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AN EQUATION FOR ASSESSING INTERACTION AMONG COMPONENTS IN COMBINED POLLUTIONS

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Abstract : In order to assess interaction among multiple pollutants, over-content ratio $r_j = x_j/A_j$ and equation $\sum r_j^{1/\mu} = r_0^{1/\mu}$ are proposed, where x_j and A_j are content of Pollutant j in a polluted material and the control value of the pollutant, respectively, and μ and r_0 are positive constants. The interaction is expressed by μ (say interaction index) as follows: independent (i.e. no) at $\mu \rightarrow 0$, somewhat independent and somewhat complementary (or negative) at $0 < \mu < 1$, completely complementary at $\mu = 1$, and stronger than the complementary case (or positive) at $\mu > 1$. The control value of dioxins is given on the assumption that the interaction among chemicals belonging to the dioxins is completely complementary. In lung cancer relating to asbestos exposure and cigarette smoking, the death rate is given by $r = \{r_A^{1/3} + r_C^{1/3}\}^3$, where r_A and r_C are over-content ratios (= death rates in this example) of asbestos and cigarette, respectively. This equation shows that the death rate of both asbestos and cigarette (ca. 600) is 12 times of the death rate of only asbestos (ca. 50), and 6 times of the death rate of only cigarette (ca. 100). The equation suggests that environmental assessments may cause risks that hazardous cases are assessed to be safe or that safe cases are assessed to be hazardous, if control values are given not taking account of interaction among pollutants. When an interaction index indicating degree of combined pollution is not known, it may be the most adequate that the interaction is assumed to be completely complementary.

Key words : Environment, Combined pollution, Interaction, Over-content ratio, Mathematics

1. Introduction

The present society bears many kinds of pollutions called as “air pollution”, “water pollution”, “soil pollution”, and so on. The global warming caused by increasing greenhouse gases is also a kind of pollution. In order to control pollutions, many laws, ordinances and agreements have been established, and give a control value for each pollutant. For example, control values in Japan are 0.06 ppm (hourly average) for photochemical oxidants in air, 25 mg/l for suspended solids in drinking water, 0.01 g/l for copper in rice field soil, and so on. Mostly, such control values are given independently of the other components. In contrast, a weighted sum of greenhouse gasses is applied for the assessment of global warming. Weighted sums are also applied for assessing air pollution, and called as “combined pollution index” by Babcock (1970) or “ambient air quality” by Wheeler (2004). Ott (1978) proposed a conceptual framework to aggregate environmental indices of multiple pollutants, and presented several functions including not only the linear sum but also the nonlinear sum and the root-sum-power in the framework (see Section 4.2 in detail).

A control value for a pollutant is determined on the basis of experiences and/or experiments, which show health risks caused

by pollutants. If it is possible to ignore interaction between a pollutant and any other pollutant, it is adequate to give the control value of the pollutant independently of the other pollutants. If a pollutant is complementary to another pollutant, the weighted sum method seems to be available for assessing the risk caused by these two pollutants. We have two questions here. The one is what pairs of pollutants are independent, and what pairs of pollutants are complementary. The other is if a relation between two pollutants is only independent and complementary. In other words, does not any pair show interaction of another type? For example, some pairs may show interaction represented by the root-sum-power. This is very important for assessing combined pollutions.

So many experiences and experiments are necessary to answer the questions. Approximately speaking, numbers of necessary data increase geometrically with increasing numbers of components, when a phase diagram is drawn. This trend seems to be same in the case of risk assessment of pollutions. It is expected, however, to reduce numbers of experiments, if a relation among pollutants can be represented by a mathematical formula. This paper proposes an equation representing relations among pollutants, and shows that the equation indicates degree

of the interaction among them.

The fundamental concept of the theme stated in this paper was already published briefly (Shoji, 2005a, b). This paper states the concept in detail, and also presents some examples based on the concept.

2. Interaction among Pollutants

2.1. Over-content Ratio

In order to assess mineral resources of polymetallic deposits, Shoji (2002) introduced a concept of enrichment ratio. The enrichment ratio (e_j) of Metal j in a deposit is given by the following equation:

$$e_j = x_j / A_j \quad (1)$$

where x_j and A_j are grade of the metal in a deposit, and crustal abundance of the metal, respectively. If a deposit consists of Metal 1 and Metal 2, the enrichment ratios of the metals are $e_1 = x_1 / A_1$ and $e_2 = x_2 / A_2$. The enrichment ratio of Metal 2 is also written as $e_2 = (x_2 / a_{2/1}) / A_1$, where $a_{2/1} = A_2 / A_1$. This means that $x_2 / a_{2/1}$ is the grade of Metal 2 normalized by Metal 1. Therefore, the grade summarizing Metals 1 and 2 is defined as $(x_1 + x_2 / a_{2/1})$ when Metal 1 is the norm. In this case, the total enrichment ratio (e_{1+2}) is given as $e_{1+2} = (x_1 + x_2 / a_{2/1}) / A_1 = e_1 + e_2$. Therefore, if a deposit consists of m kinds of metals, the total enrichment ratio (e_Σ) is given as follows:

$$e_\Sigma = \sum_{j=1}^m e_j \quad (2)$$

In order to assess a pollution caused by multiple components, let us consider x_j and A_j of Equation (1) content and control value of Pollutant j , respectively. According to the change, let us replace e_j and e_Σ by r_j and r ($= r_\Sigma$), respectively, and say r_j and r “over-content ratio” and “total over-content ratio”, respectively. When a polluted material contains Pollutants 1 and 2, Equation (2) is written as follows:

$$r = r_1 + r_2 \quad (3)$$

$$= x_1 / A_1 + x_2 / A_2 \quad (3')$$

Let us expand Equation (3) as follows:

$$r^{1/\mu} = r_1^{1/\mu} + r_2^{1/\mu} \quad (4)$$

$$r = \{r_1^{1/\mu} + r_2^{1/\mu}\}^\mu \quad (4')$$

$$= \{(x_1 / A_1)^{1/\mu} + (x_2 / A_2)^{1/\mu}\}^\mu.$$

where μ is a positive constant. Equation (3) is obtained when $1/\mu = 1$ (i.e. $\mu = 1$) in Equation (4).

Equation (4') gives a curved surface in a 3D space consisting of r_1 , r_2 and r . Therefore, the $r_1 - r_2$ relation at $r = r_0$ (i.e. the contour line of r_0) is given by the following equation:

$$r_1^{1/\mu} + r_2^{1/\mu} = r_0^{1/\mu} \quad (5)$$

Coordinates of each point (r_1 , r_2) on a curve expressed by Equation (5) are given as follows:

$$r_1 = \rho \cos \alpha_1$$

$$r_2 = \rho \cos \alpha_2$$

where ρ is the distance from the origin to the point, and $\cos \alpha_1$ and $\cos \alpha_2$ are direction cosines (i. e. $\cos^2 \alpha_1 + \cos^2 \alpha_2 = 1$) of the point. Therefore, Equation (5) is written as follows:

$$\rho^{1/\mu} \cos^{1/\mu} \alpha_1 + \rho^{1/\mu} \cos^{1/\mu} \alpha_2 = r_0^{1/\mu} \quad (6)$$

The above equation gives the following equation:

$$\rho = \frac{r_0}{(\cos^{1/\mu} \alpha_1 + \cos^{1/\mu} \alpha_2)^\mu} \quad (7)$$

Dotted lines in Figure 1 show relations given by Equation (5) under the conditions where $1/\mu$ is 1 (i.e. $\mu = 1$), and $r_0^{1/\mu}$ ($= r_0$) is 0.5, 1, 1.5 and 2. On the other hand, solid curves are obtained under the conditions where μ is 0.001 (nearly 0), 0.2, 0.5, 1, 2 and 4 ($1/\mu$ are 1000, 5, 2, 1, 0.5 is 0.25, respectively), and $r_0^{1/\mu}$ is 1 (i.e. $r_0 = 1$).

2.2. Interaction between Two Pollutants

Before discussing on interaction between two pollutants, let us consider meaning of Equation (5) in a system containing a single pollutant. If $r_0 = 1$, then $r_1 = 1$ (i.e. $x_1 = A_1$) independently of μ . This means that the content of Pollutant 1 equals to the control value of the pollutant. On the other hand, the content of Pollutant 1 is lower than the control value if $r_1 < 1$ ($x_1 < A_1$), while the content is higher than the control value if $r_1 > 1$ ($x_1 > A_1$). In other words, the content of Pollutant 1 is lower than the control value if the over-content ratio is plotted on the left side of 1 in Figure 1, and that the content of Pollutant 1 is higher than the control value if the ratio is plotted on the right side of 1.

Next, let us consider a case where a control value of each component has been determined independently of the other components. If risk caused by Pollutant 1 is independent on that of Pollutant 2 in a binary system, the safe/hazardous boundary, which is the boundary between the contents being higher and lower than the control values, are shown by a line passing through Points (0,1), (1,1) and (1,0) in Figure 1, and the inside of the boundary is assessed to be safe. The boundary line is obtained when $r_0 = 1$, and $\mu \rightarrow 0$ (i.e. $1/\mu \rightarrow \infty$) in Equation (5), because under this mathematical condition, r_2 is almost 1 when r_1 is between 0 and 1, and r_2 changes from 1 to 0 when r_1 is almost 1 but increases. Let us say this case to be completely independent.

The condition where $r_0^{1/\mu} = 1$ and $\mu = 1$ in Equation (5) gives the straight line connecting Points (0,1) and (1,0) in Figure 1. This means that a unit of the over-content ratio of Pollutant 2

is equal to that of Pollutant 1. In contrary, if it can be assumed that a unit of the over-content ratio of Pollutant 2 equals to that of Pollutant 1, the safe/hazardous boundary is given by the following equation:

$$r_1 + r_2 = 1$$

Let us say this case to be completely complementary.

The condition where $r_0^{1/\mu} = 1$ and $0 < \mu < 1$ in Equation (5) gives curves which are concave toward the origin in Figure 1. A group of curves being concave toward the origin represent cases where Pollutants 1 and 2 are not completely independent and not completely complementary, because the components are completely independent at $\mu \rightarrow 0$, and completely complementary at $\mu = 1$ as mentioned above. This means that values of μ indicate the degree of independency/complementation between Pollutants 1 and 2. For example, when $\mu = 1/2$ in Equation (5), the interaction between two pollutants is not independent and not complementary, and the safe/hazardous boundary in the system is represented by a circle as $r_1^2 + r_2^2 = r_0^2$. This case may indicate that the risk is proportional to the distance from the origin in the $r_1 - r_2$ space.

The condition where $r_0^{1/\mu} = 1$ and $\mu > 1$ in Equation (5) gives curves which are convex toward the origin in Figure 1. In the cases, interaction between Pollutants 1 and 2 is stronger than that in the completely complementary case.

Let us say μ interaction index, because μ of Equation (5) can indicate degree of interaction between two components. The interaction index classifies degree of interaction into four cases: i) completely independent if $\mu \rightarrow 0$, ii) weak (between independency and complementation) if $0 < \mu < 1$, iii) completely

complementary if $\mu = 1$, and iv) strong (higher than complementation) if $\mu > 1$. If we can determine the index, we can show degree of interaction between two pollutants quantitatively. If we consider the completely complementary case is standard, then we can say Cases ii and iv show negative and positive interactions, respectively. If we use this definition, sign of the interaction is given by $\log \mu$. This is the reason why the exponent of Equation (4) is not μ but $1/\mu$.

The relation given by Equation (4) is represented in a r_1 - r_2 - r space where Axes r_1 and r_2 are horizontal, and Axis r is vertical. In the space a surface represented by Equation (4) is convex upwards or downwards, when the exponent ($1/\mu$) is lower or higher than 1, respectively [1]. In other words, contour lines are convex and concave toward the origin at $\mu > 1$ and $\mu < 1$, respectively. It is adequate to say that if a surface is convex upward then it represents a positive interaction, and that if a surface is convex downward it represents a negative interaction.

2.3. Case Studies

Unfortunately, we have no example showing Equation (4) directly. It is possible, however, to explain a mathematical reason for giving control values of dioxins on the basis of the equation. The equation is also applicable to a case of lung cancer relating to asbestos exposure and cigarette smoking, though the control values are not given, but death rates are given.

Dioxins and furans are some of the most toxic chemicals. Initially "dioxin" was an abbreviation for a single substance, with the formidable chemical name, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD). Presently it is one member of a family of chemical compounds with an identical carbon-oxygen framework. Chlorine atoms (four in this case) are attached at specific carbon atom sites as indicated by the numbers 2,3,7,8 in the name. "Dioxins" refers to a group of several hundred of chemical compounds which are members of three closely related families: the chlorinated dibenzo-*p*-dioxins (CDDs), chlorinated dibenzofurans (CDFs) and certain polychlorinated biphenyls (PCBs). In Japan, the environmental quality standards of dioxins are 0.6 pg-TEQ/m³ in air, 1 pg-TEQ/l in water, 150 pg-TEQ/g in bottom sediment, and 1000 pg-TEQ/g in soil, where a TEQ is a dioxin toxic equivalent, calculated by looking at all toxic dioxins and furans and measuring them in terms of the most toxic form of dioxin, 2,3,7,8-TCDD. This means, therefore, that the risk caused by dioxins is assessed by a weighted sum of chemicals belonging to dioxins. It is concluded, hence, that the quality standard is given on the assumption that the interaction among the chemicals is completely complementary. Since the statement in this part treats many chemicals belonging to dioxins, exactly speaking, it is based on Equation (11) given in Section 3.2.

It is well known that each of asbestos exposure and cigarette smoking is a course of lung cancer, and that the combina-

Figure 1. Relations of over-content ratios (r_1 and r_2) of two pollutants. Dotted lines show the cases where $1/\mu = 1$ ($\mu = 1$), and $r_0^{1/\mu} = r_0$ is 0.5, 1, 1.5 and 2 in Equation (5) from the origin outwards. Solid curves show the cases that $r_0^{1/\mu} = 1$ (i.e. $r_0 = 1$) and μ is 0.001 (nearly 0), 0.2, 0.5, 1, 2 and 4 ($1/\mu$ is 1000, 5, 2, 1, 0.5 and 0.25, respectively) from the outside to the origin.

Table 1. Death rates of lung cancer relating to asbestos and cigarette (Hammond et al., 1979).

		Smoking	
		No	Yes
Asbestos Workers	No	11.3 (0)	122.6 (111.3)
	Yes	58.4 (47.1)	601.6 (590.3)

Values with parentheses are death rates after reducing the background value, which is considered to be represented by the value in the case of no asbestos and no smoking.

tion of both courses accelerate to develop the cancer (e. g. Hammond et al., 1979). Table 1 shows death rates of lung cancer relating to asbestos and cigarette. In order to apply the concept of the over-content ratio to the case, we need the following assumptions: 1) death rates relating to each of asbestos and cigarette increase linearly with the degree of asbestos exposure or cigarette smoking (exactly speaking, the assumption is enough if death rates increase monotonically with the degree, because the relation between death rates and the degree can be transformed to a linear equation in the case), and 2) A_j (j denotes asbestos or cigarette) in Equation (4') represents the degree at which the death rate is 1. These assumptions make over-content ratio equivalent to death rate. We need more three assumptions because of the data presented by Hammond et al. (1979) are limited: 3) every asbestos worker was exposed under a condition of an equal degree, 4) every smoker smoked cigarettes of an equal degree, and 5) the death rate in the case of no asbestos and no cigarette represents the background. At the first step, the assumptions give the death rates relating really to asbestos and cigarette as the values with parentheses in Table 1. At the second step, the assumptions give the following simultaneous equations:

Figure 2. Relation among death rates (parameter r) and over-content ratios of asbestos (horizontal axis) and cigarette (vertical axis). In this case, $\mu = 3$ ($1/\mu = 1/3$).

$$r_{\text{Asbestos}}^{1/\mu} + (0)^{1/\mu} = (111.3)^{1/\mu}$$

$$(0)^{1/\mu} + r_{\text{Cigarette}}^{1/\mu} = (47.1)^{1/\mu}$$

$$r_{\text{Asbestos}}^{1/\mu} + r_{\text{Cigarette}}^{1/\mu} = (590.3)^{1/\mu}$$

These equations are satisfied when $1/\mu \approx 1/3$ (exactly $1/\mu = 0.335259$).

Figures 2 and 3 show death rates caused by asbestos exposure and cigarette smoking in a linear scale and a log-log scale, respectively. In Figure 2, the line of $r = 50$ starts at Point (0,50) and ends Point (50,0), while the line of $r = 100$ starts at Point (100,0) and ends at Point (0,100). If $r_{\text{Asbestos}} = x_{\text{Asbestos}}/A_{\text{Asbestos}} = 50$, and $r_{\text{Cigarette}} = x_{\text{Cigarette}}/A_{\text{Cigarette}} = 100$, then $r \approx 600$ as shown in Figure 2 (or Figure 3). This means that the death rate of both asbestos and cigarette is 12 times of the death rate of only asbestos exposure, and 6 times of the death rate of only cigarette smoking.

3. Complex Cases

3.1. Asymmetric Case

Equation (5) is symmetric between r_1 and r_2 . It is considered, however, that interaction between two pollutants is not symmetric. In an asymmetric case, Equation (5) is expanded as follows:

$$r^{1/\mu_0} = r_1^{1/\mu_1} + r_2^{1/\mu_2}$$

$$r = \{r_1^{1/\mu_1} + r_2^{1/\mu_2}\}^{\mu_0} \quad (8)$$

Coordinates of each point (r_1, r_2) on a contour of r_0 are given as follows:

$$r_1^{1/\mu_1} + r_2^{1/\mu_2} = r^{1/\mu_0} \quad (9)$$

$$r_2 = (r^{1/\mu_0} - r_1^{1/\mu_1})^{\mu_2}$$

In this case, the equation corresponding to Equation (6) is given

Figure 3. Relation among death rates (parameter r) and over-content ratios of asbestos (horizontal axis) and cigarette (vertical axis) on a log-log diagram.

Figure 4. Relations of over-content ratios (r_1 and r_2) in asymmetrical cases given by Equation (4). The constants of Equation (8) are as follows: $\mu_1 = 1.5$ and $\mu_2 = 0.2$, $\mu_1 = 20$ and $\mu_2 = 0.2$, and $\mu_1 = 5$ and $\mu_2 = 0.7$ at $r_0^{1/\mu} = 1$ (i.e. $r_0 = 1$) from the top downwards.

as follows:

$$\rho^{1/\mu_1} \cos^{1/\mu_1} \alpha_1 + \rho^{1/\mu_2} \cos^{1/\mu_2} \alpha_2 = r_0^{1/\mu_0} \quad (10)$$

Equation (10) does not give ρ as a simple form as Equation (7) [2]. For this reason, the exponent ($1/\mu_0$) of the term r_0 is not important.

Figure 4 shows curves given by Equation (9) in cases where μ_1 and μ_2 are 25 and 0.2, 2 and 0.2, and 10 and 0.5, respectively (i.e. $1/\mu_1$ and $1/\mu_2$ are 0.04 and 5, 0.5 and 5, and 0.1 and 2, respectively). The S-shaped curve ($1/\mu_1 = 0.04$ and $1/\mu_2 = 5$) is the most interesting among the curves. The curve means that the risk caused by the interaction rises rapidly when Pollutant 1 is added into Pollutant 2, while the risk scarcely rises when Pollutant 2 is added into Pollutant 1.

3.2. Interaction among Multiple Pollutants

When a system contains m species of pollutants, Equation (4) is expanded as follows:

$$r = \left\{ \sum_{j=1}^m r_j^{1/\mu_j} \right\}^\mu = \left\{ \sum_{j=1}^m (x_j/A_j)^{1/\mu_j} \right\}^\mu \quad (11)$$

In this system, Equation (5) is written as follows:

$$\sum_{j=1}^m r_j^{1/\mu_j} = r_0^{1/\mu} \quad (12)$$

In order to satisfy Equation (12) in a ternary system, the interaction index should be equal at least in every binary system constituting the ternary system. The interaction in each binary system is expressed as follows:

$$r_1^{1/\mu_{12}} + r_2^{1/\mu_{12}} = r_{012}^{1/\mu_{12}}$$

$$r_1^{1/\mu_{13}} + r_3^{1/\mu_{13}} = r_{013}^{1/\mu_{13}}$$

$$r_2^{1/\mu_{23}} + r_3^{1/\mu_{23}} = r_{023}^{1/\mu_{23}}$$

where the subscripts consisting of two or three figures correspond to the binary systems as shown by the example that subscript 12 and subscript 012 represent the binary system between Components 1 and 2 and the constant in the system, respectively. In the equations, if any of μ_{12} , μ_{13} and μ_{23} is not equal to the others, these three equations cannot be summarized as Equation (12).

If three components are complementary, the following equation is obtained:

$$r_1 + r_2 + r_3 = 1. \quad (13)$$

Equation (13) shows a flat plane crossing at 1 of each of Axes r_1 , r_2 and r_3 . In this case, interaction in each binary system is expressed as a crossing line between the plane and each of the $r_1 - r_2$, $r_1 - r_3$ and $r_2 - r_3$ planes, and then they are complementary as follows:

$$r_1 + r_2 = 1 \quad (\text{be } r_3 = 0 \text{ in } r_1 + r_2 + r_3 = 1) \quad (14)$$

$$r_1 + r_3 = 1$$

$$r_2 + r_3 = 1.$$

The proposition contrary to this is not necessarily satisfied: i.e. even if every binary system is complementary, a ternary system is not always complementary. This is easily understood by considering that Equation (14) is satisfied for $r_1 + r_2 + r_3 + r_1 r_2 r_3 = 1$: the plane given by this equation passes near Point (0.322, 0.322, 0.322), but the plane given by Equation (13) passes near Point (0.333, 0.333, 0.333) [3]. This means that even if any pair of Pollutants 1 and 2, Pollutants 1 and 3 and Pollutants 2 and 3 is complementary, the interaction among Pollutants 1, 2 and 3 is not always complementary.

If pollutants can be divided into several groups on the basis of the interaction index, the total over-content ratio of a group (e.g. Group k) is written as follows:

$$r_k = \left\{ \sum_{j=1}^{m_k} r_{kj}^{1/\mu_k} \right\}^{\mu_k}$$

In the case, therefore, Equation (11) is expanded as follows:

$$r = \left\{ \sum_{k=1}^{m_G} r_k^{1/\mu_G} \right\}^{\mu_G} = \sum_{k=1}^{m_G} \left[\left\{ \sum_{j=1}^{m_k} r_{kj}^{1/\mu_k} \right\}^{\mu_k / \mu_G} \right]^{\mu_G} \quad (15)$$

where m_G and μ_G are number of groups, and interaction index among groups, respectively. If the interaction among groups is independent, $\mu_G \rightarrow 0$ in Equation (15). If the interaction among groups is complementary, $\mu_G = 1$.

4. Discussions

4.1. Simplicity and Complexity of Interaction

If interaction between two pollutants is expressed by Equation (5), the interaction index seems to be easily determined by only a data set including two variables as shown by the example of lung cancer. In practice, however, we need many experiments and/or experiences in order to obtain the data set. This will be easily understood by considering phase diagrams. A phase boundary is given as a point on a temperature-pressure diagram under an isobaric condition. However, we need many experiments at temperatures higher and lower than the point in order to find the point. As clear by this example, simplicity and complexity expected to be seen in interaction among pollutants will be easily understood by comparison with phase diagrams.

A phase diagram has a eutectic point and is simple, if a high form is completely miscible and low forms are completely immiscible in a binary system. On the other hand, if a high form is completely miscible and a low form is also completely miscible, the phase diagram does not have a eutectic point, but it is also simple. We can liken these simple diagrams to the independent or complementary interaction among pollutants. If low forms are partly miscible, we need experiments to draw a phase diagram. If interaction among pollutants is not completely independent and not completely complementary, we have to collect data in order to know the degree of the interaction. When a compound which incongruently melts exists in a binary system, the phase diagram becomes complex. This seems to be similar to an asymmetric interaction.

As stated previously, interaction among Pollutants 1, 2 and 3 is not necessarily complementary, even if any pair of Pollutants 1 and 2, Pollutants 1 and 3 and Pollutants 2 and 3 is complementary. This is similar to the fact that we observe frequently a ternary phase diagram which is different from that expected from the binary diagrams.

Generally, phase diagrams are determined experimentally when a component was added. In practical cases, interaction among pollutants should be also determined experimentally in each system. It is expected, however, that Equation (5) gives a starting point for the experiments. In thermodynamics, a concept of activity is introduced for applying equations of ideal mixing to real mixing. Similarly, a concept of over-activity ratio may be acceptable when Equation (12) is not satisfied.

4.2. Risks Caused by Interaction Models

Ott (1978) presented a conceptual framework that was designed to embrace many environmental indices existing at the time, and discussed on air pollution indices and water pollution indices using the framework. The aggregation functions involving in the framework are $I = \sum I_j$ (linear sum), $I = \sum w_j I_j$ (weighted sum), $I = [\sum I_j^\nu]^{1/\nu}$ (root-sum-power), $I = \max(I_1, \dots, I_j, \dots, I_m)$ (maximum operator), $I = \prod w_j I_j$ (weighted product), and $I = \min(I_1, \dots, I_j, \dots, I_m)$ (minimum operator), where I and I_j are environmental index (“integrated environmental index” according to de Boer et al., 1991) and subindex of Pollutant j , respectively, and $\sum w_j = 1$. Ott (1978) defined the eclipsing region and the ambiguous region in the $I_1 - I_2$ diagram. The eclipsing region is the outside of the given aggregation function, and the inside of the safe/hazardous boundary in the independent case. On the other hand, the ambiguous region is the inside of the given aggregation function, and the outside of the safe/hazardous boundary in the independent case. This definition implies that Ott (1978) considered that a completely independent case was standard, and did not take into account of interaction among pollutants. In practice, he did not give any comment for the interaction. However, the eclipsing and the ambiguity have another meaning as stated in the following paragraphs.

Commonly, multiple pollutants have been assessed by any of two ways as stated previously. The one is that a control value of each pollutant is given independently of the other components, and that the risk is also assessed individually, i.e. regardless of the others. This means that the assessment is based on the independency among pollutants. Accordingly, we can say this way “assessment based on the independency assumption”. The other is that the risk is assessed by the weighted sum of contents of pollutants, where the weight of each pollutant is reciprocal of the control value. “Combined pollution index” of Babcock (1970) and “ambient air quality” of Wheeler (2004) are examples of this. This means that the assessment is based on the complementation among pollutants (say “assessment based on the complementation assumption”).

Let us discuss on the relation between the assumptions and the misjudgments. The misjudgments are classified into two types (Table 2): the first type misjudgment (say alpha misjudgment or alpha type) is that a safe case is assessed to be hazardous, and the second type misjudgment (beta misjudgment or

Table 2. Definition of types of misjudgments in pollution assessment.

		True	
		Safe	Hazardous
Judgment	Safe Hazardous	Alpha Misjudgment (Type I error)	
		Beta Misjudgment (Type II error)

Figure 5. Relations of over-content ratios (r_1 and r_2) in the cases that each curve passes through Point (1,1). The constants of Equation (5) are as follows: $\mu \rightarrow 0$ ($r_0 = 1$), $\mu = 1/2$ ($r_0 = \sqrt{2}$), $\mu = 1$ ($r_0 = 2$) and $\mu = 2$ ($r_0 = 4$) at $r_0^{1/\mu} = 2$ from the origin outwards.

beta type) is that a hazardous case is assessed to be safe.

If Pollutants 1 and 2 are assessed on the basis of control values given under the independency assumption, Figure 1 gives the conclusion that the inner side of the curve given by $\mu \rightarrow 0$ is safe. If the interaction between Pollutants 1 and 2 is not independent, e.g. complementary ($\mu = 1$), however, the safe condition is represented by the inside of a curve determined by a values of interaction index (μ) in Figure 1 (straight line if $\mu = 1$). In this case, the area surrounded by the two curves determined by $\mu \rightarrow 0$ and $\mu > 0$, which corresponds to the Ott's (1978) ambiguous region, is practically hazardous, but is assessed to be safe. The misjudgment is always the beta type under the independency assumption.

When Pollutants 1 and 2 are assessed on the basis of control values given under the complementation assumption, Figure 1 gives the conclusion that the origin side of the line given by $\mu = 1$ is safe. If the real interaction between Pollutants 1 and 2 is negative (or weak: $0 < \mu < 1$), however, the safe condition is represented by the inside of a curve concave toward the origin in Figure 1. In this case, the area surrounded by the two curves determined by $\mu = 1$ and $0 < \mu < 1$ is practically safe, but is assessed to be hazardous. This misjudgment is the alpha type. In contrast, if the interaction between Pollutants 1 and 2 is positive (or strong: $\mu > 1$), the safe condition is represented by the inside of a curve convex toward the origin in Figure 1. In this case, the area surrounded by the two curves determined by $\mu = 1$ and $\mu > 1$ is practically hazardous, but is assessed to be safe. This misjudgment is the beta type.

If a binary system is symmetric, the safest judgment in a pollution assessment is realized under the condition where a sufficiently large value is applied as the interaction index, and con-

trol values are given as the safe/hazardous boundary passes through Point (1,1). Figure 5 shows four safe/hazardous boundaries in cases where $\mu \rightarrow 0$ ($r_0 \rightarrow 1$), $\mu = 1/2$ ($r_0 = \sqrt{2}$), $\mu = 1$ ($r_0 = 2$) and $\mu = 2$ ($r_0 = 4$) from the origin outwards. All of them pass through Point (1,1). If the safe/hazardous boundary passes through Point (1,1), always $r_0^{1/\mu} = 2$ (because, if $r_1 = 1$ and $r_2 = 1$, then $r_1^{1/\mu} + r_2^{1/\mu} = 1 + 1 = r_0^{1/\mu}$). If a system includes m kinds of pollutants, the safest judgment is realized under the condition where control values are given as the safe/hazardous boundary passes through Point (1, ..., 1) in a m -dimensional space. In a system consisting of m kinds of pollutants, $r_0^{1/\mu} = m$ (i.e. $r_0 = m^\mu$).

Each curve shown in Figure 5 gives the safest judgment under a given interaction index. If pollutants are independent (i.e. $\mu \rightarrow 0$), the curve gives the real safe/hazardous boundary. In contrast, if pollutants are not independent (i.e. $\mu > 0$), the curve does not give the real safe/hazardous boundary: the area between the curve and the line passing through Points (1,0), (1,1) and (0,1), which corresponds to the Ott's (1978) eclipsing region, are assessed to be hazardous although the area is safe (i.e. alpha misjudgment). The control values are too strict, if a polluted material is almost free from other pollutants. If the interaction is complementary ($\mu = 1$), the safe/hazardous boundary passes through Points (2,0), (1,1) and (0,2). Under this condition, if a polluted material is almost free from Pollutant 2, the content of Pollutant 1 is acceptable until the value two times of the present value. If interaction is stronger (e.g. $\mu = 2$), the control value of a pollutant should be four times of the present value where the other pollutants are free.

A hazardous case is assessed to be safe in the beta misjudgment. This type of misjudgment must be clearly eliminated. In contrast, a safe case is assessed to be hazardous in the alpha misjudgment. Misjudgment of this type seems to be acceptable. If misjudgment of this type is acceptable, control values will be given as the safe/hazardous boundary passes through Point (1, ..., 1). When a content of a pollutant is higher than the control value, we will take some action. The action needs energy, and the energy demand causes other environmental problems (e.g. global warming). This example suggests that the risks caused by the alpha misjudgment are scarcely recognized. Accordingly we can say risks of the type to be invisible. In contrast, the risks caused by the beta misjudgment are visible. Anyway, the alpha misjudgment must be also eliminated, even though the risks caused by the misjudgment are invisible.

Finally we have to consider how to determine control values when we have no datum concerning interaction among pollutants. Unfortunately, we have no answer of this question. The complementation assumption seems to give a starting point for determining control values, although we do not have any reason for accepting this assumption. It is clear, however, that the complementation assumption takes combined pollutions into account

compared with the independency assumption.

4.3. Comments for Applying Interaction Index Concept

This paper has introduced and developed a concept of over-content ratio and interaction index to assess risks caused by interaction among multiple pollutants considering environmental pollutions. This concept, however, is applicable for assessing risks in many fields other than environmental pollutions, as shown in the case study of lung cancer relating to cigarette smoking and asbestos exposure.

When the interaction between cigarette smoking and asbestos exposure was discussed, it was assumed that A_j in Equation (4') represented the degree at which the death rate was 1, because detailed data for this topic had not been presented. When Equation (4') is applied to such kind of topics, A_j (control value) and x_j (content) should be given by ADI (Acceptable Daily Intake) or TDI (Tolerable Daily Intake) and actual exposure of chemicals, respectively: for example, the TDI of dioxins, whose control values were shown previously, is 4 pg-TEQ/kg/d. In this case, it seems to be adequate to say r_j (over-content ratio) "over-exposure ratio".

Equation (4') may be also applied for assessing metabolic risks. Obesity is said for the condition that BMI (=weight/height² in kg/m²) is higher than 26 in Japan. Hyperlipidemia is defined as a condition where total cholesterol is more than 220 mg/dl or triglycerides are over 150 mg/dl. Hypertension (high blood pressure) is defined as repeatedly elevated blood pressure exceeding 140 mmHg (systolic pressure) and 90 mmHg (diastolic pressure). Diabetes is defined as a condition where fasting glucose is over 126 mg/dl. According to the National Cholesterol Education Program- Adult Treatment Panel (NCEP-ATP III) and the WHO diagnostic criteria (stated with parentheses), in contrast, the metabolic syndrome is identified as the presence of three or more of the following components: 1) waist circumference > 102 cm for men and 88 cm for women (waist/hip ratio > 0.90 for men and 0.85 for women, or BMI > 30 kg/m²; waist circumference > 85 cm and 90 cm in Japan), 2) triglycerides $\geq$ 150 mg/dl, 3) HDL ("good") cholesterol < 40 mg/dl for men and 50 mg/dl for women (< 35 mg/dl and 39 mg/dl, respectively; < 40 mg/dl in Japan), 4) blood pressure $\geq$ 130/85 mmHg ($\geq$ 140/90 mmHg; $\geq$ 130/85 mmHg in Japan), and 5) fasting glucose $\geq$ 110 mg/dl (e.g. Deen, 2004; Ministry of Health, Labour and Welfare <http://www.mhlw.go.jp/shingi/2005/08/s0826-9d.html>). This implies that metabolic syndrome is developed by combination of or interaction between several components. It is expected, therefore, that Equation (4') is applicable for assessing risks of metabolic syndrome. If the concept of interaction index is considered to be applicable for many fields, it may be adequate to say r_j (over-content ratio) "excess ratio", "saturation ratio" or any other word as a commoner term.

This paper has stated risks commonly. When health risks are discussed, endpoints are important, because each risk depends on not only chemicals but also endpoints. It is easily supposed, accordingly, that an interaction among chemicals depends largely on the combination of an endpoint and the chemicals. This means that A_j in Equation (4') should be given for each combination of endpoints and chemicals. The supposition also suggests that interaction indices depend on combinations of endpoints and chemicals.

5. Conclusions

In order to assess interaction among multiple pollutants, a concept of over-content ratio $r_j = x_j/A_j$ is proposed, and the following conclusions are obtained: 1) the interaction between two pollutants can be assessed by the equation $r_1^{1/\mu} + r_2^{1/\mu} = r_0^{1/\mu}$, where μ (say interaction index) and r_0 are positive constants; 2) the interaction is independent at $\mu \rightarrow 0$, somewhat independent and somewhat complementary (or negative) at $0 < \mu < 1$, completely complementary at $\mu = 1$, and stronger than the complementary case (or positive) at $\mu > 1$; and 3) environmental assessments which do not take account of interaction among pollutants may cause risks that hazardous cases are assessed to be safe or that safe cases are assessed to be hazardous.

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Notes

[1] If $r_1 = r_2 = \dots = r_m = \bar{r}$ in Equation (11), then $r = m^{\mu} \bar{r}$. The

equation presents straight lines whose slopes are m^{μ} . When the line of $\mu = 1$ is considered to be standard (completely complementary), lines of $\mu > 1$ (i.e. $1/\mu < 1$) pass above the standard line (i.e. positive interaction), while lines of $\mu < 1$ ($1/\mu > 1$) pass below the standard line (negative interaction).

[2] In order to solve the equation corresponding to Equation (10), the following logarithmic equation may be considered instead of Equation (9):

$$\mu_1 \log r_1 + \mu_2 \log r_2 = \mu_0 \log r_0.$$

However, the equation is not acceptable for assessing interaction between two components, because the equation means that r_1 diverges when $r_2 \rightarrow 0$. In contrast, r_1 converges when $r_2 \rightarrow 0$ in Equation (9) as shown in Figure 3.

[3] $0.322 \approx \{(1+\sqrt{5})/2\}^{1/3} + \{(1-\sqrt{5})/2\}^{1/3}$ (root of $r^3+3r-1=0$), and $0.333 \approx 1/3$ (root of $3r-1=0$).

要 旨

複合汚染における原因物質間の相互作用を評価するための数式

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汚染物質 j の濃度を x_j , その規制値を A_j として過濃度比 $r_j = x_j/A_j$ を定義したとき, μ と r_0 を正の定数としてそれらの組合せで得られる数式 $\sum r_j^{1/\mu} = r_0^{1/\mu}$ で, 汚染物質間の相互作用を評価することができる. すなわち, 相互作用はその程度を μ (相互作用指数) で表すことができ, 1) $\mu \rightarrow 0$ ならば独立, 2) $0 < \mu < 1$ ならば独立と相補的の間 (あるいは負), 3) $\mu = 1$ ならば完全に相補的であり, 4) $\mu > 1$ ならば相補的よりも強い (あるいは正である). この式に従うと, ダイオキシン類の環境基準は, ダイオキシン類に属す化学物質間の相互作用が完全に相補的であるという前提の下に与えられている. 石綿と喫煙に関連する肺癌の死亡率 r は, r_A と r_C をそれぞれ石綿のみおよび喫煙のみの場合の死亡率として, $r = [r_A^{1/3} + r_C^{1/3}]^3$ なる式で与えられる. この式は, 両方による死亡率 (約 600) が, 石綿のみの場合 (約 50) の 12 倍で, 喫煙のみの場合 (約 100) の 6 倍であることを示している. また, 環境基準が汚染物質間の相互作用を考慮することなく決められると, 危険な場合が安全と評価されたり, 安全な場合が危険と評価されるリスクが生じることが, 上述の式から導かれる. 相互作用指数を定量的に与えることができないときは, 相互作用が完全に相補的であると仮定することが妥当なようである.

キーワード：環境, 複合汚染, 相互作用, 過濃度比, 数学

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