



ISSN 0250-3301 CODEN HCKHDV
HUANJING KEXUE

- 主办 中国科学院生态环境研究中心
- ■出版科学出版社



2019

Vol.40 No.3 第40卷 第3期

ENVIRONMENTAL SCIENCE

第40卷 第3期 2019年3月15日

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兰州市农牧业源氨排放清单及其时空分布特征

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摘要:通过实地调研等方式获取农牧业源的活动水平,采用 NARSES 模型确定氮肥施用排放因子,其它排放因子通过文献调研确定,建立了 2016 年兰州市农牧业源氨排放清单,并进一步分析了农牧业源氨排放的时空分布特征. 2016 年兰州市农牧业源大气氨排放量为9 356.90 t;其中畜禽养殖源氨排放量7 584.03 t,分担率81.05%;永登县是氨排放量最大的区县,氨排放量为2 820.59 t,分担率为 30.14%. 在兰州市各区县氨排放量分担率中,畜牧业源氨排放的分担率在65.83%~97.38%之间;氮肥施用源的分担率在2.27%~28.66%之间. 从空间分布来看,兰州市农牧业源氨排放主要集中在皋兰县西北部与中部、红古区东南部、七里河区东西两部与榆中县东部. 从时间分布来看,畜牧业源氨排放主要集中在4~9月,氮肥施用源的氨排放主要集中在3~7月和9月,其它月份排放量相对较小.

关键词:农牧业源; 氨排放清单; NARSES 模型; 时空分布; 兰州市

中图分类号: X51 文献标识码: A 文章编号: 0250-3301(2019)03-1172-07 DOI: 10.13227/j. hjkx. 201807190

Inventory and Spatiotemporal Distribution of Ammonia Emission from Agriculture and Animal Husbandry in Lanzhou City

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Abstract: After obtaining data on the activity of agriculture and animal husbandry by field investigation and from statistical data, an inventory of ammonia emission from agriculture and animal husbandry in Lanzhou was established based on literature investigation to determine the emission factors. The emission factor related to the application of nitrogen fertilizer was calculated using the NARSES model. The spatiotemporal distribution of the amount of ammonia emission was also analyzed. The total amount of ammonia emission from agriculture and animal husbandry was 9 356. 90 t. Among this emission, the amount from livestock and poultry was 7 584. 03 t, accounting for 81.05%. Yongdeng County was the region with the largest amount of ammonia emission, which was 2 820. 59 t, accounting for 30. 14%. Considering the sharing rate of ammonia emissions in all districts and counties, the sharing rates of livestock and poultry were between 65. 83% and 97. 38%; and the sharing rates for nitrogen fertilizer application were between 2. 27% and 28. 66%. From the perspective of spatial distribution, the sources of ammonia emissions from agriculture and animal husbandry were mainly concentrated in the northwest and central parts of Gaolan County, the southeast of Honggu District, the east and west of Qilihe District, and the east of Yuzhong County. From the perspective of temporal distribution, the ammonia emissions from livestock and poultry were higher from April to September. The emission from nitrogen fertilizer application was higher from March to September, except in August.

Key words: agriculture and animal husbandry source; ammonia emission inventory; NARSES model; spatiotemporal distribution; Lanzhou City

氨是大气中参与氮循环的主要成分^[1]. 在水体、土壤、大气等环境的复杂系统中有重要作用. 氨作为碱性气体,能够与大气中的 NO_{*} 、 SO_{2} 等酸性气体发生中和反应生成的二次无机气溶胶是大气中 $PM_{2.5}$ 重要组成部分^[2]. 研究表明,在欧洲铵盐占 PM_{10} 和 $PM_{2.5}$ 质量的 30%以上^[3];在美国一些地区则占到 $PM_{2.5}$ 质量的 47% ^[4];在我国部分地区可以占 PM_{10} 和 $PM_{2.5}$ 质量的 40% ^[5,6].

农牧业源作为氨排放贡献最大的子源,在我国人为源氨排放中平均占比可高达80%以上^[7-9].在国内已建立的部分氨排放清单中^[10,11],针对农牧业源的活动水平划分较粗,且未考虑本地气象条件与环境条件对氨挥发的影响,致使计算结果与实际

偏差较大. 兰州市是细颗粒物污染严重的城市之一, 其中铵盐的 PM_{2.5} 贡献率高达 37%^[12]. 国务院在颁布的打赢蓝天保卫战三年行动计划中指出, 在未来要提高化肥利用率, 强化畜禽粪污资源化利用, 减少氨排放^[13]. 因此在本研究中, 在更为细致划分活动水平的基础上, 通过实地调查等方式获取活动水平, 并结合合理的估算模型及文献调研的排放因子, 建立了兰州市 2016 年农牧

收稿日期: 2018-07-23; 修订日期: 2018-09-12

基金项目: 半干旱气候变化教育部重点实验室(兰州大学)开放课题 基金项目; 中央高校基本科研业务费专项(lzujbky-2017kb02)

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业源氨排放清单,并进一步分析了氨排放的时空分布特征.通过对兰州市二次颗粒物污染形成机制研究提供基础数据,以期为制定科学合理的氨减排方案提供决策依据.

1 材料与方法

1.1 研究区域及对象

本研究以 2016 年为基准年, 研究区域为兰州市六区三县. 研究的对象包括畜牧业、氮肥施用、

土壤本底与秸秆堆肥排放这4大类农牧业源,具体如表1所示.

1.2 估算方法

本研究利用活动水平和排放因子,采用"自下而上"的方法计算农牧业氨源排放总量. 计算公式为:

$$E_{i,j} = A_{i,j} \times \mathrm{EF}_{i,j} \tag{1}$$

式中, E 为排放量, A 为活动水平, EF 为排放因子, i 为地区, i 为排放源类.

表 1 氨排放源分类

Table 1 Classification of ammonia emission sources

一级排放源	二级排放源	三级排放源			
	畜牧业	肉牛、乳牛、肉猪、母猪、山羊、绵羊、肉鸡鸭鹅、蛋鸡鸭鹅、马驴骡			
农牧业	氮肥施用	尿素、碳铵、磷酸二铵、三元素复合肥, 其它氮肥			
水	土壤本底	- 00			
	秸秆堆肥	-			

1.2.1 畜牧业源

各阶段总铵态氮的活动水平依据文献[14]进 行计算. 2016 年畜禽年内饲养量来源于兰州市农委 农业数据平台[15],对于饲养周期大于1 a 的牛、 羊、母猪和蛋禽,畜禽年内饲养量取年底存栏数, 对于饲养周期小于1 a 的肉猪、肉禽, 畜禽年内饲 养量取年内出栏数[16]. 单位畜禽排放量、含氮量、 铵态氮比例和存储过程中氮的损失参数来自文献 [14]; 散养畜禽排泄物在室内户外各占50%, 集约 化养殖畜禽排泄物在室内占 100%; 散养畜禽液态 粪肥比重为 11%, 集约化养殖畜类液态粪肥比重 为50%, 禽类液态粪肥比重为0; 粪肥用作生态饲 料的比重主要考虑集约化养殖过程, 牛、羊、猪、 鸡取值分别为 20%、20%、30%、50%, 其它畜禽 取值为 0. 兰州市位于干旱半干旱区, 放牧所需的 草原植被资源匮乏[17]. 实地调查发现母猪、肉猪、 母鸡、肉鸡、绵羊、山羊、肉牛与母牛养殖方式为 集约化养殖与散养, 马、驴和骡的养殖方式仅为散 养: 并且永登县、榆中县、皋兰县、新区的产业结 构、畜牧业发展水平等相似,而城关区、红古区、 七里河区、西固区、安宁区相似[18]. 因此, 本研究 选取红古区与榆中县的畜禽集约化养殖比例进行了 实地调查以代表其他区县集约化养殖比例,实际调 查结果如表 2 所示. 此外, 调查还发现母猪、奶牛 与蛋鸡因需要繁衍后代与生产, 所以养殖的温度不 低于 10℃;集约化养殖各类畜禽的温度不低于 10℃, 其他条件温度参考室外平均温度. 2016 年兰 州市日平均温度数据来源于中国气象局国家气候中 心[19]. 排放因子依据文献[14]计算不同养殖方式 的氨排放量.

表 2 红古区与榆中县畜禽集约化养殖比例/%

Table 2 Ratios of the intensive culture of livestock

in Honggu District and Yuzhong County/%

	畜禽种类	红古区	榆中县
	奶牛	45. 16	67. 33
	肉牛	13.38	11. 10
	肉猪	33. 10	42.61
	母猪	33.10	42. 61
	蛋鸡	28. 74	43. 84
	肉鸡	50.00	15. 96
)	山羊	28.53	58. 08
	绵羊	28. 53	58. 08
	<u> </u>	<u> </u>	

1.2.2 氮肥施用源

氮肥施用过程的氨挥发是肥料氮损失的一个重要途径. 氮肥的活动水平通过公式计算:

$$M_i = \sum_{j=1}^n (m_{i,j} \times S_j)$$
 (2)

式中, M_i 为j 类作物 i 类氮肥的施用折纯量总和, $m_{i,j}$ 为j 作物每亩 i 类氮肥施用折纯化量, S_j 为j 作物的播种面积.

$$A_{\mathfrak{MH}_{i}} = \frac{M_{i}}{\sum_{i=1}^{n} M_{i}} \times M \tag{3}$$

式中, $A_{\text{MR}i}$ 为 i 类氮肥的活动水平,M 为当年氮肥施用折纯量总和. $m_{i,j}$ 来源于全国农产品成本收益资料汇编 $^{[20]}$, S_j 与 M 来源于兰州市 2017 年统计年鉴 $^{[18]}$. $m_{i,j}$ 在甘肃地区仅统计了三大类主要作物的每亩氮肥施用折纯量,因此计算出的 M_i 低于实际氮肥的施用折纯量,不能用来代替活动水平,需要式(3)补充计算.

氮肥的排放因子利用 Misselbrook 等^[21]开发的 国家尺度模型 NARSES 估算,该模型根据田间实验 研究的数据确定不同氮肥的最大排放百分比,选取 土壤 pH 值、温度、降水、氮肥施用率以及土地利 用方式作为影响氮肥施用源氨排放的主要因素. 计 算公式如式(4)所示:

$$\begin{split} EF_{\text{MR}} &= EF_{\text{max}} \times RF_{\text{soil pH}} \times RF_{\text{landuse}} \times \\ &RF_{\text{rate}} \times RF_{\text{rainfall}} \times RF_{\text{temperature}} \end{split} \tag{4}$$

式中, EF_{gen} 是各氮肥的排放因子, EF_{max} 是各类型的最大排放因子,RF 是和每个影响因子相关的消减因子。 $RF_{\text{soil pH}}$ 为土壤酸碱性消减因子, RF_{landuse} 为土地利用消减因子, RF_{rate} 为氮肥施用率消减因子,

RF_{rainfall}为降水消减因子,RF_{temperature} 为温度消减因子.对于 RF_{landuse},本研究认为蔬菜、薯类、豆类、瓜类、草莓是矮作物,其它作物则认为是高作物,分别取 1 和 0.7,基肥需要深施使其与根系生长的主要土层混合^[22],取 0.2;追肥则以撒施、滴灌为主^[23],取 1.利用王激清等^[24]统计的 2004 年我国小麦与玉米的基肥和追肥用量计算两种作物的总基追比代表兰州市作物的基追比,最终取值如表 3 所示.

最终 NARSES 模型各参数的计算结果如表 4 所示.

表 3 我国小麦玉米基追肥用量、兰州市作物基追比、高作物比例与 RF_{landuse}

Table 3 Amount of basal fertilizer and topdressing fertilizer on wheat and corn in China, dressing

ratios of crops, ratios of high crops, and RF _{landuse} in Lanzhou City							
氮肥种类	小麦:	小麦×10 ⁴ /t		玉米×10 ⁴ /t		兰州市高	RE
须几个大	基肥	追肥	基肥	追肥	物基追比	作物比例/%	$\mathrm{RF}_{\mathrm{landuse}}$
尿素	82.00	208.90	75.70	331.40	1:3.43	48.10	0.59
碳铵	254.90	139.90	261.10	154.20	1:0.57	48.10	0.38
磷酸二铵	140.10	2.80	140.00	13.40	1:0.06	48.10	0.23
其它氮肥	171.90	12.50	203.00	33.30	1:0.12	48.10	0.25

表 4 NARSES 模型各参数计算结果

Table 4 Parameters calculated in the NARSES mode

氮肥种类	FF [21]	RF _{ooil nu} [25]	4 RF	RF[21]	RFi.g.n [19,26]	RF. [19,26]
/ 1 /	LT _{max}	son pii	landuse	4/ 6	rainian	temperature
尿素	0. 45	1.00	0. 59	0. 53	1.00	0.58
碳铵	0.04	1.00	0.38	1. 00	1.00	0.60
磷酸二铵	0.45	1.00	0. 23	0.40	1.00	0. 58
其它氮肥	0.04	1.00	0. 25	1. 00	1. 00	0. 60

1.2.3 秸秆堆肥源

本研究通过草谷比法^[27]估算活动水平,如式(5)所示.

$$A_{\text{fift}} = W_{\text{P}} \times S_{\text{G}} \times M_{\text{R}} \tag{5}$$

式中,活动水平为田间堆肥秸秆量农作物产量, W_P 为农作物产量, S_G 为草谷比, M_R 为田间堆肥秸秆量质量分数.其中农作物产量数据来源于兰州市

2017 年统计年鉴^[18];用作田间堆肥的秸秆质量分数来源于甘肃省"十三五"秸秆饲料化利用规划统计^[28],其取值与不同农作物草谷比见表 5,排放因子取 0.32 kg·t^{-1 [14]}.

1.2.4 土壤本底源

活动水平为 2016 年兰州市各区县的耕地面积^[15],排放因子取 180 kg·km^{-2[14]}.

表 5 草谷比与秸秆堆肥比例

Table 5 Ratios of straw to grain and straw compost

秸秆种类	小麦	玉米	油料	瓜类	秸秆堆肥比例/%
草谷比	1. 13	1. 32	2. 40	0. 10	21.57

1.3 时间分配方法

由于兰州市氨排放时间变化特征的相关资料较为缺乏,本研究仅对畜牧业源与氮肥施用源的氨排放进行时间变化特征的研究,气象资料选取 2016 年榆中站每日间隔 3h 的数据资料,榆中站位于兰州市郊区,其数据能更好地代表兰州地区畜禽养殖和氮肥使用的环境温度.根据经验公式(6)^[29]计算小时排放速率,进一步依据式(7)按月求和得到了兰州市畜牧业源的氨排放月变化系数,基于月变化系数对全年排放总量进行按月分配:

$$E_{\rm h} = \left(2.36^{\frac{T_{\rm h} - 273}{10}}\right) \times V_{\rm h}$$
 (6)

$$P_{\rm h} = E_{\rm h} / \sum_{1}^{2928} E_{\rm h} \tag{7}$$

式中, E_h 为畜牧业源某小时排放率; T_h 为某小时环境的温度(K); V_h 为某小时的风速($\mathbf{m} \cdot \mathbf{s}^{-1}$), P_h 为畜牧业源某小时排放率在全年排放率中的占比.

根据甘肃省每月施肥比例^[30]计算各类氮肥每月氨排放量,采用1.2.2节中的方法,利用公式^[26]对每月氨排放量进行订正,分析兰州市氮肥施用源

的氨排放时间特征:

$$RF_{temperature} = \frac{e^{\left[0.2197225 \times (t_{month} - t_{annual})/3\right]}}{3} \tag{8}$$

$$RF_{temperature} = \frac{e^{\left[0.1386 \times (\iota_{month} - \iota_{year})/3\right]}}{2}$$
 (9)

式中, t_{month} 和 t_{annual} 是每月和每年温度的当地平均值 ($^{\circ}$ C),是中国的年平均气温($^{\circ}$ C).尿素和磷酸二铵 的氨排放率用式($^{\circ}$ 9)进行订正,其它氮肥用式($^{\circ}$ 8).

1.4 空间分配方法

畜牧业源、氮肥施用源、秸秆堆肥源与土壤本 底源的氨排放属于面源.考虑畜禽养殖的区域多分 布于村庄,因此基于兰州市各区县农村居民点的分 布对其氨排放量进行空间分配;而对于秸秆堆肥 源、氮肥施用源与土壤本底源的氨排放的实际范 围,则基于兰州市耕地类型占土地总面积的比重,对各区县的氨排放量进行空间分配.

2 结果与讨论

2.1 兰州市农牧业源大气氨排放清单及空间分布

兰州市 2016 年农牧业源大气氨排放清单如表 6 所示,排放总量为9 356.90 t,畜牧业源是兰州市 氨排放量最大的农牧业源,排放量达7 584.03 t;其 次是氮肥施用源,排放量为1 353.32 t;秸秆堆肥和土壤本底的排放量为 419.55 t. 永登县是兰州市氨排放量最大的区县,排放量为2 820.59 t;其次是榆中县,排放量为2 562.16 t;然后为七里河区,排放量为1 034.22 t. 安宁区是兰州市氨排放量最小的区县,排放量为 23.18 t.

表 6 2016 年兰州市农牧业源排放清单/t

Table 6 Agriculture and animal husbandry ammonia emission inventory for the year 2016 in Lanzhou City/t

排放源	秸秆堆肥	土壤本底	畜牧业	氮肥施用	总排放量
城关区	0. 74	1. 96	163. 03	4. 35	170.08
七里河区	2. 87	18. 17	911. 17	102. 01	1 034, 22
西固区	2. 50	6. 54	478. 70	33. 83	521.57
安宁区	0.00	0.08	22. 57	0. 53	23. 18
红古区	5. 30	9,61	873. 80	84. 10	972.81
永登县 //	13. 63	134. 61	2 491. 36	180. 99	2 820. 59
皋兰县	7.37	35. 18	539. 78	136. 91	719. 24
榆中县	17. 95	123. 06	1 686. 78	734. 37	2 562. 16
兰州新区	3. 36	36. 62	416. 84	76. 23	533. 05
总排放量	53. 72	365. 83	7 584. 03	1 353. 32	9 356. 90

通过在 1.3.2 节中所述的空间分配方法, 将各 子源的氨排放量叠加到同一网格中, 进而得到兰州 市农牧业源氨排放空间分布(图1). 由图1可知, 兰州市农牧业源氨排放主要集中在皋兰县西北部与 中部、红古区东南部、七里河区东西两部与榆中县 东部,这4个区县农牧业源氨排放的分担率也较 大. 其中原因为:皋兰县农牧业资源主要分布在西 北部与中部省道周围;红古区与七里河区的畜禽饲 养量较大, 因此出现高值点; 榆中县西部的兴隆山 与马衔山平均海拔在2000 m^[31]以上,有兴隆山自 然保护区, 该区域没有农牧业活动, 因此氨排放主 要集中在农牧业资源丰富的东部. 西固区的氨排放 量不大, 但由于农牧业活动主要集中在西部与东南 部,因此西固区西部与东南部出现了氨排放高值 点;而城关区与安宁区的面积较小,农牧业活动较 少, 故这两个地区的排放量较低.

兰州市各区县氨排放量分担率见图 2.

从图 2 看出, 畜牧业源氨排放的占比在65.83%~97.38%之间, 其中安宁区的占比最大, 榆中县最小; 氮肥施用源的占比在2.27%~28.66%之间; 榆中县的占比最大, 安宁区最小; 市

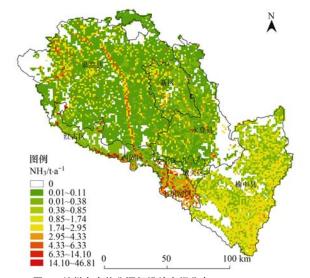


图 1 兰州市农牧业源氨排放空间分布(1 km×1 km)

Fig. 1 Spatiotemporal distribution of agriculture and animal husbandry ammonia emission of Lanzhou City($1~\rm{km} \times 1~\rm{km}$)

区种植业资源较少,难以开展种植活动,故氮肥施 用源分担率较小,畜牧业源分担率较大.

进一步分析第四级子源. 绵羊对大气氨排放量的贡献量最大,分担率为30.27%;各区县绵羊的氨排放占比在10.85%~54.31%之间. 主要因为绵

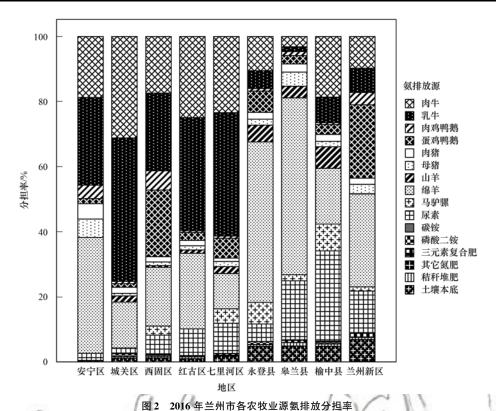


Fig. 2 Emission contribution of agriculture and animal husbandry sources by categoriy for the year 2016 in Lanzhou City

羊的需求量较大,年末存栏数较大;其次为肉牛,分担率为 15.84%;各区县肉牛的氨排放占比在 3.15%~31.23%之间. 尿素施用为第三大排放源,分担率为 13.60%;各区县尿素施用的氨排放占比在 1.64%~27.74%之间.

2.2 时间分布特征

兰州畜牧业源氨排放呈单峰分布,主要集中在4~9月,其他月份排放量相对较小(见图3).从1月开始,畜牧业源的氨排放量逐渐上升,在8月达到最大值,随后又开始下降,到1月排放量回落到最小值.兰州市氮肥施用源的氨排放主要集中在3~7月和9月,其它月份排放量均较小.由于畜牧业源氨排放受到温度的影响,因此可以看到春、冬季温度由于较低,不利于氨的挥发;随着气温的上升,氨的挥发也随之增大,故在夏季达到最大值.氦肥施用源的氨排放受到温度与种植活动的共同影响,因此可以看到氮肥施用源的氨排放在8月处于较低水平.除此之外,根据兰州市榆中站的气象数据显示,2016年兰州地面风速较小,故风速对畜牧业源氨排放的影响也较小.

2.3 不确定性分析

本研究中,不确定性主要来自于部分活动水平的缺失与排放因子的不确定.活动水平调查获取的过程中,对于畜牧业源,各区县的特种禽类未列人本清单中;在氮肥施肥部分中,作物的每亩氮肥/

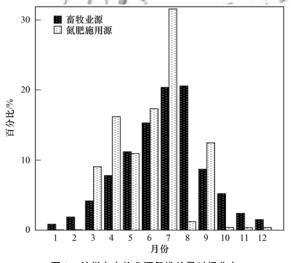


图 3 兰州市农牧业源氨排放量时间分布

Fig. 3 Time distribution of agriculture and animal husbandry ammonia emission of Lanzhou City

复合肥施用的折纯化量统计的作物种类较少,计算各类氮肥的活动水平时可能会导致相应的偏差;其余活动水平数据基于政府部门或统计年鉴,来源较为可靠.不确定性范围的量化需要获取大量与清单估算过程涉及数据类型一致^[29],而活动水平数据的来源单一有限,本研究不对其进行定量分析.基于蒙特卡洛不确定性传递,随机重复抽样50000次估算排放清单的不确定性大小,在95%的置信度下得到清单排放总量的不确定范围为-2.69%~39.67%.建议进一步研究将 NARSES 模型中的参

数进行本地化, 进而提高氨排放清单的准确性.

3 结论

- (1) 兰州市 2016 年农牧业源大气氨排放总量为 9 356.90 t; 其中永登县氨排放量最大为 2 820.59 t; 其次分别为榆中县、七里河区、红古区、皋兰县、新区、西固区与安宁区.安宁区氨排放量最小为 23.18 t. 畜禽养殖源是兰州市氨排放量最大的农牧业源,排放量达7 584.03 t; 其次是氮肥施用源,排放量为 419.55 t.
- (2)在各区县氨排放量分担率中,畜禽养殖源 氨排放的占比在65.83%~97.38%之间,其中安宁 区的占比最大,榆中县最小;氮肥施用源的占比在 2.27%~28.66%之间;榆中县的占比最大,安宁区 最小.从空间分布来看,兰州市农牧业源氨排放主 要集中在皋兰县西北部与中部、红古区东南部、七 里河区南北两部与榆中县东部.从时间分布来看, 畜牧业源氨排放主要集中在4~9月,氮肥施用源 的氨排放主要集中在3~7月和9月,其它月份排 放量相对较小.
- (3)绵羊对大气氨排放量的贡献量最大,分担率为30.27%;各区县绵羊的氨排放占比在10.85%~54.31%之间.其次为肉牛,分担率为15.84%;各区县肉牛氨排放占比在3.15%~31.23%之间.尿素施用为第三大排放源,分担率为13.60%;各区县尿素施用的氨排放占比在1.64%~27.74%之间.

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Environmental Science (monthly)

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