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Methodological study applying three-mode factor analysis to three-way chemical data sets

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ABSTRACT

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A multivariate data analysis method, three-mode factor analysis (TMFA), has primarily been employed in the social sciences. Although it has not been extensively used in the natural sciences, TMFA provides the opportunity to examine data that are collected in form of a three-way matrix. With TMFA, one can simultaneously examine system variations in the three dimensions to determine the causal factors that control the system. This approach has been applied to the receptor modeling problem that attempts to relate ambient air quality to sources of pollution. By simulating an air pollution system, the relationships between the results of TMFA and the underlying physical model are investigated. In this way, the physical interpretation of the results of TMFA has been found. The model can be generalized to suit many three-way chemical data sets. It is also found that varimax rotation of the initially derived factors greatly improves their interpretability. The rotated solution can reflect the nature of the underlying physical system.

INTRODUCTION

In many chemical experiments or environmental measurement programs, data are gathered in such a way that they may be organized as a three-way table. For example, chemical species may be determined in airborne particulate samples collected at several sampling sites during different periods of time. Similar situations can be easily found in chemical studies. Measurement of absorption spectra of different wavelength could be made at certain positions in a system (e.g. along a chromatography column, within a reactor used in kinetics studies) under various experimental conditions. The measured properties, locations, and conditions can then serve as three modes of the three-way matrices. These data matrices may contain a wealth of information that could be revealed by appropriate data analysis methods.

Factor analysis [1–3] is a widely used multivariate method for finding causal factors that control the system. In the air pollution example mentioned above, factor analysis can be applied to a

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two-way matrix (chemical species at various locations or chemical species in different periods, i.e. a plane in the three-way matrix). The results of factor analysis can: (1) determine the number of types of pollution sources (factor number) in the system; (2) identify the sources by their chemical characteristics; and (3) investigate their influences in different situations [3,4]. A special factor analysis technique called target transformation factor analysis (TTFA) [2,3] can quantitatively obtain the mass contribution of each source to each sample [5]. Many similar applications for chemical data can be found in the monograph by Malinowski and Howery [2].

However, conventional (two-mode) factor analysis can only be applied to a two-way matrix. The analysis is thus only on a single plane oriented in certain direction in the complete three-way data block. Since only a limited part of the data is used, the results may be distorted and some information about the interactions among the modes may be lost. There is another factor analysis method called three-mode factor analysis [6,7] (denoted as TMFA hereafter). This method works directly on the full three-way data matrix as its input, and thus can examine variations among all three modes simultaneously. The ability to examine three-way observational data suggest the potential utility of TMFA.

TMFA was originally introduced by Tucker in the 1960s for interpretation of psychological studies [6]. Since then, many applications have appeared in the social science literature [7]. In these applications, there was no underlying physical model for the studied systems. Therefore, the interpretation of the results has only been presented in statistical sense. Because of a lack of an underlying physical model, application of TMFA in natural sciences has been very limited. Only a few applications are found in the literature [8–11]. The model in these applications "is not a physical, but a mathematical-statistical model" [10]. TMFA is still not a clear model that natural scientists can meaningfully relate to their physical systems. However, it is an attractive method because it opens a new dimension to extract more information. In order to explore the application of TMFA to three-way environmental or chemical data and examine its interpretation in terms of the physical and chemical processes operating within the system, a methodological study on data with a known, well defined structure has been performed.

THEORY

The basic concept of TMFA is to extend the two-mode (conventional) factor analytical model to three-way data. Each mode corresponds to a class of variables, i.e. a 'mode'. The term 'mode' is used to mean a "set of indices by which data might be classified" [6]. Through TMFA, the data cube is decomposed into three two-way matrices (called factor loading matrices) and one three-way matrix (called the core matrix). Similar to conventional factor analysis, most of the variation of measured variables is compressed into a few factors according to the covariance among the variables. The results can determine: (1) how many underlying causal factors are controlling the system; (2) what the relationship is between the factors and variables; and (3) how much of the system variance is accounted for by the factors.

TMFA model is presented in terms of elements of the factor loading matrices, **A**, **B**, and **C**, and the core matrix, **G**.

$$x_{ijk} = \sum_m \sum_p \sum_q a_{im} b_{jp} c_{kq} g_{mpq} \tag{1}$$

The model can also be expressed in matrix form with a Kronecker product [12].

$${}_i\mathbf{X}_{(jk)} = {}_i\mathbf{A}_m \mathbf{G}_{(pq)} \left({}_p\mathbf{B}_j \otimes {}_q\mathbf{C}_k \right) \tag{2}$$

The notational device that Tucker [6] employed has been used here. The subscript (e.g. i) is used in several related, but distinct roles: (1) as a general identification of the mode; (2) as a subscript identifying the mode to which an element belongs, (3) as a variable identification symbol for the elements in the mode. The pre-subscript letter denotes the row mode while the post-subscript letter is the designation for the column mode. Reversal of the subscripts indicates transposition, for example, ${}_i\mathbf{A}_j$ is the transpose of ${}_j\mathbf{A}_i$. The matrix ${}_i\mathbf{X}_{(jk)}$ is the three-mode data matrix rearranged as a two-mode matrix by sequentially

placing all the vertical planes, that are parallel to the side planes of the three-mode matrix, side by side along the row direction. The designation (jk) is called combination mode. The order (jk) may be read as j-outer loop, k-inner loop.

The Kronecker product of ${}_p\mathbf{B}_j$ and ${}_q\mathbf{C}_k$, denoted as ${}_p\mathbf{B}_j \otimes {}_q\mathbf{C}_k$, yields ${}_{(pq)}\mathbf{H}_{(jk)}$. Then ${}_{(pq)}\mathbf{H}_{(jk)}$ can be represented as below as a supermatrix containing submatrices proportional to the matrix ${}_q\mathbf{C}_k$

$$ {}_{(pq)}\mathbf{H}_{(jk)} = \begin{pmatrix} (b_{11q}\mathbf{C}_k) & (b_{12q}\mathbf{C}_k) & \cdots \\ (b_{21q}\mathbf{C}_k) & (b_{22q}\mathbf{C}_k) & \cdots \\ \vdots & \vdots & \cdots \end{pmatrix} \tag{3} $$

Using these definitions and notations, the three-mode problem is reduced to a two-mode problem as in eq. (2). Tucker [6] showed that the matrices ${}_i\mathbf{A}_m$, ${}_j\mathbf{B}_p$, and ${}_k\mathbf{C}_q$ in eq. (2) could be obtained from the matrices ${}_i\mathbf{M}_i$, ${}_j\mathbf{P}_j$, and ${}_k\mathbf{Q}_k$. These latter matrices are analogous to covariance matrices in conventional factor analysis.

$$ {}_i\mathbf{M}_i = {}_i\mathbf{X}_{(jk)}\mathbf{X}_i \tag{4a} $$

$$ {}_j\mathbf{P}_j = {}_j\mathbf{X}_{(ik)}\mathbf{X}_j \tag{4b} $$

$$ {}_k\mathbf{Q}_k = {}_k\mathbf{X}_{(ij)}\mathbf{X}_k \tag{4c} $$

The factor loading matrices ${}_i\mathbf{A}_m$, ${}_j\mathbf{B}_p$, and ${}_k\mathbf{C}_q$ can be obtained in this way: m significant eigenvectors from ${}_i\mathbf{M}_i$ constitute ${}_i\mathbf{A}_m$; p significant eigenvectors from ${}_j\mathbf{P}_j$ constitute ${}_j\mathbf{B}_p$; and q significant eigenvectors from ${}_k\mathbf{Q}_k$ constitute ${}_k\mathbf{C}_q$.

The core matrix ${}_m\mathbf{G}_{(pq)}$ is given by

$$ {}_m\mathbf{G}_{(pq)} = {}_m\mathbf{A}_i^+ \mathbf{X}_{(jk)} \left({}_j\mathbf{B}_p^+ \otimes {}_k\mathbf{C}_q^+ \right) \tag{5} $$

where

$$ {}_m\mathbf{A}_i^+ = \left({}_m\mathbf{A}_i\mathbf{A}_m \right)^{-1} {}_m\mathbf{A}_i \tag{6a} $$

$$ {}_p\mathbf{B}_j^+ = \left({}_p\mathbf{B}_j\mathbf{B}_p \right)^{-1} {}_p\mathbf{B}_j \tag{6b} $$

$$ {}_q\mathbf{C}_k^+ = \left({}_q\mathbf{C}_k\mathbf{C}_q \right)^{-1} {}_q\mathbf{C}_k \tag{6c} $$

Since matrices **A**, **B**, and **C** are column-wise sections of orthonormal,

$$ {}_m\mathbf{A}_i\mathbf{A}_m = {}_m\mathbf{I}_m, \quad {}_p\mathbf{B}_j\mathbf{B}_m = {}_p\mathbf{I}_p, \quad {}_q\mathbf{C}_k\mathbf{C}_q = {}_q\mathbf{I}_q. \tag{7} $$

Then,

$$ {}_m\mathbf{A}_i^+ = {}_m\mathbf{A}_i, \quad {}_p\mathbf{B}_j^+ = {}_p\mathbf{B}_j, \quad {}_q\mathbf{C}_k^+ = {}_q\mathbf{C}_k \tag{8} $$

Eq. (5) becomes

$$ {}_m\mathbf{G}_{(pq)} = {}_m\mathbf{A}_i \mathbf{X}_{(jk)} \left({}_j\mathbf{B}_p \otimes {}_k\mathbf{C}_q \right) \tag{9} $$

In principle, all the system information is contained in the three factor loading matrices (**A**, **B**, and **C**) and one core matrix (**G**) in terms of system causal factors. As in conventional factor analysis, each loading matrix (e.g. ${}_i\mathbf{A}_m$) will show the relationship between the variables (i) and factors (m) in that mode. The squares of the elements in the core matrix are the indication of how much the system variance is accounted for by the corresponding combination of factors (m, p, and q) from each mode. This relationship can be seen from following equation:

$$ \sum_i \sum_j \sum_k x_{ijk}^2 = \sum_m \sum_p \sum_q g_{mpq}^2 \tag{10} $$

In conventional factor analysis, a rotation is usually needed before interpreting the results. Tucker [6] also presented a transformation scheme. Let the matrices ${}_m\mathbf{T}_{m^*}$, ${}_p\mathbf{T}_{p^*}$, and ${}_q\mathbf{T}_{q^*}$ be square, non-singular matrices, and let

$$ {}_i\mathbf{A}_m\mathbf{T}_{m^*} = {}_i\mathbf{A}_{m^*} \tag{11a} $$

$$ {}_j\mathbf{B}_p\mathbf{T}_{p'} = {}_j\mathbf{B}_{p^*} \tag{11b} $$

$$ {}_k\mathbf{C}_q\mathbf{T}_{q^*} = {}_k\mathbf{C}_{q^*} \tag{11c} $$

The m^*, p^*, and q^* are transformed derivational modes. For core matrix:

$$ {}_{m'}\mathbf{G}_{(p^*q^*)} = \left({}_m\mathbf{T}_{m^*} \right)^{-1} {}_m\mathbf{G}_{(pq)} \left[\left({}_{p^*}\mathbf{T}_p \right)^{-1} \otimes \left({}_{q^*}\mathbf{T}_q \right)^{-1} \right] \tag{12} $$

In transformed form, eq. (2) becomes

$$ {}_i\mathbf{X}_{(jk)} = {}_i\mathbf{A}_{m^*} \mathbf{G}_{(p^*q^*)} \left({}_{p^*}\mathbf{B}_j \otimes {}_{q^*}\mathbf{C}_k \right) \tag{13} $$

In terms of algorithms, there are several approaches other than the method described above to perform TMFA. Kroonenberg [7] presented these approaches, particularly an alternating least squares algorithm (TUCKALS3, see Appendix).

APPLICATION

In applying TMFA to the natural sciences, it is desirable not only statistically, but physically to

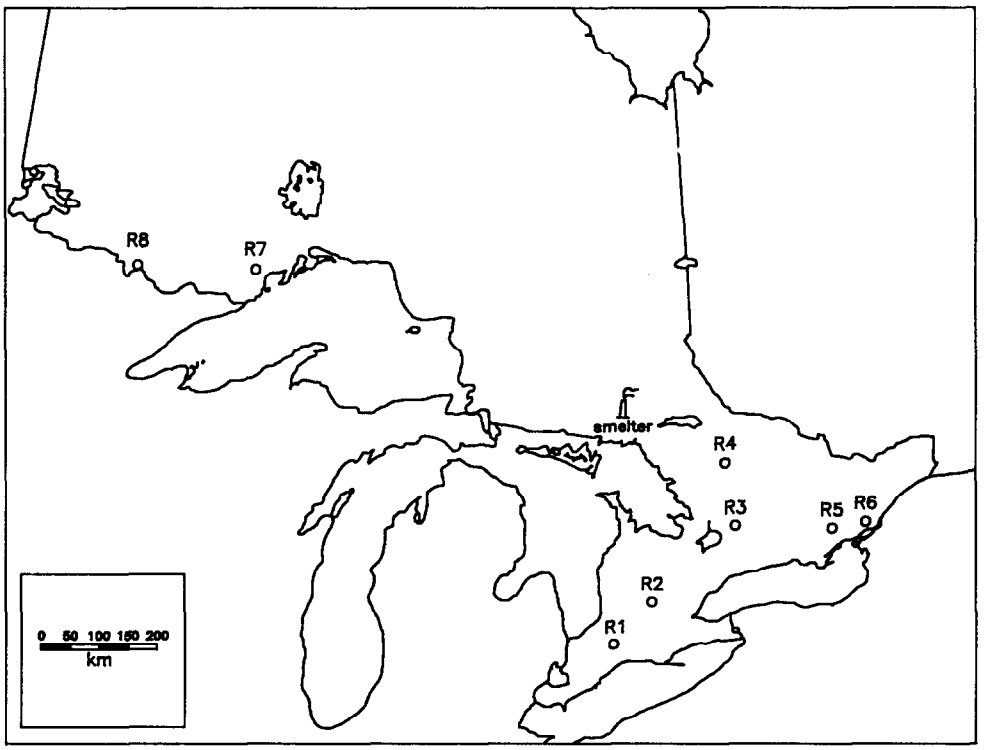

Fig. 1. Map of the simulated system showing the Great Lakes and Southern Ontario, Canada.

relate the TMFA model to the actual physical system. In order to examine these relationships, it is extremely useful to employ a simulated data set with a known underlying structure.

Description of the simulated data

An air pollution example can be simulated without loss of generality. A simulated physical system has been developed based on Ontario, Canada. The system is assumed to consist of eight sampling sites (R1–R8) as shown in Fig. 1. Three classes of particulate pollution sources are considered. The first one (S1) is emissions from local coal-fired power plants. The sources in this class are considered as area sources and exist in all directions around the sampling sites. The second source type (S2) is assumed to be long-range transported power plant emissions from the Midwest of the United States. The third source (S3) is a point source simulating a nickel smelter in Sudbury, Ontario. Within a source class, the chemical composition of the emitted particles is assumed to be the same. The compositions given in Table 1 were obtained by modifying reported data [3,13].

The samples are assumed to be collected during 12 periods that have different wind directions and wind strengths (Fig. 2). The particles from differ-

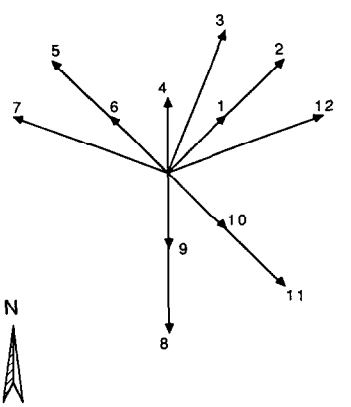

Fig. 2. Wind directions and strengths in the twelve periods.

TABLE 1

Chemical compositions of the particles emitted from three classes of sources

No.	Element	Source 1: local coal (S1)	Source 2: regional coal (S2)	Source 3: Ni smelter (S3)
1	Na	9.7	5.9	1.4
2	Al	26	8.8	2.5
3	Si	150	47	5
4	S	15	71	40
5	Cl	11	12	11
6	K	10	6.3	5
7	Ca	23	11.5	5
8	Ti	2.6	1.7	2.8
9	V	0.48	1.7	0.01
10	Cr	0.4	0.9	1
11	Mn	0.48	6.5	0.22
12	Fe	18	10.7	44
13	Ni	0.2	0.7	250
14	Cu	1.3	3.2	2
15	Zn	3.2	32	1.9
16	As	0.3	3	0.05
17	Se	0.11	1.1	0.05
18	Br	0.2	1.3	0.84
19	Sb	0.03	0.9	0.05
20	Pb	2	8.2	10

ent sources are transported to the sampling sites. Under different wind condition, the mass contributions of the sources to the sites will vary. The assigned contributions are tabulated in Table 2, and they are constructed according to the system layout and the wind condition for each period.

A simulated data set then can be produced from the chemical compositions and the mass contributions by the following equation,

$$x_{ijk} = \sum_{h=1} \alpha_{ih}\beta_{bjk} \tag{14}$$

where x_{ijk} = concentration of element i at sampling site j in period k (ng/m^3 air); α_{ih} = content of element i in the particles coming from source h (Table 1) (ng/μg); β_{bjk} = mass contribution of source h at site j in period k (Table 2) (μg/m^3 air). Actually, eq. (14) is the physical model of the system. Therefore, the method developed in this work will be applicable to other chemical data as long as a physical model in the form of eq. (14) can be established.

Results

The 'observed' data set created through eq. (14) is taken as the input data matrix to the TMFA computer program, TUCKALS3 [14]. Modes 1, 2, and 3 (i, j, and k) correspond to chemical elements, sampling sites, and time periods, respectively. The data are centered and scaled in such a way that the mean of each frontal plane equals zero and the sum of squares per frontal plane equals one. The retained numbers of factors in each mode are determined according to the model fitting and the residuals (Table 3). Since the numbers of factors need to be determined for all three

TABLE 2

Mass contributions of the sources in each sample

Period	R1			R2			R3			R4		
	S1	S2	S3	S1	S2	S3	S1	S2	S3	S1	S2	S3
1	777	385	1	685	308	3	608	178	13	534	133	29
2	298	803	0	274	827	2	235	791	4	201	753	8
3	239	777	1	254	835	3	198	809	3	222	843	5
4	631	377	3	582	282	2	555	156	10	498	99	20
5	209	281	2	234	268	1	209	217	3	229	198	5
6	555	298	2	567	289	3	520	245	5	502	208	8
7	192	259	1	213	269	2	197	203	3	203	201	3
8	194	131	56	201	125	43	162	109	19	143	103	7
9	491	172	70	482	156	92	421	148	67	393	162	41
10	472	161	53	456	165	88	403	153	186	402	148	223
11	209	153	8	211	171	11	189	151	22	162	139	17
12	274	772	2	268	805	1	253	811	3	233	790	5

R5			R6			R7			R8		
S1	S2	S3	S1	S2	S3	S1	S2	S3	S1	S2	S3
437	123	15	485	109	11	207	82	2	248	78	1
222	729	3	219	693	2	138	79	1	149	101	2
199	632	2	183	591	1	109	108	2	132	171	1
389	95	7	444	92	6	227	79	2	272	97	3
173	235	3	176	219	2	93	523	183	91	489	125
378	209	4	409	198	3	219	111	29	252	102	22
151	212	1	149	199	1	90	309	301	85	345	339
96	111	3	89	99	2	59	67	1	55	73	1
387	151	20	378	148	18	198	77	2	179	65	1
375	133	85	362	129	79	174	70	1	181	59	2
108	117	25	93	119	27	89	63	1	76	58	2
230	811	2	228	823	2	197	71	1	182	70	1

modes, it is a more complicated problem as compared to conventional factor analysis. To simplify the situation, one mode is examined at a time with the other two modes assigned a sufficiently high number of factors to insure that those two modes fit perfectly (five factors are used in this case). From the sum of squares of individual modes [$SS(Fit_I)$, $SS(Fit_{II})$, and $SS(Fit_{III})$], the overall fit [SS(Fit)], and SS(Res) (see Appendix and Table 3), the role of each added factor can be seen. For mode 1, $SS(Fit_I)$ becomes larger and larger as more factors are taken. In other words, more variation is accounted for. Meanwhile, the SS(Res) decreases and the overall model fit is improved. From eq. (A.15) in the Appendix, the overall fit, SS(Fit), is restricted by $SS(Fit_I)$, that is the minima among all $SS(Fit_I)$, $SS(Fit_{II})$, and $SS(Fit_{III})$, when 1, or 2, or 3 factors are taken for mode 1. As

TABLE 3

Sum of squares (SS) with different number of retained factors (standardized such that SS(Total) = 100)

Number of retained factors	$SS(Fit_I)$ (mode 1)	$SS(Fit_{II})$ (mode 2)	$SS(Fit_{III})$ (mode 3)	SS(Fit) (overall)	SS(Res) (residuals)
Test for mode 1 ($N2 = 5$, $N3 = 5$)					
$N1 = 1$	85.39	99.92	99.10	85.37	14.63
$N1 = 2$	93.16	99.92	99.06	92.70	7.30
$N1 = 3$	97.40	99.92	99.06	96.57	3.43
$N1 = 4$	100.00	99.92	99.06	99.06	0.94
$N1 = 5$	100.00	99.92	99.08	99.08	0.92
Test for mode 2 ($N1 = 5$, $N3 = 5$)					
$N2 = 1$	100.00	88.49	99.10	88.49	11.51
$N2 = 2$	100.00	98.07	99.06	98.04	1.96
$N2 = 3$	100.00	99.35	99.10	98.47	1.26
$N2 = 4$	100.00	99.82	99.07	99.04	0.96
$N2 = 5$	100.00	99.92	99.08	99.08	0.92
Test for mode 3 ($N1 = 5$, $N2 = 5$)					
$N3 = 1$	100.00	99.82	85.98	85.98	14.02
$N3 = 2$	100.00	99.82	93.32	93.31	6.69
$N3 = 3$	100.00	99.82	97.40	97.39	2.61
$N3 = 4$	100.00	99.82	98.59	98.56	1.44
$N3 = 5$	100.00	99.92	99.08	99.08	0.92

TABLE 4

Factor loadings of the first mode

i	Element	Unrotated			Varimax rotated		
		$m=1$ (S1)	$m=2$ (S3)	$m=3$ (S2)	$m^*=1$ (S1)	$m^*=2$ (S3)	$m^*=3$ (S2)
1	Na	−0.046	0.047	0.009	−0.053	0.038	0.012
2	Al	0.051	0.024	−0.050	0.059	0.044	−0.014
3	Si	**0.871**	−0.112	−0.274	**0.920**	0.024	0.019
4	S	0.333	0.386	**0.745**	0.028	−0.004	**0.902**
5	Cl	−0.004	0.096	0.062	−0.037	0.057	0.092
5	K	−0.041	0.059	0.005	−0.049	0.050	0.016
7	Ca	0.051	0.048	−0.002	0.041	0.044	0.036
8	Ti	−0.103	0.052	−0.007	−0.102	0.050	−0.018
9	V	−0.115	0.048	0.010	-0.118	0.039	−0.010
10	Cr	−0.119	0.049	−0.002	−0.119	0.044	−0.021
11	Mn	−0.090	0.066	0.071	−0.117	0.026	0.057
12	Fe	0.043	0.183	−0.054	0.029	0.187	0.048
13	Ni	0.015	**0.854**	−0.453	0.020	**0.966**	−0.003
14	Cu	−0.102	0.058	0.021	−0.111	0.042	0.008
15	Zn	0.054	0.153	0.380	−0.088	−0.039	0.402
16	As	−0.109	0.053	0.028	−0.119	0.035	0.009
17	Se	−0.120	0.047	0.004	−0.121	0.040	−0.017
18	Br	−0.118	0.050	0.005	−0.110	0.043	−0.014
19	Sb	−0.121	0.047	0.002	−0.122	0.041	−0.019
20	Pb	−0.069	0.100	0.067	−0.100	−0.058	0.075

factors are added to mode 1 beyond the third one, SS(Fit) is limited by mode 3. This result means that the factors following the third one are unnecessary and that three factors should be retained for mode 1. Similarly, two factors are retained for mode 2.

There must be one mode that will restrict SS(Fit) even when the highest number of factors are retained. Mode 3 is this limiting mode in this case. The criterion described above is not suitable for this mode. The methods used in conventional factor analysis may be used to determine the number of factors in this mode. In Table 3, $SS(Fit_{III})$ increases as $N3$ increases. The increments after $N3 = 3$ are less than 1.19% and tend to stabilize while the previous two increments are 7.34% and 4.08%. Three factors are therefore retained for this mode. Thus, the numbers of factors are determined to be 3, 2, and 3 for modes 1, 2, and 3, respectively. The final SS(Fit) is 95% of SS(Total), and the SS(Res) is 5%. The output from the program is unrotated loading matrices **A**, **B**, **C**, and core matrix **G** (Tables 4–7, respectively).

In conventional factor analysis, factor axis rotation is performed. Usually, a rotation transforms the results to 'simple structure' [4] and make them easier to interpret. A rotation can also be applied to the loading matrices of TMFA in the same manner because the loading matrices are two-way matrices as in conventional factor analysis. Several rotation methods (both orthogonal and oblique) have been tested (Table 8). These methods can be

TABLE 5

Factor loadings of the second mode

j	Site	Unrotated		Varimax rotated	
		$p=1$ (zone 1)	$p=2$ (zone 2)	$p^*=1$ (zone 1)	$p^*=2$ (zone 2)
1	R1	**0.459**	−0.095	**0.468**	0.023
2	R2	**0.455**	−0.100	**0.465**	0.017
3	R3	**0.405**	−0.133	**0.425**	−0.028
4	R4	**0.382**	−0.126	**0.401**	−0.027
5	R5	**0.326**	−0.082	**0.336**	0.003
6	R6	**0.322**	−0.078	**0.331**	0.005
7	R7	0.175	**0.691**	−0.004	**0.713**
8	R8	0.181	**0.676**	0.006	**0.700**

TABLE 6

Factor loadings of the third mode

k (period)	Unrotated			Varimax rotated		
	$q=1$ (regime ?)	$q=2$ (regime ?)	$q=3$ (regime ?)	$q^*=1$ (regime 1)	$q^*=2$ (regime 2)	$q^*=3$ (regime 3)
1	0.301	0.071	−0.207	**0.372**	−0.014	0.013
2	0.291	0.153	0.452	−0.006	0.016	**0.559**
3	0.289	0.1452	0.482	−0.027	0.004	**0.579**
4	0.303	0.047	−0.207	**0.368**	−0.037	0.008
5	0.263	−0.542	0.039	0.064	**−0.599**	0.038
6	0.308	0.014	−0.147	**0.329**	−0.075	0.051
7	0.208	−0.762	0.063	−0.040	**−0.790**	−0.028
8	0.301	0.092	−0.131	**0.331**	0.000	0.079
9	0.303	0.099	−0.291	**0.429**	0.019	−0.046
10	0.284	0.166	−0.380	**0.481**	0.095	−0.112
11	0.307	0.088	−0.062	**0.295**	−0.011	0.137
12	0.291	0.149	0.449	−0.005	0.012	**0.555**

applied with or without row normalization. In row normalization, the factor loadings for each variable are normalized by the corresponding communality. The similarities of the different rotated loadings to the true source compositions are listed in Table 9 in forms of correlation coefficients. In this particular form, the 'simple structure' means that loadings of one factor tend to relate to only one source composition. The higher in uniqueness and correlation to one source, the better is the solution. Therefore, the correlation coefficients can be used to select the best rotation method. Since the results of all the orthogonal rotation without normalization are equal, only one set (VARU) of values appears in Table 9. Also, the results of the oblique rotations are similar to that of the varimax rotation so that only the results of the oblimax rotation are listed in the table.

Table 9 shows that the correlations increased after rotation, and the loadings become more identifiable, particularly for factors 1 and 3 (1 and 2 in oblimax case). It should be noted that corresponding source compositions themselves are moderately correlated with correlation coefficient of 0.53. Varimax without normalization (VARU) and oblimax give the best results. Since orthogonal rotations have the advantage of maintaining factor loadings as representation of the weights of factors on variables, and the squares of core values can be considered as explained variance, VARU is preferable to an oblique rotation. Therefore, varimax without row normalization has been selected, and is referred to as varimax for simplicity. The varimax rotated factor loadings were obtained and are given in Tables 4–6 along with the corresponding unrotated values. An inverse

TABLE 7

Unrotated core matrix

	Regime 1 ($q=1$)		Regime 2 ($q=2$)		Regime 3 ($q=3$)	
	Zonel ($p=1$)	Zone 2 ($p=2$)	Zone 1 ($p=1$)	Zone 2 ($p=2$)	Zone 1 ($p=1$)	Zone 2 ($p=2$)
Source 1 ($m=1$)	40.091	0.299	−0.198	−4.234	−0.062	−0.685
Source 2 ($m=3$)	0.036	−1.591	0.158	0.431	8.164	−2.010
Source 3 ($m=2$)	−1.136	−0.886	−0.218	−10.862	0.483	0.297

TABLE 8

The rotations used in this work

Orthogonal rotations:	
Varimax with row normalization	(VARN)
Varimax without row normalization	(VARU)
Quartimax with row normalization	(QUAN)
Quartimax without row normalization	(QUAU)
Equamax with row normalization	(EQUN)
Equamax without row normalization	(EQUU)
Oblique rotations:	
Quartimin with row normalization	
Quartimin without row normalization	
Direct oblimin with row normalization	
Direct oblimin without row normalization	
Oblimax *	

* The program used for oblimax is from SOUPAC package at University of Illinois. Others are taken from IMSL Stat/Library.

transformation given by eq. (12) is needed to satisfy eq. (2) as eq. (13). The corresponding varimax rotated core matrix is given in Table 10.

Discussion

The interpretation of TMFA result is primarily based on: (1) the factor loading (e.g. a_{im}) being the weight of the factor (m) on the variable (i); (2) the square of a element in the core matrix, g^2_{mpq}, being a measure of the system variance accounted for by the combination of factors m, p, and q.

The first mode

Mode 1 is associated with the chemical properties. From the unrotated loading matrix ${}_iA_m$ in Table 4, it can be seen that factor 1 is highly related with element Si that is a major constituent of source 1 (Table 1). The loading pattern of factor 1 and composition of source 1 are similar. In other words, the values of these two columns are close if they are respectively rescaled by dividing by the maximum value of each column. The correlation coefficient between the two columns can be used as a measure of their similarity. This coefficient is 0.93 (Table 9). The varimax rotated loadings of factor 1 have stronger correlation with source 1. In summary, factor 1 in mode 1 is the

TABLE 9

Correlation coefficients between source profiles and loadings of mode 1

	Unrotated			VARN			QUAN			EQUN			VARU			Oblimax		
	F1	F2	F3	F1	F2	F3	F1	F2	F3	F1	F2	F3	F1	F2	F3	F1	F2	F3
Source 1 (S1)	0.93	−0.28	−0.26	0.97	0.26	−0.14	0.98	0.28	−0.04	0.95	0.26	−0.23	0.99	−0.10	−0.01	0.99	−0.02	−0.08
Source 2 (S2)	0.78	0.10	0.59	0.67	0.06	0.77	0.60	0.08	0.83	0.73	0.06	0.71	0.54	−0.21	0.85	0.55	−0.85	−0.20
Source 3 (S3)	0.07	0.94	−0.40	−0.25	0.94	0.01	−0.26	0.93	−0.02	−0.24	0.94	0.03	0.06	0.98	0.05	0.09	−0.04	0.98

TABLE 10

Varimax rotated core matrix

	Regime 1 ($q^* = 1$)		Regime 2 ($q^* = 2$)		Regime 3 ($q^* = 3$)	
	Zone 1 ($p^* = 1$)	Zone 2 ($p^* = 2$)	Zone 1 ($p^* = 1$)	Zone 2 ($p^* = 2$)	Zone 1 ($p^* = 1$)	Zone 2 ($p^* = 2$)
Source 1 ($m^* = 1$)	29.497	7.745	−11.196	−5.580	18.317	4.694
Source 2 ($m^* = 3$)	6.168	−0.121	−3.473	−5.881	13.445	−0.109
Source 3 ($m^* = 2$)	1.818	−2.906	2.714	−9.013	−3.009	−1.808

representation of source 1 in the physical system. Similarly, factor 2 represents source 3 and factor 3 represents source 2. In this way, the sources in the system can be recognized by their chemical characteristics reflected through the factor loadings in mode 1.

The second mode

The pattern of the unrotated loadings in this mode is clear (Table 5), i.e.sites R1–R6 have large loadings of factor 1 and small loadings of factor 2, while sites R7 and R8 have large loadings of factor 2 and small loadings of factor 1. This contrast becomes stronger after rotation (Table 5). Referring to Fig. 1, all the sites can be divided into two groups according to the loadings. The concept of a 'pollution zone' can be introduced to refer a geographic region in which all the sites are polluted in a similar manner. Sites R1–R6 belong to zone 1, R7 and R8 to zone 2. By using the term 'zone', the sites are characterized.

The third mode

Based on the unrotated loadings in this mode (Table 6), the periods may be divided into three groups: one group of periods 5 and 7 (large loadings of factor 2), one of 2, 3, and 12 (large loadings of factor 3), and one of the remaining. However, it is hard to say what group factor 1 represents because all the loadings of factor 1 are essentially same. This mode is difficult to interpret because the factors cannot be directly related to physical situations. Fortunately, this problem is resolved by the rotation. The varimax rotated results (Table 6) clearly show that factor 1 represents the group of periods, 1, 4, 6, 8, 9, 10, and 11; factor 2 represents periods 5 and 7, and factor 3 represents periods 2, 3, and 12. The concept of 'primary meteorological regime' is then applied to refer to a class of meteorological conditions that govern the transport of pollutants to the receptor sites. Based on the rotated factor loadings (Table 6) and the wind conditions (Fig. 2), three primary

TABLE 11

Primary meteorological regime and its relation with the factors and the periods

Primary meteorological regime	1		2	3
Corresponding unrotated factor	Factor 1? ($q = 1$)		Factor 2? ($q = 2$)	Factor 3? ($q = 3$)
Corresponding varimax rotated factor	Factor 1 ($q^* = 1$)		Factor 2 ($q^* = 2$)	Factor 3 ($q^* = 3$)
Wind direction	All directions	Northern	SE	SW
	OR			
Wind speed	Weak	Strong	Strong	Strong
Corresponding periods	1, 4, 6, 9, 10	8, 11	5, 7	2, 3, 12

meteorological regimes are defined as presented in Table 11. Regime 1 (factor 1) includes weak winds in all directions or strong northern winds; regime 2 (factor 2) represents strong southeastern wind; and regime 3 (factor 3) is strong southwestern wind. It will be seen below that the concepts of 'zone' and 'regime' are very helpful to understand the system.

The core matrix

The relationships among the factors can be seen in the core matrix **G** (Tables 7 and 10), where g^2_{mpq} indicates the importance of the combination of the corresponding factors from each mode. In this sense, the unrotated core matrix (Table 7) is extremely difficult to interpret. Because of the complex three dimensional multiplication, it is hard to discern the physical variations in the system. For instance, in regime 1, the local sources (S1) appear to be totally dominant ($g_{111} = 40.1$) such that the other sources are negligible. This result does not agree with the 'physical model' in which the regional coal sources (S2) also have made significant contributions to most sites in this regime (Table 2 or Fig. 3). The elements of the unrotated core matrix (Table 7) are apparently not well partitioned. In particular, it can be seen that the third mode cannot be directly associated with the defined meteorological regimes.

The rotation significantly improves the ability to directly relate the results to the physical system. The varimax rotated core matrix (Table 10) can be interpreted using the concepts of 'source', 'pollution zone', and 'meteorological regime'.

Under the regime 1, the influence of source 1 on zone 1 is the most important ($g_{111} = 29.5$). The effect of source 1 on zone 2 and source 2 on zone 1 are also significant. Figure 3 is constructed based on the true mass contributions of the sources given in Table 2 averaged over the time periods and the sites that belong to the specific regimes and zones. Under regime 1, Fig. 3 shows that a large amount of particulate mass in zone 1 is contributed by source 1, and a significant amount by source 2. In zone 2, a significant contribution is from source 1. The information obtained from the core matrix is consistent with that shown in Fig. 3. It seems that the core matrix is related semi-

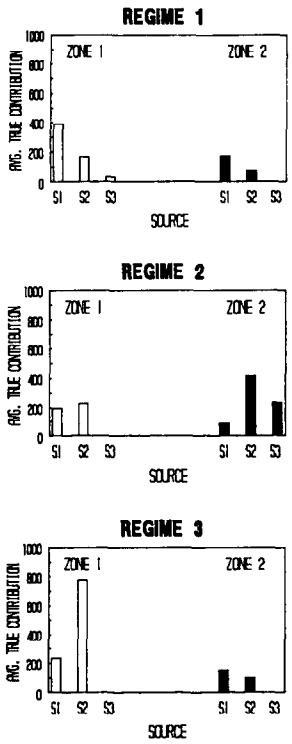

Fig. 3. The average contributions calculated from true values of corresponding sites and periods that belong to the specific zone and regime.

quantitatively to the source contribution in terms of zones under a given regime. It is obvious (Figs. 1 and 2) that long-range transport is not very important in weak wind conditions. The major contributors to all sites are the local sources (S1). Since upper Ontario is assumed to be a clean area, no long-range transported particles arrive at the sites in strong northern wind conditions (periods 8 and 11). Therefore, periods 8 and 11 have similar loadings to those of the weak wind periods (Table 6), and they belong to one regime (Table 11).

Under regime 2, the strong southeast wind decreases the contributions of local sources (S1), $g_{112} = -11.2$ and $g_{122} = -5.6$, and brings pollutants from the distant source (S2) and the smelter (S3) to zone 2, $g_{222} = -5.9$ and $g_{322} = -9.0$. Under regime 3, strong southwestern winds bring particles from the midwestern of the United States (S2) to zone 1, $g_{213} = 13.4$, supplementing the local source effect observed for the other condi-

tions. Referring to regime 1 and 2 shown in Fig. 3, it can be seen that these results basically reflect the variation of the source contributions as they were defined in the simulated system.

CONCLUSION

The method of analyzing three-way chemical data sets with three-mode factor analysis (TMFA) has been explored using a simulated data set. Since the underlying physical model is known, it is possible to physically interpret the results of the TMFA, and thus further develop this methodology. This study begins the advance of TMFA from a purely statistical model to a statistical-physical model. In general, the three modes of the data set are associated with generalized 'measured variable', 'position', and 'condition'. With TMFA, the data set can be decomposed into three two-way factor loading matrices and one three-way core matrix. This decomposition is based on simultaneous variations of causal factors in the system. The factors in the mode corresponding to 'measured variable' can be recognized as generalized 'sources' (e.g. pollution sources, chemical species if measured variable is absorption spectrum, etc.); the factors in 'position' mode as generalized 'zones' (e.g. pollution zones, concentration boundary layers, etc.); the factors in 'condition' mode as generalized 'regime' (e.g. wind condition, experimental conditions, etc.). In this way, the whole complicated physical system can be summarized by a few 'sources', 'zones', and 'regimes', and their relationships are shown in the core matrix.

Axis rotation of the initially derived factor solution is a very important step to make the results more interpretable. Varimax rotation without row normalization has been selected as the best rotation method. Good agreement between the TMFA model and the simulated physical system is achieved by this rotation. Qualitative interpretation and semi-quantitative results are obtained by this TMFA method. The research to develop a quantitative model by incorporating TMFA with target transformation [2] is in progress.

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APPENDIX

In practical applications, Tucker's model [eq. (1) or (2)] is not exactly true unless all of the components are used (i.e. $m = i$, $p = j$, and $q = k$). As in conventional factor analysis, only a few of principal components in each mode are retained ($m < i$, $p < j$, and $q < k$) in order to obtain a parsimonious model. Then an approximate solution is obtained

$$ {}_i\mathbf{X}_{(jk)} \approx {}_i\tilde{\mathbf{X}}_{(jk)} = {}_i\mathbf{A}_m\mathbf{G}_{(pq)}\left({}_p\mathbf{B}_j \otimes {}_q\mathbf{C}_k\right) \qquad (A.1) $$

It is desireable to make ${}_i\tilde{\mathbf{X}}_{(jk)}$ approach ${}_i\mathbf{X}_{(jk)}$ in at least squares sense. However, Tucker's procedures described in the theory section "do not produce a least squares approximation to the data" [6]. Kroonenberg and De Leeuw [15] developed an algorithm, TUCKALS3, that adopts an alternating least squares (ALS) approach. TUCKALS3 can provide least squares estimates of the parameters in the three-mode model. A outline of TUCKALS3 algorithm is given below without proof (see ref. 7 or 15 for details and proof).

A mean-squared loss function, f, is defined as

$$ f(\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{G}) = \|\mathbf{X} - \tilde{\mathbf{X}}\|^2 = \|\mathbf{X} - \mathbf{AG}(\mathbf{B}' \otimes \mathbf{C}')\|^2 \qquad (A.2) $$

The subscripts are omitted for simplicity unless the clarification is needed. A superscript t is then

needed for transpose of a matrix. The loss function f will be minimized by searching for an approximate factorization $\bar{\mathbf{X}}$. For fixed **A**, **B**, and **C**, **G** can be estimated by eq. (9),

$$\hat{\mathbf{G}} = \mathbf{A}'\mathbf{X}(\mathbf{B} \otimes \mathbf{C}) \tag{A.3}$$

A new function, u, can be obtained by substituting (A.3) into (A.2),

$$\begin{aligned} u(\mathbf{A}, \mathbf{B}, \mathbf{C}) &= \|\mathbf{X} - \tilde{\mathbf{X}}\|^2 \\ &= \|\mathbf{X} - \mathbf{A}\mathbf{A}'\mathbf{X}(\mathbf{B} \otimes \mathbf{C})(\mathbf{B}' \otimes \mathbf{C}')\|^2 \\ &= \|\mathbf{X} - \mathbf{A}\mathbf{A}'\mathbf{X}(\mathbf{B}\mathbf{B}' \otimes \mathbf{C}\mathbf{C}')\|^2 \end{aligned} \tag{A.4}$$

The problem is sequentially modified to search for only **A**, **B**, and **C** in such a way that the loss function u is minimized. Unfortunately, u is a function of the cross-product term of unknown **A**, **B**, and **C**, and it is not possible to solve such nonlinear problems explicitly to attain a minimum with ordinary methods. The ALS approach is used to solve this problem. With ALS, each set of parameters is estimated in turn by applying least squares procedures holding the other parameters fixed. After all sets have been estimated once, the procedure is repeated until convergence.

To solve (A.4) with ALS, matrices **A**, **B**, and **C** are initialized to ${}^0\mathbf{A}$, ${}^0\mathbf{B}$, and ${}^0\mathbf{C}$, respectively (presuperscripts indicate the iteration times). In principle, ${}^0\mathbf{A}$, ${}^0\mathbf{B}$ and ${}^0\mathbf{C}$ could be arbitrary, but use of the eigenvector matrices produced through Tucker's method (see theory section) can optimize the whole procedure. A new ${}^1\mathbf{A}$ is obtained by minimizing u [eq. (A.4)] with fixed ${}^0\mathbf{B}$ and ${}^0\mathbf{C}$; a new ${}^1\mathbf{B}$ is obtained with ${}^0\mathbf{C}$ and the just computed ${}^1\mathbf{A}$ fixed; ${}^1\mathbf{C}$ is produced with ${}^1\mathbf{A}$ and ${}^1\mathbf{B}$. In general, the $(n+1)$th main iteration step is described as follow.

A substep

$${}^n\mathbf{M} = \mathbf{X}({}^n\mathbf{B}{}^n\mathbf{B}' \otimes {}^n\mathbf{C}{}^n\mathbf{C}')\mathbf{X}' \quad \text{with} \quad \mathbf{X} = {}_i\mathbf{X}_{(jk)} \tag{A.5}$$

$${}^{n+1}\mathbf{A} = {}^n\mathbf{M}{}^n\mathbf{A}({}^n\mathbf{A}'{}^n\mathbf{M}^2{}^n\mathbf{A})^{-1/2} \tag{A.6}$$

B substep

$${}^n\mathbf{P} = \mathbf{X}({}^n\mathbf{C}{}^n\mathbf{C}' \otimes {}^{n+1}\mathbf{A}{}^{n+1}\mathbf{A}')\mathbf{X}' \quad \text{with} \quad \mathbf{X} = {}_j\mathbf{X}_{(ki)} \tag{A.7}$$

$${}^{n+1}\mathbf{B} = {}^n\mathbf{P}{}^n\mathbf{B}({}^n\mathbf{B}'{}^n\mathbf{P}^2{}^n\mathbf{B})^{-1/2} \tag{A.8}$$

C substep

$${}^n\mathbf{Q} = \mathbf{X}({}^{n+1}\mathbf{A}{}^{n+1}\mathbf{A}' \otimes {}^{n+1}\mathbf{B}{}^{n+1}\mathbf{B}')\mathbf{X}' \quad \text{with} \quad \mathbf{X} = {}_k\mathbf{X}_{(ij)} \tag{A.9}$$

$${}^{n+1}\mathbf{C} = {}^n\mathbf{Q}{}^n\mathbf{C}({}^n\mathbf{C}'{}^n\mathbf{Q}^2{}^n\mathbf{C})^{-1/2} \tag{A.10}$$

Eqs. (A.6), (A.8), and (A.10) compute eigenvectors using the Bauer–Rutishauser method, which is more efficient for this case [6,15]. It has been shown [6,15] that the iteration will converge toward at least a local minimum u, so that a least squares solution will be obtained.

The following relationships may be useful for examining the residuals of fitting. Eq. (A.2) or (A.4) can be rewritten as

$$\begin{aligned} f'(\mathbf{A}, \mathbf{B}, \mathbf{C}) &= \sum_i \sum_j \sum_k (x_{ijk} - \tilde{x}_{ijk})^2 \\ &= \sum_i \sum_j \sum_k x_{ijk}^2 - \sum_i \sum_j \sum_k \tilde{x}_{ijk}^2 \end{aligned} \tag{A.11}$$

This equation can be expressed with a conventional statistical notation, sum of squares (SS).

$$\mathrm{SS(Res)} = \mathrm{SS(Total)} - \mathrm{SS(Fit)} \tag{A.12}$$

where

$$\mathrm{SS(Fit)} = \sum_m \sum_p \sum_q g_{mpq}^2 \tag{A.13}$$

For each mode,

$$\begin{aligned} &\mathrm{SS(Fit_I)} = \sum_m \lambda_m, \quad \mathrm{SS(Fit_{II})} = \sum_p \mu_p, \\ &\mathrm{SS(Fit_{III})} = \sum_q \nu_q \end{aligned} \tag{A.14}$$

where λ_ms, μ_ps, and ν_qs are the eigenvalues with the extent of the summation corresponding to the number of factors retained for each mode, respectively. $\mathrm{SS(Fit_I)}$ is an approximation of the amount of variation explained by the factors of a two-mode factor analysis on the first mode. $\mathrm{SS(Fit_I)}$ is a function of retained number of factors in mode 1. $\mathrm{SS(Fit_{II})}$ and $\mathrm{SS(Fit_{III})}$ have analogous meanings. The relationship among them is

$$\begin{aligned} \mathrm{SS(Fit)} &\le \min\{\mathrm{SS(Fit_I)}, \mathrm{SS(Fit_{II})}, \mathrm{SS(Fit_{III})}\} \\ &\le \mathrm{SS(Total)} \end{aligned} \tag{A.15}$$

The proofs of these relationships can be found in further technical references [7,15,16].

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