder et al. [5]. Some examples of "vector performance" are the Hasse diagram

by Halfon et al. [1], Copeland score method (Al-Sharrah [6]), and the comprehensive multi-index comparison (Ren and Xiong [7]).

The aim of this paper is to identify the chemicals that are hazardous to human health in Southeast China and to determine the hazard ranking of these chemicals. The results may provide realistic information that can be used in developing hazard control policies and management. In this study, human health effects, environmental effects, octanol-water partitioning, bioaccumulation, human exposure concentration, and frequency of detection were used as indicators to determine the chemical hazards. The human exposure concentration data of these chemicals were obtained by the project team that completed the previous measurement. The other data for the study can be found in the "Case study" section. The Copeland score method and comprehensive multi-index comparison method were used in this study.

Principle of the Method

Copeland score method

The Copeland score method was proposed by A. H. Copeland (1951) in a seminar on applications of mathematics to the social sciences at the University of Michigan [8]. It is a simple nonparametric method that has been used to evaluate the election results after voting. To use the method outside the voting field, candidates are replaced by objects, and votes are replaced by indicators. Two papers reported on the investigation of the properties and flaws of the Copeland score method [9,10].

The Copeland score method is based on a comparison of one

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indicator with another for each pair of objects. For example, assume that the number of chemicals is m , and each chemical has n indicators. From there, we can create a matrix X , as follows:

$$X = \begin{bmatrix} x_{11} & x_{12} & \cdots & x_{1n} \\ x_{21} & \cdots & \cdots & \vdots \\ \vdots & \cdots & \cdots & \vdots \\ x_{m1} & \cdots & \cdots & x_{mn} \end{bmatrix}$$

The next step is to compare each indicator for chemical i and j . $S_{k,ij}$ is the result of the k indicator comparison. The original Copeland has elements of the comparison matrix of (1,0,-1), so we quote this matrix in our method. Equivalent to the matrix are the (1, 1/2, 0) and (1/3, 1/6, 0) [9].

$$S_{k,ij} = \begin{cases} 1 & x_{ik} > x_{jk} \\ 0 & x_{ik} = x_{jk} \\ -1 & x_{ik} < x_{jk} \end{cases} \quad k = 1, 2, \dots, n$$

The sum of the comparisons is set as a comparison matrix A . a_{ij} is the sum of comparisons for chemicals i and j . The comparison matrix is a special matrix; it has all zeros as its diagonal elements because a comparison of a variable to itself always results in zero. The element in any row i and j is the negative of the element in rows j and i , respectively [6].

$$A = \begin{bmatrix} a_{11} & a_{12} & \cdots & a_{1m} \\ a_{12} & \cdots & \cdots & \vdots \\ \vdots & \cdots & \cdots & \vdots \\ a_{m1} & \cdots & \cdots & a_{mm} \end{bmatrix}$$

$$a_{ij} = \sum_{k=1}^n S_{k,ij}$$

After evaluating all the matrix elements, the sum of the row forms the Copeland score for each chemical:

$$CS_i = \sum_{j=1}^m a_{ij}$$

Consequently, the chemicals are ranked according to the Copeland score values. However, the method does not guarantee that the chemicals would have different scores; some objects may have the same rank. The Copeland scores can be easily calculated using a computer.

Comprehensive multi-index comparison method

Again, assume that the number of chemicals is m , and each chemical has n indicators. From there, we can create a matrix X :

$$X = \begin{bmatrix} x_{11} & x_{12} & \cdots & x_{1n} \\ x_{21} & \cdots & \cdots & \vdots \\ \vdots & \cdots & \cdots & \vdots \\ x_{m1} & \cdots & \cdots & x_{mn} \end{bmatrix}$$

The minimum and maximum values of each column of matrix X is normalized to [0 1], as follows:

$$Z = \begin{bmatrix} z_{11} & z_{12} & \cdots & z_{1n} \\ z_{21} & \cdots & \cdots & \vdots \\ \vdots & \cdots & \cdots & \vdots \\ z_{m1} & \cdots & \cdots & z_{mn} \end{bmatrix}$$

$$z_{ij} = \frac{x_{ij} - \min x_j}{\max x_j - \min x_j}$$

We then create the transposed matrix Z^T :

$$Z^T = \begin{bmatrix} z_{11} & z_{21} & \cdots & z_{m1} \\ z_{12} & \cdots & \cdots & \vdots \\ \vdots & \cdots & \cdots & \vdots \\ z_{1n} & \cdots & \cdots & z_{mn} \end{bmatrix}$$

We then determine the weights of each indicator. In our case study, the indicators were given equal weight by assigning a value of 1.

$$W = (w_1 \quad w_2 \quad \cdots \quad w_n)^T$$

We then create the comparison matrix A .

$$A = \begin{bmatrix} a_{11} & a_{21} & \cdots & a_{m1} \\ a_{12} & \cdots & \cdots & \vdots \\ \vdots & \cdots & \cdots & \vdots \\ a_{1n} & \cdots & \cdots & a_{mn} \end{bmatrix}$$

$$= \begin{bmatrix} w_1 a_{11} & w_1 a_{21} & \cdots & w_1 a_{m1} \\ w_2 a_{12} & \cdots & \cdots & \vdots \\ \vdots & \cdots & \cdots & \vdots \\ w_n a_{1n} & \cdots & \cdots & w_n a_{mn} \end{bmatrix}$$

Ideal point:

$$P^* = \max_j \{a_{ij} \quad i = 1, 2, \dots, m\} \\ = (p_1^* \quad p_2^* \quad \cdots \quad p_n^*)^T$$

We can then calculate the d_i and T_i values, as follows:

$$d_i = \sqrt{\sum_{j=1}^n (a_{ij} - p_j^*)^2} \quad i = 1, 2, \dots, m$$

$$T_i = 1 - \frac{\sum_{j=1}^n a_{ij} p_j^*}{\sum_{j=1}^n (p_j^*)^2} \quad i = 1, 2, \dots, m$$

Consequently, the chemicals are ranked according to T_i . The lower the T_i value, the more accurate the ranking. When the T_i are equal in size, then we can use d_i to rank, the lower the more accurate the ranking [7].

Case Study

A total of 79 chemicals were selected for screening; all these chemicals were requested by the 2008 Commonwealth and

Environmental Protection Project of the Ministry of Environmental Protection of the People's Republic of China (MEP): "Bioconcentration of Toxic Hazardous Substances in body adipose tissues and risk analysis on human health.. Sample collection and detection were performed in previous. The human exposure concentration and detection frequency data were reported in two articles [11,12]. Human exposure concentration experiments show that eight chemicals were not detected in human adipose tissue samples. Therefore, these eight chemicals are not potential hazards. Thus, at first screening, these eight chemicals can be ignored. The remaining chemicals were then used in the subsequent experiments.

Table 1 presents the toxicological and exposure endpoints used in this article. Four human health effects, two environmental effects, octanol-water partition, bioaccumulation, human exposure concentration, and frequency of detection are included. Human health effects and environmental effects are important indicators that reflect the hazards or toxicities of chemicals; these effects are indispensable in risk assessment. Octanol-water partition is also an important parameter. Octanol is a long-chain alcohol that can reflect the transmission and distribution capacity of organisms.

The experimental data were obtained from the Hazardous Substances Data Bank, Pesticide Properties Database, U.S EPA Aggregated Computational Toxicology Resource, and U.S EPA ECOTOX Database whenever possible. Structure-activity relationships (SARs) and quantitative structure-activity relationships (QSARs) such as EPI Suite™ v3.20 and ECOSAR™ v 1.00 were used to estimate missing data. This estimation depends on the availability of reliable SARs or QSARs. If an SAR or QSARs was not available, the missing data were decided through expert judgment. The human exposure concentration and detection frequency data were quoted from previously published two articles.

Toxicological values such as rodent oral LD50, fish LC50, and bird LD50 have negative correlations with the hazards of human health. To simplify the comparison calculations, we used the negative number of the values. Moreover, the octanol-water partition (Kow) and bioconcentration factor (BCF) values were sometimes too large to calculate. Thus, we used log Kow and log BCF. The properties of each chronic effect were divided into three classes: recognized, suspected, and not likely. These parameters were qualitative and could not be calculated using formulas. For this reason, the parameters were

assigned the quantitative values of 5, 3, and 0, respectively. At this step, the original dataset was established.

To apply the Copeland method and the multi-index comparison method, programs were written using the MATLAB mathematical software.

After the original data set was constructed, the Copeland and multi-index comparison methods were used to determine the ranking order. The results are listed in Table 2.

We calculated the correlation coefficient of these two methods from the results. The correlation coefficient can explain the similarities between the ranking orders. If the correlation coefficient is close to 1, the two ranking orders are highly similar. However, the correlation coefficients of the Copeland and multi-index comparison methods are 0.8424, the two ranking orders are not much similar. Why is there a big difference between the Copeland method and the multi-index comparison method? The principles of these two methods can provide some explanations. The Copeland method only focuses on the numerical magnitude between two values. Therefore, the results of 1<100 and 1<1000 are equivalent. On the other hand, in the multi-index comparison method, the numerical magnitude and numerical distribution significantly affect the result. Calculation of the ideal point is a key step in this method. The ideal point is directly related to the maximum values of each indicator. When a value is considerably bigger than the others, a lower T value may be obtained but may have a smaller effect on the Copeland method.

Value pretreatment was performed to improve the results of these two methods. Each indicator to the oral LD50, fish LC50, and bird LD50 toxicity terms can range from zero to five. A cutoff value set for each indicator so that the hazard value for very high or low toxicities would not exceed five or be below zero.

The hazard value for the acute oral toxicity (HVOR) was based on the oral LD50 and was calculated using a continuous, linear function, with the cutoff values at 5,000 and 5 mg/kg:

$$HV_{OR} = 6.165 - 1.667 \log(\text{oral LD50}) \text{ for } 5 \text{ mg/kg} < \text{oral LD50} \leq 5,000 \text{ mg/kg}$$

$$HV_{OR} = 0 \text{ for oral LD50} > 5,000 \text{ mg/kg}$$

$$HV_{OR} = 5 \text{ for oral LD50} \leq 5 \text{ mg/kg}$$

Type	Endpoint	Definition
Human health effects		
Acute	Rodent oral LD50	The mass of the substance administered per unit mass of the test subject that will kill half of the test subjects within 14 days when orally administered as a single dose.
Chronic	Carcinogenicity	Based on supporting evidence.
Chronic	Reproductive and Developmental Toxicity	
Chronic	Endocrine Toxicity	
Environmental effects		
Aquatic, acute	Fish LC50	The concentration of a substance in water that will cause 50% of fish deaths in the 96 h test.
Terrestrial, acute	Bird LD50	The mass of the substance administered per unit mass of the test subject that will kill half of the test subjects within 14 days when orally administered as a single dose.
Exposure potential		
Partition	Octanol-water Partition (Kow)	The ratio of the distribution of a substance between octanol and water.
Bioaccumulation	Bioconcentration factor (BCF)	The ratio of the concentration of a chemical in a biological tissue to that in the water surrounding that tissue.
	Human exposure concentration	The concentration of a chemical in human adipose tissues.
	Frequency of detection	Frequency of detection in human adipose tissue samples.

Table 1: Toxicological and exposure endpoints.

NO.	Chemical	Copeland method		Multi-index comparison	
		Original data	Pretreatment data	Original data	Pretreatment data
1	2,4'- DDD	19	19	25	15
2	2,4'- DDE	13	13	12	12
3	2,4'- DDT	1	1	1	1
4	4,4'- DDD	10	10	9	11
5	4,4'- DDE	3	3	6	9
6	4,4'- DDT	2	2	3	2
7	alpha-Hexachlorocyclohexane	35	35	26	28
8	beta-Hexachlorocyclohexane	8	9	2	6
9	gamma-Hexachlorocyclohexane	24	23	32	21
10	Lindane	44	44	33	45
11	Hexachlorobenzene	7	6	4	3
12	Mirex	18	18	14	18
13	Aldrin	9	8	10	8
14	Endrin	5	5	15	5
15	Heptachlor	12	11	8	10
16	Chlordane	36	37	35	47
17	Chlordane	26	26	31	41
18	Acenaphthylene	63	63	67	64
19	Acenaphthene	58	58	49	58
20	Anthracene	49	46	46	48
21	1,2-Benzanthracene	25	22	29	23
22	Benzo[a]pyrene	6	7	7	7
23	Benzo[b]fluorathene	38	38	30	31
24	Benzo(ghi)perylene	29	27	39	40
25	Benzo[k]fluorathene	11	12	17	13
26	Dibenz[a,h]anthracene	23	25	21	25
27	Fluoranthene	37	36	59	37
28	Fluorene	53	53	41	57
29	Indeno[1,2,3-cd]Pyrene	16	15	18	14
30	naphthalene	32	32	11	24
31	Phenanthrene	22	24	23	32
32	Pyrene	40	41	42	46
33	Butyl benzyl phthalate	52	52	28	43
34		59	59	38	59
35	Dibutyl phthalate	30	30	27	30
36	Dicofol	14	14	13	17
37	Methamidophos	51	51	65	56
38	Chlordimeform	56	56	34	50
39	Acetamiprid	67	67	70	67
40	Alachlor	43	42	22	35
41	Amitraz	45	45	20	39
42	Buprofezin	60	60	58	60
43	Machette	61	61	61	62
44	Carbofuran	57	57	63	55
45	Chlorothalonil	42	43	24	29
46	Clorpyrifos	17	16	36	16
47	Clomazone	71	71	68	71
48	Cyfluthrin	46	48	54	44
49	Cypermethrin	28	28	45	22
50	Deltamethrin	48	49	56	51
51	Diazinon	55	54	50	52
52	Thiosulfan I	34	34	48	38
53	Thiosulfan II	31	31	47	36
54	Mocap	21	20	37	20
55	Phenvalerate	41	39	57	34
56	Esfenvalerate	15	17	40	26
57	Hexythiazox	64	65	44	63
58	Isoproturon	68	68	64	68
59	Cyhalothrin	39	40	53	42

60	Metolachlor	62	62	52	61
61	Nitfen	33	33	16	27
62	O,O-Dimethyl-S-methylcarbamoymethyl phosphorothioate	65	64	66	65
63	Oxyfluorfen	47	47	43	49
64	Parathion-methyl	50	50	51	53
65	Pirimicarb	66	66	71	66
66	Prometryn	70	70	62	69
67	Pyridaben	54	55	60	54
68	Triazophos	27	29	55	33
69	Tricyclazole	69	69	69	70
70	Trifluralin	20	21	19	19
71	Chlorobiphenyl	4	4	5	4

Table 2: Ranking order of the Copeland method and the multi-index comparison method after data pretreatment.

	R
Copeland method on original data vs. pretreatment data	0.9989
multi-index comparison of original data vs. multi-index comparison of pretreatment data	0.8784
Copeland method on original data vs. multi-index comparison of original data	0.8424
Copeland method on original data vs. multi-index comparison of pretreatment data	0.9718
Copeland method on pretreatment data vs. multi-index comparison of original data	0.8419
Copeland method on pretreatment data vs. multi-index comparison of pretreatment data	0.9736

Table 3: Correlation coefficients of the Copeland method and the multi-index comparison method.

The hazard value for the acute aquatic toxicity (HV_{FA}) was based on the acute fish LC50 and was calculated using a continuous, linear function, with the cutoff values at 100 and 0.01 mg/l:

$$HV_{FA} = 2.5 - 1.25 \log(LC50) \text{ for } 0.01 \text{ mg/l} < LC50 \leq 100 \text{ mg/l}$$

$$HV_{FA} = 0 \text{ for } LC50 > 100 \text{ mg/l}$$

$$HV_{FA} = 5 \text{ for } LC50 \leq 0.01 \text{ mg/l}$$

The hazard value for the acute bird toxicity (HV_{BA}) was based on the acute bird LD50 and was calculated using a continuous, linear function, with the cutoff values at 5,000 and 5 mg/kg:

$$HV_{BA} = 6.165 - 1.667 \log(\text{bird LD50}) \text{ for } 5 \text{ mg/kg} < \text{bird LD50} \leq 5,000 \text{ mg/kg}$$

$$HV_{BA} = 0 \text{ for } \text{bird LD50} > 5,000 \text{ mg/kg}$$

$$HV_{BA} = 5 \text{ for } \text{bird LD50} \leq 5 \text{ mg/kg}$$

Kow and BCF can range from one to five. Cutoff values were also set for the indicators so that the hazard value would not exceed five or be less than one. The Kow hazard value (HV_{Kow}) was calculated using

$$HV_{Kow} = 0.6667 \log Kow + 0.3333 \text{ for } 1 < \log Kow \leq 7$$

$$HV_{Kow} = 1 \text{ for } \log Kow \leq 1 \text{ and } \text{for } LD50 > 7$$

The BCF hazard value (HV_{BCF}) was calculated using

$$HV_{BCF} = 1.3333 \log BCF - 0.3333 \text{ for } 1 < \log Kow \leq 4$$

$$HV_{BCF} = 1 \text{ for } \log Kow \leq 1 \text{ and } \text{for } LD50 > 4$$

The hazard value of the frequency of detection (HV_{FD}) was also calculated using continuous, linear functions, with the cutoff values at 0.001 and 1:

$$HV_{FD} = \log(FD) + 3 \text{ for } 0.001 < FD < 1 \text{ and } \text{for } FD \leq 0.001$$

On the other hand, the hazard value of human exposure concentration (HV_{HEC}) was calculated using continuous, linear functions without cutoff values.

$$HV_{HEC} = \log(HEC)$$

The carcinogenicity, reproductive and developmental toxicity, and endocrine toxicity data did not need pretreatment; the values used in this step are the same as those in the original data.

After data pretreatment, a new dataset was created. The Copeland method and the multi-index comparison method were then used.

Table 2 shows the ranking order of the Copeland method and the multi-index comparison method after data pretreatment, as well as the ranking order of the Copeland method using original data. The correlation coefficients were calculated and are listed in Table 3. The correlation coefficient between the original data and pretreatment data was much higher when the Copeland method was used, indicating that the two results have a high degree of similarity (Figure 1). The correlation coefficient between the original data and pretreatment data in multi-index comparison method is 0.8784, it seem that the pretreatment of the data have much influence on the ranking result. Besides, the correlation coefficient between the multi-index comparison with original data and Copeland method are all about 0.84. While the correlation coefficient between the multi-index comparison with pretreatment data and Copeland method are all about 0.97, which also indicated that the pretreatment of the data have significant influence on the ranking result.

Although the correlation coefficients between the Copeland method and the multi-index comparison method reached 0.97, the ranking orders were not very similar, particularly the middle part of the rank orders (Figure 2). However, the top and last parts of the rank order are highly similar. This similarity is sufficient in identifying the hazardous chemicals.

As a result, the Copeland method seems much more convenient, it can give a good ranking result without data pretreatment. The multi-index comparison method is not very good at deal with original data, but it also can give a good ranking result with data pretreatment.

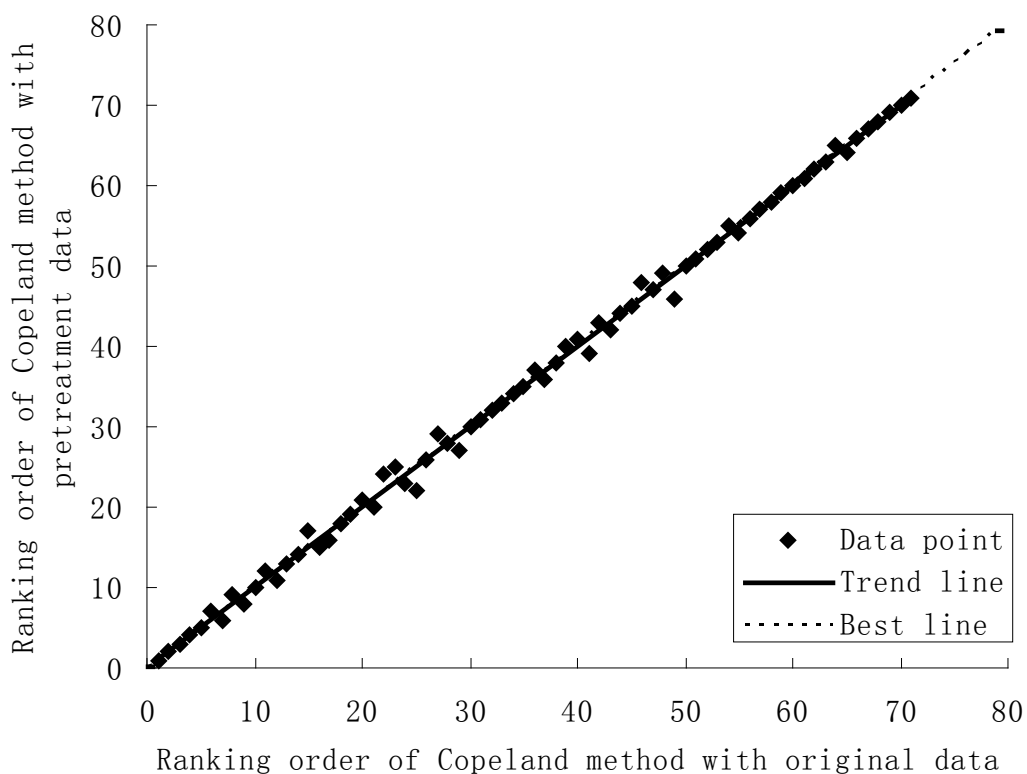


Figure 1: Relationship between the Copeland method results using original data and pretreatment data.

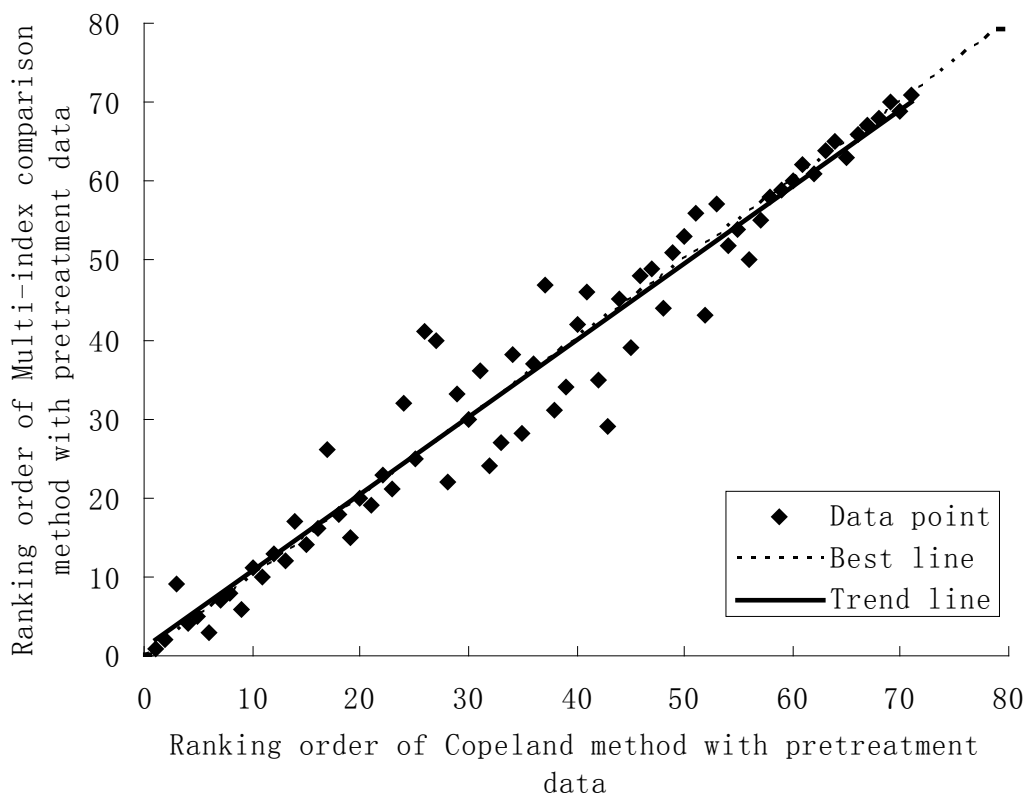


Figure 2: Relationship between the Copeland method and the multi-index comparison method results using pretreatment data.

Rank	Copeland method		Multi-index comparison	
	Original data	Pretreatment data	Original data	Pretreatment data
1	2,4'- DDT	2,4'- DDT	2,4'- DDT	2,4'- DDT
2	4,4'- DDT	4,4'- DDT	beta-Hexachlorocyclohexane	4,4'- DDT
3	4,4'- DDE	4,4'- DDE	4,4'- DDT	Hexachlorobenzene
4	Chlorobiphenyl	Chlorobiphenyl	Hexachlorobenzene	Chlorobiphenyl
5	Endrin	Endrin	Chlorobiphenyl	Endrin
6	Benzo[a]pyrene	Hexachlorobenzene	4,4'- DDE	beta-Hexachlorocyclohexane
7	Hexachlorobenzene	Benzo[a]pyrene	Benzo[a]pyrene	Benzo[a]pyrene
8	beta-Hexachlorocyclohexane	Aldrin	Heptachlor	Aldrin
9	Aldrin	beta-Hexachlorocyclohexane	4,4'- DDD	4,4'- DDE
10	4,4'- DDD	4,4'- DDD	Aldrin	Heptachlor
11	Benzo[k]fluorathene	Heptachlor	naphthalene	4,4'- DDD
12	Heptachlor	Benzo[k]fluorathene	2,4'- DDE	2,4'- DDE
13	2,4'- DDE	2,4'- DDE	Dicofol	Benzo[k]fluorathene
14	Dicofol	Dicofol	Mirex	Indeno[1,2,3-cd]Pyrene
15	Esfenvalerate	Indeno[1,2,3-cd]Pyrene	Endrin	2,4'- DDD
16	Indeno[1,2,3-cd]Pyrene	Clorpyrifos	Nitfen	Clorpyrifos
17	Clorpyrifos	Esfenvalerate	Benzo[k]fluorathene	Dicofol
18	Mirex	Mirex	Indeno[1,2,3-cd]Pyrene	Mirex
19	2,4'- DDD	2,4'- DDD	Trifluralin	Trifluralin
20	Trifluralin	Mocap	Amitraz	Mocap

Table 4: Top 20 Hazardous Chemicals.

Discussion

The top 20 ranked chemicals are presented in Table 4. In the top 20, the three ranking results share 18 common chemicals. These chemicals were then classified into four general groups: six kinds of dichlorodiphenyltrichloroethane (DDTs), seven kinds of other POPs, three kinds of polycyclic aromatic hydrocarbons, and two kinds of pesticides. These substances should be regarded as chemicals of concern for human health, and appropriate management should be taken. From the result, the ranking of 2,4'-DDT, 4,4'-DDT, 4,4'-DDE, chlorobiphenyl, endrin, benzo[a]pyrene, hexachloro cyclohexane, and aldrin were relatively high because of their chronic effects and high human exposure concentrations. Some of these pesticides are still being used in some regions of China. Thus, the use of these products may increase the concern on the potential health hazards to humans.

The results of this study show that the Copeland method is a simple and effective ranking and screening method. The ranking order of the Copeland method using original data can rationally explain the hazard relationships between 71 chemicals. In our study, the indicators of human exposure concentration and detection frequency are more significant for ranking results. The Copeland method can be easily performed using software, thus making the ranking, screening, and assessing of hazardous chemicals more convenient. If necessary, expert judgment can be used to add weight to the indicators.

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