呼和浩特市大气气相和颗粒物中有机污染物的定性分析*

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摘要 选用聚氨基甲酸酯泡沫塑料(PUFP)吸附块和玻璃纤维滤膜(GF)构成的大气全态采样头,以内蒙古四子王旗草原为对照点,于冬夏两季捕集呼和浩特市(呼市)大气气相和颗粒物中的有机污染物。样品经提取分离后用色质联机(GC-MS)、气相色谱仪(GC)、液相色谱仪(LC)鉴定出3类(烃类、芳烃类、酞酸酯类)近80种有机污染物,对其中45种进行定量分析。结果表明呼市地区属于大气有机污染物高浓度地区。

关键词 大气污染,颗粒物,有机污染物。

近几年来,国内一些学者相继对大气中芳烃类有机污染物在气相和颗粒物上的分布规律[1]以及大气全态采样器[2]开展了研究。在此基础上,笔者对典型煤烟型污染城市呼和浩特市大气气相和颗粒物中的有机污染物(侧重于烃类、芳烃类、酞酸酯类)进行了系统的分析研究,确定了污染物种类、污染水平和污染来源,并探讨了污染物的气/固分布规律,化合物之间的相互关系及污染特点等。

1 实验

1.1 采样

(1)采样地点与采样时间 在呼市市内布5个采样点(优化定点):小召点(居民区)、糖厂点(工业区)、第四毛纺厂点(商业交通区)、公安厅点(文化区)、牧机所点(清洁区)。对照点选在距市区170km,人为污染源极少的四子王旗草原。于1990年1月和8月在市内5个点采集大气颗粒物样品,1990年1月、8月,1991年11月和1990年3月、8月分别在小召点和草原点采集大气全态样品。

(2)采样设备与采样方式 颗粒物样品采样设备为TH-1000型总悬浮颗粒物国际标准型大气采样器,采样高度约5m,流量1.2m³/min,24h连续采样,采集5d。大气全态样品采样设备为KB-120型大气采样泵加GP-Ⅱ型采样头[2],采样高度约5m,流量0.1m³/min,间歇式采样(采样30—40min,间隔40—60min),连续采集3d。吸附材料GF和PUFP块的预处理同文献[3]。

1.2 分析

(1)样品的提取和预分离同文献[3] 分离后的各级淋洗液均浓缩至0.5—1.0ml,用于上机测定(作GC-MS分析的样品不过柱分离)。

(2)仪器分析 结果见表1。

2 结果与讨论

2.1 定性鉴定结果

经仪器分析,计算机检索,谱图解析及标准样品核对,给出呼市和四子王旗草原大气有机污染物定性鉴定结果,见表2—3、图1—2。

2.2 讨论

(1)呼市大气气相样品中鉴定出烷烃类26种,苯系物15种,多环芳烃类17种,酞酸酯类2种,共60种有机物。颗粒物样品中鉴定出烷烃类24种,苯系物3种,多环芳烃类23种,酞酸酯类2种,共52种有机物。气相样品中低碳化合物所占比例较大,如GC-MS在气相样品中鉴定出的芳烃多为3环以下,而4环以上的芳烃均是在颗粒物中鉴定出来的。这说明呼市大气中3环以下

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表 1　仪器分析条件[1)]

仪器名称	测定项目	测定条件	备注
GC-9A 气相色谱仪	正构烷烃 (C_5—C_{33})	FID 检测器，OV-101 石英毛细管色谱柱(25m×φ0.2mm) 进样口和检测器温度 330℃，柱初温 100℃，保持 5min，以 4℃/min 速率升温至 280℃，保持 15min。载气 N_2 40ml/min。	采用混合外标法对所有样品进行定性测定。求出各烷烃对 C_{23} 烷的响应因子，以 C_{23} 烷为外标进行定量测定。
	酞酸酯类	FID 检测器，OV-101 石英毛细管色谱柱(25m×φ0.2mm)，进样口和检测器温度 300℃，柱温 280℃，载气 N_2 30ml/min。	用混合标样外标法对所有样品作定性、定量测定。
LC-5A 高压液相色谱仪	多环芳烃类	RF-530 荧光检测器，ODS 反相柱，柱温 35℃，流动相：甲醇加水 83∶17，流速 1.0ml/min。检测波长：激发 250nm，发射 400nm	同上
Finnigan-MAT 4510 型质谱仪	烃类，芳烃类 酞酸酯类等	SE-54 毛细管柱(30m×φ0.25mm)，EI 离子源，电离能 70eV，发射电流 0.25mA，EM 电子倍增管电压 1300V	对呼市冬季全态样品作定性测定。
GC-MiNi2 携带式气相色谱仪	正构烷烃 (C_6—C_9) 苯系物	SUPELCO-WAX10 毛细管柱，FID 检测器，此仪器可自动采样、自动测试，详细测定条件见文献[4]	用混合标样外标法对呼市冬季空气气相样品作定性、定量测定。

1)　使用的均为进口标样

表 2　呼和浩特市大气中鉴定出的有机化合物种类[1)]

序号	化合物名称	分子式	分子量	检出样品种类		鉴定方法					定量测定
				气态	颗粒物	GC-MS	GC-9A	GC-MiNi2	LC-5A	标样	
1	正己烷	C_6H_{14}	86	✓				✓		✓	✓
2	正庚烷	C_7H_{16}	100	✓				✓		✓	✓
3	正辛烷	C_8H_{18}	114	✓				✓		✓	✓
4	正壬烷	C_9H_{20}	128	✓				✓		✓	✓
5—23	正十五烷至正三十三烷	$C_{15}H_{32}$至$C_{33}H_{68}$	212—464	✓	✓		✓			✓	✓
24	2,2,3,4-四甲基戊烷	C_9H_{20}	128		✓	✓					
25	2,2,6-三甲基庚烷	$C_{10}H_{22}$	142	✓		✓					
26	2,5,6-三甲基辛烷	$C_{11}H_{24}$	156	✓		✓					
27	2,4,6-三甲基辛烷	$C_{11}H_{24}$	156		✓	✓					
28	3,7-二甲基壬烷	$C_{11}H_{24}$	156		✓	✓					
29	2,6-二甲基辛烷	$C_{10}H_{22}$	142		✓	✓					
30	2,2,6-三甲基辛烷	$C_{11}H_{24}$	156		✓	✓					
31	2,5,6-三甲基癸烷	$C_{13}H_{28}$	184	✓		✓					
32	苯	C_6H_6	78	✓			✓	✓		✓	✓
33	甲苯	C_7H_8	92	✓			✓	✓		✓	✓
34	1,4-二甲苯	C_8H_{10}	106	✓	✓	✓		✓		✓	✓
35	乙苯	C_8H_{10}	106	✓				✓		✓	✓
36	1,3-二甲苯	C_8H_{10}	106	✓				✓		✓	✓
37	1,2-二甲苯	C_8H_{10}	106	✓				✓		✓	✓
38	1,3,5-三甲苯	C_9H_{12}	120	✓	✓	✓		✓		✓	✓
39	丙基苯	C_9H_{12}	120	✓				✓		✓	✓
40	1,4-二乙基苯	$C_{10}H_{14}$	134	✓				✓		✓	✓
41	1,3-二乙基苯	$C_{10}H_{14}$	134	✓				✓		✓	✓
42	1,2-二乙基苯	$C_{10}H_{14}$	134	✓				✓		✓	✓
43	1,2,3-三甲基苯	C_9H_{12}	120	✓				✓		✓	✓
44	1,2,4-三甲基苯	C_9H_{12}	120	✓				✓		✓	✓
45	1-甲基-2-乙基苯	C_9H_{12}	120	✓		✓					
46	1-乙基-3-甲基苯	C_9H_{12}	120	✓		✓					
47	联苯	$C_{12}H_{10}$	154		✓	✓					

续表 2

48	萘	$C_{10}H_8$	128	✓		✓						
49	1-甲基萘	$C_{11}H_{10}$	142	✓		✓						
50	2-甲基萘	$C_{11}H_{10}$	142	✓		✓						
51	1,3-二甲基萘	$C_{12}H_{12}$	156	✓		✓						
52	苊烯	$C_{12}H_8$	152	✓		✓						
53	苊	$C_{12}H_{10}$	154		✓	✓						
54	芴	$C_{14}H_{10}$	178	✓	✓	✓						
55	氧芴	$C_{13}H_{10}O$	182	✓		✓						
56	蒽	$C_{14}H_{10}$	178	✓	✓	✓				✓	✓	✓
57	4-甲基芴	$C_{15}H_{12}$	192	✓		✓						
58	菲	$C_{14}H_{10}$	178	✓	✓	✓						
59	1-甲基菲	$C_{15}H_{12}$	192		✓	✓						
60	2-甲基菲	$C_{15}H_{12}$	192		✓	✓						
61	4-甲基菲	$C_{15}H_{12}$	192		✓	✓						
62	9-甲基菲	$C_{15}H_{12}$	192		✓	✓						
63	荧蒽	$C_{16}H_{10}$	202	✓	✓	✓						
64	芘	$C_{16}H_{10}$	202	✓	✓	✓				✓	✓	✓
65	2-苯基萘	$C_{16}H_{12}$	204		✓	✓						
66	二甲基菲	$C_{16}H_{14}$	206		✓	✓						
67	苯并[ghi]荧蒽	$C_{18}H_{10}$	226		✓	✓						
68	䓛	$C_{18}H_{12}$	228	✓	✓	✓				✓	✓	✓
69	苯并[a]蒽	$C_{18}H_{12}$	228		✓	✓						
70	苯并[a]芘	$C_{20}H_{12}$	252	✓	✓	✓						
71	苯并[e]芘	$C_{20}H_{12}$	252		✓	✓				✓	✓	✓
72	苯并[b]荧蒽	$C_{20}H_{12}$	252		✓	✓						
73	苯并[j]荧蒽	$C_{20}H_{12}$	252		✓	✓						
74	苯并[k]荧蒽	$C_{20}H_{12}$	252		✓	✓						
75	苝	$C_{20}H_{12}$	252	✓	✓	✓				✓	✓	
76	苯并[ghi]苝	$C_{22}H_{12}$	276	✓	✓	✓				✓	✓	✓
77	二苯并[ah]蒽	$C_{22}H_{14}$	278	✓	✓	✓				✓	✓	✓
78	酞酸二正丁酯	$C_{16}H_{22}O_4$	278	✓	✓	✓	✓				✓	✓
79	酞酸二异辛酯	$C_{24}H_{38}O_4$	390	✓	✓	✓	✓				✓	✓
合计				60	52	45	21	17		7	45	45

1) 64、68、70、75、76、77 号化合物仅在颗粒物样品中被 GC-MS 检出，

1—31 为烷烃类，32—47 为苯系物，48—77 为多环芳烃，78—79 为酞酸酯类

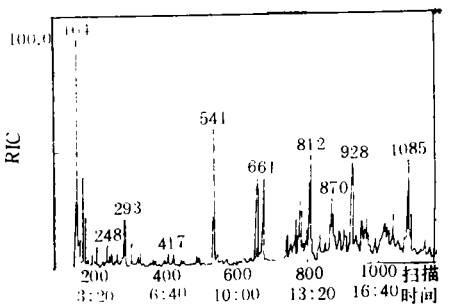

图 1　呼和浩特市冬季大气中气态有机污染物离子流色谱图

1,4-二甲苯(164),1-乙基-3-甲基苯(248),

1,3,5-三甲基苯(293),萘(541),2-甲基萘(661),

苊烯(812),氧芴(870),芴(928),菲(1085)

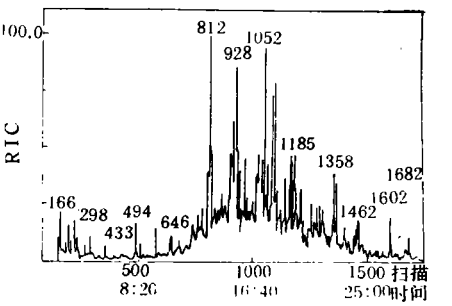

图 2　呼和浩特市冬季大气颗粒物中有机污染物离子流色谱图

1,4-二甲基苯(166),2,4,6-三甲基辛烷(298),

联苯(494),芴(646),菲(812),1-甲基菲(928),

荧蒽(1052),苯并[a]蒽(1358),苯并[j],荧蒽(1602)

表3　内蒙古四子王旗草原大气中鉴定出的有机化合物种类[1)]

序号	化合物名称	分子式	分子量	检出样品种类		鉴定方法			定量测定
				气态	颗粒物	GC—9A	LC—5A	标样	
	烷烃类								
1—19	正十五烷至正三十三烷	$C_{15}H_{32}$至$C_{33}H_{68}$	212—464	✓	✓	✓		✓	✓
	多环芳烃类								
20	蒽	$C_{14}H_{10}$	178	✓	✓		✓	✓	✓
21	芘	$C_{16}H_{10}$	202	✓	✓		✓	✓	✓
22	菌	$C_{18}H_{12}$	228	✓	✓		✓	✓	✓
23	苯并[a]芘	$C_{20}H_{12}$	252		✓		✓	✓	✓
	酞酸酯类								
24	酞酸二正丁酯	$C_{16}H_{22}O_4$	278	✓	✓	✓		✓	✓
25	酞酸二异辛酯	$C_{24}H_{38}O_4$	390	✓	✓	✓		✓	✓

1) 因采样条件所限，样品量不多，未能作GC-MS鉴定分析

的芳烃多存在于气相中，而4环以上的芳烃均存在于颗粒物中，LC定量检测的结果也证实了这一点，如蒽(3环)的平均浓度冬季在气相中为427ng/m³，颗粒物中为355ng/m³，夏季为40ng/m³和21ng/m³，无论冬夏季，均是气相中浓度高于颗粒物中浓度。而芘(4环)的平均浓度冬季为152ng/m³和626ng/m³，夏季为9ng/m³和34ng/m³，均是颗粒物中浓度高于气相中浓度。这与文献[1]所述基本一致，只是4环芳烃的气固比有所不同，这可能与呼市地区的气温偏低有关[5]。

(2)在检出的有机物中，除烷烃类对人体健康的影响尚待进一步研究外，其余几类污染物中的不少化合物均被证明具有致癌、致突活性(其中苯并[a]芘等至少4种多环芳烃为强致癌物)。笔者对其中45种进行了定量分析，结果表明，呼市地区空气中大部分有机污染物含量与已知数据地区比属于高浓度水平；冬季大气颗粒物中多环芳烃类浓度是太原的17倍，兰州的18倍，北京的20倍，广州的76倍，达到11209ng/m³，其中化合物芘所占的浓度比重最大，为60%以上；正构烷烃类浓度是北京和广州的3倍，达1981ng/m³；酞酸酯类也较北京地区高出3倍多。而就呼市本地区进行比较，冬夏两季在各功能区大气颗粒物中均定性定量检出19种正构烷烃，7种多环芳烃，2种酞酸酯；污染程度冬季以居民区最为严重，夏季是工业区较重。各功能区冬夏季有机污染物浓度的差别以居民区最大，冬季约是夏季的8—10倍，而工业区的季节差别不明显。尽管呼市地区夏季大气污染程度较冬季轻许多，但污染物浓度水平仍不低，颗粒物中多环芳烃类浓度仍达3245ng/m³，正构烷烃类浓度为747ng/m³也都远远高出北京、广州等地。人们长期生活在这样的大气环境中，对身体健康的危害是显而易见的。形成目前这种污染状况的主要原因是呼市地区的工业和民用能源结构以烧煤为主，而且大多是烧散煤。

(3)四子王旗草原冬夏季大气全态样品中2种酞酸酯、19种正构烷烃(C_{15}—C_{33})均被检出，多环芳烃类在气相中仅被检出较低环的蒽、芘、菌颗粒物中检出了蒽、芘、菌和苯并[a]芘。以上污染物中除正构烷烃类外(植物在生长过程中要释放出一部分烷烃类化合物)，芳烃类和酞酸酯类化合物的浓度水平均很低，并且冬夏2季差异不大[3,6]，说明该地区未受到人为污染，以上2类大气有机物浓度值可提供作环境背景参考值。

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Abstracts

residence time (HRT) of 40 h with an influent having a COD_{Cr} concentration of 1101mg/L, then the second stage of the A/O system and the biological activated carbon unit had their effluents with a COD_{Cr} concentration of 82 mg/L and 55 mg/L respectively, and a corresponding COD_{Cr} removal of 92% and 95%, respectively. When the pilot plant was operated at a HRT of 32 h with an influent having a COD_{Cr} concentration of 1112 mg/L, the second stage of the A/O system and the biological AC unit had their effluents with a COD_{Cr} concentration of 86 mg/L and 64 mg/L respectively, and a corresponding COD_{Cr} removal of 92% and 95%, respectively. Under the above conditions of operation, the second stage of the A/O system had a TA (total acid) removal of 93% for both cases.

Key words: A/O biofilm process, terephthalic acid wastewater, wastewater treatment, pilot plant.

Study on the Acid Hydrolysis-Anaerobic-Aerobic Fixed Biofilm System for the Treatment of Wastewater from Coking Plant. Shao Lin' guang et al. (Dept. of Environ. Eng., Wuhan College of Construction, Wuhan 430070): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 51—53

An acid hydrolysis-anaerobic-aerobic fixed biofilm system filled with a YDT elasto-steric packing was used to treat wastewater from coking plant. The results show that when the system was operated with an influent in which the concentrations of COD and NH_3-N were 1065 mg/L and 253 mg/L respectively, a total hydraulic residence time (HRT) of 33.5 h, and a mixed liquor recirculating ratio of 3.6, then there was an effluent with a COD concentration of about 180 mg/L and a NH_3-N concentration of 5 mg/L, namely that the removals of COD and NH_3-N were up to 83% and 98%, respectively.

Key words: acid hydrolysis-anaerobic-aerobic treatment system, fixed bed biofilm, wastewater from coking plant.

Study on the Treatment of wastewater from the Production Process of Vitamin C by Using a Complex Reactor of Upflow Anaerobic Sludge Blanket-Filter. Yang Jingliang et al. (Dept. of Environ. Eng., Hebei Institute of Light and Chemical Industries, Shijiazhuang 050018): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 54—57

A complex reactor of upflow anaerobic sludge blanket-filter (UASB + AF) with an available volume of $6m^3$ was used to treat a highly concentrated organic wastewater from the production process of Vitamin C. During the period of its stable operation, it had a volumetric COD loading of up to 10—12 kg COD/(m^3 · d), a COD removal of over 80%, and a volumetric biogas yield of over 3.0 $Nm^3/(m^3 \cdot d)$, as well as a high start-up speed and a stronger resistance to shock loading.

Key words: upflow anaerobic sludge blanket-filter reactor, wastewater, Vitamin C, synthetic fiber filter.

Feasibility Study on the Use of Several Devices for Sampling Natural Hydrocarbons. Bai Yuhua, Li Jinlong et al. (Dept. of Technical physics, Environ. Sci. Center, Peking University, Beijing 100871): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 58—62

The tests were conducted on the performances of several devices such as polished canister, steel tank, sampling bag and Tenax GC stainless steel tube for sampling HCs emitted from natural sources. It was found that the polished canister is well sealed and has a less wall effect. The Tenax GC tube has excellent performances in absorption and desorption. It is insufficient for steel tank and sampling bag to sample natural HCs over C_6 because of their serious wall absorption. However, the sampling bag is useful for sampling and saving HCs bellow C_6.

Key words: natural hydrocarbons, sampling devices, feasibility.

Determination of Polychlorinated Biphenyls (PCBs) in the Effluent from a Pulp Bleaching Process. Yang Chun, Yao Weixi (Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 63—65

Trace PCBs in the effluent from a pulp bleaching process with liquid chlorine or deactivated chlorinated lime were quantitatively determined with a method in which the samples were enriched by using a resin GDX-501, then extracted, washed with a concentrated sulfuric acid, and cleaned up over a deactivated alumina-silver nitrate-silica gel column before determination using a gas chromatography with a capillary and an electron capture detector. The PCBs in the effluent were found to be at a level in the range of 10^{-12}— 10^{-9} and to consist of dominant PCBs substituted with less (2 or 3) chlorines.

Key words: PCBs, pulp bleaching effluents, determination.

Qualitative Determination of Organic Pollutants in Atmospheric Vapor Phase and Particulate Phase in the Urban Area of Huhhot. Feng Shen' ying et al. (Inner Mongolia Center Station of Environ. Monitoring, Huhhot 010010): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 66—69

Full air samples were collected from the urban area

Abstracts

of Huhhot and the grassland belt in Siziwan Qi (a county), Inner Mongolia, as a control site in both winter and summer with a sampler using a polyurethane foam plug and a glass-fiber filter, both of which were then extracted with solvents. The extracts were analyzed with GC/MS, GC and HPLC to identify the organic pollutants in both vapor phase and particulate phase. Eighty eight pollutants in three kinds of organic compounds (n-alkanes, aromatic nydrocarbons and phthalate esters) were qualitatively identified and 45 of them were quantified. The results show that the Huhhot City is an area polluted with higher concentrations of organic pollutants in the atmosphere in its urban area.

Key words: atmospheric pollution, organic pollutant, vapor phase, particulate phase.

Study on the Assay of the Acute Toxicity and Mutagenicity of Industrial Wastewaters by Using the Bacterial Luminescence Test and the Ames Test. Huang Zheng and Wang Jialing (Institute of Environ. Medicine, Tongji Medicine University, Wuhan 430030): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 70—71

The industrial wastewaters from the Yijiadun and Huangxiaohe Wastewaters Irrigated Areas in Wuhan City were comparatively investigated for their acute toxicity and mutagenicity by using the bacterial luminescence test and the Ames test. At the same time, the composition of organic pollutants in the wastewaters was determined with the GC/ MS technique. The results show that, of four channels studied, the Luojiaqu Channel has the strongest acute toxicity and mutagenicity in its wastewater flow. The pollutants responsible for the acute toxicity of the wastewater mainly include phenol, benzyl benzoate, etc., and the pollutants responsible for the mutagenicity and carcinogenicity of the wastewater were found to be biphenyl, naphthalene. etc.

Key words: industrial wastewater, acute toxicity, mutagenicity, Gc/MS.

Validity of the Data from Automatic Air Quality Monitoring Systems. Fan Shaojia et al. (Dept. of Atmospheric Sciences, Zhongshan University, Guangzhou 510275): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 72—73

Based on the theory of statistics, a theoretical formula in which the acceptable missing hours and acceptable errors of validable daily averages are related to the variance of daily hourly averages has been derived. The monitored data on hourly concentrations of sulfur dioxide in winter from the Beijing Automatic Air Quality Monitoring System were taken to perform a calculation based on the derived formula and it was found that at a confidence of 95% the maximum random missing 6 hours were acceptable and in compliance with 75% of data captured as required by the National Environmental Protection Agency for a data validity check.

Key words: automatic air quality monitoring system, validity of monitored data, Beijing.

Homogeneous Reaction Mechanisms of Nitrous Oxide Formation and Decomposition in the Fluidized Bed of Coal Combustion. Chen Hongwei, Jin Baoshen et al. (Institute of Heat Energy, Southeast University, Nanjing 210018): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 74—78

The homogeneous reaction mechanisms of nitrous oxide formation and decomposition in the fluidized bed of coal combustion were studied, indicating that the paths of nitrous oxide (N_2O) formation from nitrogen contained in fuel mainly include: $NCO + NO \longrightarrow N_2O + CO$ and $NH + NO \longrightarrow N_2O + H$; and the main reaction of N_2O decompesition are: $N_2O + H \longrightarrow N_2 + OH$. An analysis was also made for the effects of the factors such as composition of fuel-bounded nitrogen compounds in volatiles, kinds of coal, temperature in the fluidized bed, coefficient of excess air, number of stages, and the combustion pattern in a single stage, on the emission of N_2O from the fluidized bed coal combustor.

Key words: fluidized bed combustion, coal, nitrous oxide, reaction mechanism.

Advance in the Study on Compounded Pollutions. He Yongtian and Xiong Xianzhe (Institute of Applied Ecology, Chinese Academy of Sciences, Shen' yang 110015): *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 79—83

Based on reviewing the current status of domestic and international research efforts in the field of compounded pullutions as a universal phenomenon of environmental pollution, this article dealt with the concepts and classifications of compounded pollutions, and the types of interactions of compounded pollutions and the criteria for their judgements, focusing on the factors affecting the ecological effects of compounded pollutions (i. e., pollutants factors, biological factors, and environmental factors) and the action mechanisms of compounded pollutions (i. e., influencing the structures of biotic cells, disturbing physiological activities and functions, competing for active sites, and complexing or chelating).

Key words: compounded pollutions, environmental pollution, ecological effects, review.