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我国城市饮用水中 *N*-亚硝基二甲胺的健康风险评估及水质标准制定

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摘要: 亚硝胺类物质, 特别是亚硝基二甲胺 (NDMA), 其高致癌性及在我国饮用水中高检出率引起了国内媒体和管理层的广泛关注. 为了评价饮水中 NDMA 造成的健康风险, 进而提出我国饮水标准的建议, 本文利用近几年全国饮用水水质调查的 NDMA 数据, 以伤残调整寿命年 (DALYs) 为风险评价终点, 并结合疾病模型, 对饮水途径摄入 NDMA 造成的健康风险进行估算. 结果表明, 我国城市饮用水中由于 NDMA 造成的终身癌症发病率为 5.69×10^{-6} , 人均 DALYs 损失为 6.27×10^{-7} 人·a⁻¹. 以 WHO 所推荐的风险可接受水平 (DALYs) (10^{-6} 人·a⁻¹) 来制定饮用水中 NDMA 浓度安全标准, 应为 $6.12 \text{ ng} \cdot \text{L}^{-1}$. 考虑到我国水厂工艺情况, 未来 NDMA 的安全标准设在 $6 \sim 40 \text{ ng} \cdot \text{L}^{-1}$ 范围内更合理. 最终的水质标准仍需进一步考虑其他经济和技术水平等因素.

关键词: *N*-亚硝基二甲胺; 饮用水安全; 伤残调整寿命年; 健康风险; 水质标准

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Estimation of Health Risk and Enaction of Safety Standards of *N*-nitrosodimethylamine (NDMA) in Drinking Waters in China

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Abstract: Nitrosamines such as nitrosodimethylamine (NDMA) in drinking water have recently attracted great attention because of their high carcinogenicity and high detection rate. Nitrosamines have also been repeatedly detected in drinking water in our country, leading to a lot of concerns about our drinking water safety. However, China has not yet formulated the relevant drinking water safety standards. In order to evaluate the health risks caused by NDMA in drinking water and to provide recommendations for the development of drinking water safety standards, the method of disability-adjusted life years (DALYs) and the two-stage disease model were used to estimate the health risk of liver cancer caused by intake of NDMA in drinking water. The data of this study were collected from two large-scale water quality surveys conducted in 35 cities in China from November 2009 to May 2012, and the detection conducted by Chen Chao *et al.* in 23 cities in China from 2012 to 2014, with a total of 146 water plants data. The results showed that mean ($8.97 \text{ ng} \cdot \text{L}^{-1}$) and median ($2.90 \text{ ng} \cdot \text{L}^{-1}$) NDMA concentrations were both not very high except in some special areas. The incidence of life-long cancer was 5.69×10^{-6} and 5.69 times as high as the negligible risk value (1×10^{-6}) specified by the US EPA. The total disease burden of NDMA was 844.15 person-years, of which the death loss was 818.31 person-years, accounting for 96.9%. The incapacity loss was 25.84 person-years, accounting for 3.1% in comparison. Death loss was greater than the loss of incapacity. The disease burden was highest in the age group of 55-60 years (129.40 person-years), followed by 45-50 years (120.44 person-years). The burden of disease was higher in middle-aged and elderly people. The averaged loss was 6.27×10^{-7} DALYs per person per year in our country. Only considering the health risk factors, NDMA concentration safety standards should be $6.12 \text{ ng} \cdot \text{L}^{-1}$. According to the specific national conditions, the NDMA safety standard in the range of $6 \sim 40 \text{ ng} \cdot \text{L}^{-1}$ was recommended. On one hand, we can control the concentration of NDMA in drinking water, to reduce health risks as far as possible, and on the other hand, we can also ensure the effectiveness of disinfection of drinking water treatments. China's economic and water treatment technological level and other factors should also be taken into consideration in the near future. In view of potential health risks of NDMA, it's necessary to adopt more effective, economical and also environmental water treatment techniques and develop reasonable safety standards to ensure the quality of drinking water and people's health.

Key words: *N*-nitrosodimethylamine (NDMA); drinking water safety; disability-adjusted life years (DALYs); health risk; drinking water safety standards

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亚硝胺类物质 (NAs) 由于具有强致癌效力, 并在饮用水和废水中频繁检出, 被多个国家列为法定监测指标^[1]. 近些年, 我国也曾经多次在水体中检测出亚硝胺^[2-6], 特别是最近清华大学对我国饮用水系统中亚硝胺类消毒副产物进行的普查, 引起了国内媒体和管理层的关注. 中国是世界上亚硝胺检出情况最多样的国家, 其中 *N*-亚硝基二甲胺 (NDMA) 的浓度最高. 亚硝胺在中国出厂水和龙头水中的检出率是美国环保署在 2012 年公开的一项大规模普查结果的 3.6 倍, 而西欧国家的饮用水亚硝胺浓度处于较低水平^[7]. NDMA 的毒性极强, 被国际癌症研究机构 (IARC) 认证为 2A 级^[8], 被美国环境保护署综合风险信息系统 (US EPA IRIS) 列为 B2 级^[9]. 饮水中若存在 NDMA, 即使浓度很低, 也有产生遗传毒性的风险^[10]. 流行病学研究已经表明, NAs 污染严重的饮用水与消化系统的癌症密切相关^[11-13].

亚硝胺已被列为饮用水中重要的消毒副产物之一, 特别是 NDMA, 但是我国尚缺乏饮用水中的管理标准. 目前部分发达国家和地区建立了饮用水中 NDMA 的标准. 世界卫生组织在 2006 年提出了 100 ng·L⁻¹ 的推荐值, 加拿大、澳大利亚都有了国家标准, 分别是 40 ng·L⁻¹、100 ng·L⁻¹, 加拿大安大略省、美国麻省和加州的标准更加严格, 分别是 9、10、10 ng·L⁻¹^[14-17]. 虽然我国饮用水中部分地区亚硝胺检出情况很严重, 由于缺乏对饮用水中亚硝胺类物质进行的风险评估, 至今尚未制定相关水质标准.

健康风险评价过程中, 伤残调整寿命年 (DALYs) 常被选作风险评价的终点. 该指标以时间作为度量单位, 综合考虑从患病到死亡所损失的全部健康寿命年, 能够对致癌性和非致癌性的污染进行风险定量和比较, 便于进行优先控制污染物及标准制定^[18].

本研究根据近 7 年来在重点城市的水厂进行的水质连续调查中 NDMA 的浓度数据, 评估导致肝癌的发病率, 以 DALYs 作为健康风险评价的终点, 计算饮水途径摄入 NDMA 导致的疾病负担, 并提供水质标准所存在的合理范围, 以期为我国饮用水水质标准提供支持.

1 材料与方法

1.1 数据来源

本研究的数据来源于 2009 年 11 月 ~ 2012 年 5

月在我国 35 个城市进行的两次大规模水质调查, 以及陈超等人 2012 ~ 2014 年对我国 23 个省市进行的水体亚硝胺检测^[7], 共计 146 个出厂水 NDMA 浓度数据. 采样地点、水样采集、准备、分析及质量控制等细节可以参考文献^[7, 19].

1.2 暴露评价

根据 NDMA 的浓度数据进行暴露参数分布拟合. 由于目前没有充分的证据认为呼吸和皮肤摄入是其致癌的重要途径, 所以只考虑饮水暴露途径^[20, 21]. 慢性每日摄入量 (chronic daily intake, CDI) 即个体终生暴露于某种致癌物质下单位时间单位体重的平均日摄入量. 饮水暴露途径的 CDI 可以通过公式 (1) 计算^[22]:

$$CDI = \frac{c_w \times IR \times EF \times ED}{BW \times AT} \quad (1)$$

式中, c_w 表示饮用水中 NDMA 的浓度 (ng·L⁻¹), IR 表示饮水量 (L·d⁻¹)^[23], BW 表示人体体重 (kg)^[24], AT 表示平均暴露时间 (d)^[24], EF 表示暴露频率 (d·a⁻¹)^[22], ED 表示暴露时长 (a)^[24].

在整个风险评价过程中, 采用蒙特卡洛模拟软件 (Oracle® Crystal Ball 11. 1. 1. 3. 00) 进行随机模拟, 并进行不确定性分析.

1.3 疾病终点的确定

自来水的消毒过程是饮用水中 NDMA 的主要来源^[25]. 目前, NDMA 导致的人类癌症仍缺乏相关的流行病学数据积累, 因此采用动物毒性数据来确定致癌的强度. NDMA 会引起小鼠肝癌发病率的升高^[26], 所以将肝癌确定为疾病终点.

1.4 终身癌症发病率的计算

将慢性每日摄入量乘以致癌斜率因子 (slope factor, SF) 可以获得 NDMA 饮水暴露途径导致的癌症风险 (终身癌症发病率) CR:

$$CR = CDI \times SF \quad (2)$$

SF 是使用各种模型拟合动物毒性数据估计得出的癌症效能的 95% 置信区间的上限值^[27]. 因此, CR 可以被认为是人类终身暴露于致癌物发生癌症的概率的上限值^[28]. NDMA 的致癌斜率因子采用 IRIS 中的数据, 即 51 (kg·d)·mg⁻¹^[29].

1.5 疾病模型及 DALYs 的计算

为评估饮用水中 NDMA 的疾病负担, 将肝癌发病率估计结合 WHO 设计的两阶段疾病模型^[28, 30] 计算 DALYs. 疾病模型是整个疾病历程的简化. 癌症一般会带来两种结果, 即死亡或存活 (可能伴有后遗症), 疾病历程如图 1 所示, 需要分别考虑相应的

死亡损失或失能损失.

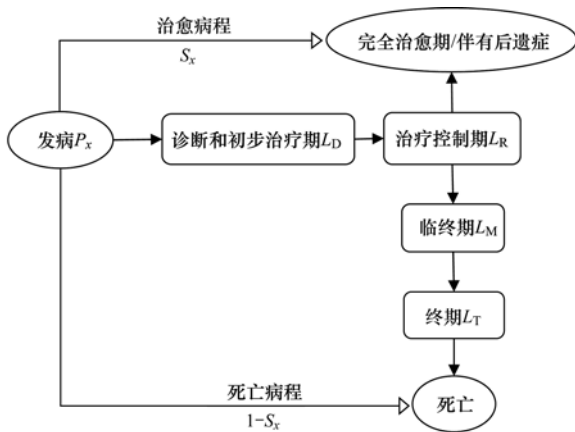


图1 两阶段疾病模型

Fig. 1 Two-stage disease history model

死亡损失 (years of life lost, YLLs) 和失能损失 (years of lived with disability, YLDs) 的计算公式如式 (3) 和式 (4) 所示^[31], DALYs 是两部分损失之和.

$$YLLs = \sum_x n_x P_x (1 - S_x) (e_x^* - T_D) \quad (3)$$

$$YLDs = \sum_{x,y} n_x P_x [(1 - S_x) DW_y L_y + S_x DW_y L_y] \quad (4)$$

$$DALYs = YLLs + YLDs \quad (5)$$

其中, x 即年龄阶段 (划分为 18 个年龄组: 0~5, 5~10, 10~15, ..., 80~85, 85 及以上); y 即肝癌不同的疾病阶段; n 即人口数, 基于 2011 年我国人口结构的调查数据^[32]; P 即肝癌发病率, S 即肝癌生存率, 利用我国癌症登记地区的肝癌年龄别发病率和死亡率计算^[33]; e^* 即标准期望寿命, 基于我国模型寿命表^[34]; T_D 即肝癌死亡经历的病程时间^[35]; DW 即失能权重^[28]; L 即肝癌病程持续时间, 划分为诊断和初步治疗期 (L_D)、治疗控制期 (L_R)、临终期 (L_M) 和终期 (L_T)^[31]. 计算过程和参数值参考文献 [28].

2 结果与分析

2.1 NDMA 的浓度分布

本研究共计 146 个出厂水 NDMA 浓度数据. 浓度范围为 ND ~ 162.9 ng·L⁻¹, 平均值为 8.97 ng·L⁻¹, 中值为 2.90 ng·L⁻¹. 浓度数据的最佳拟合分布为对数正态分布, 拟合参数为 Ln(1.37, 0.55), 如图 2 所示.

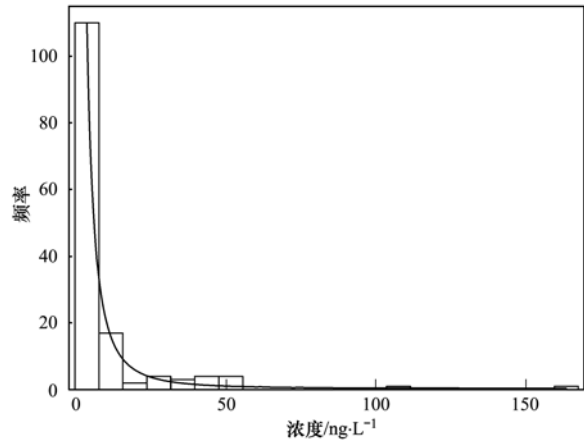


图2 NDMA 浓度拟合分布

Fig. 2 Distribution plot of NDMA concentration

2.2 NDMA 导致的终身癌症发病率

NDMA 的致癌斜率因子高达 51 (kg·d)·mg⁻¹, 因此, 在 NDMA 浓度较高的水体里, 其致癌风险值得关注. 通过计算得到, NDMA 造成的肝癌发病率中值为 5.69 × 10⁻⁶, 为美国 EPA 规定的癌症的可接受水平 (1 × 10⁻⁶) 的 5.69 倍^[36], 其 5% 及 95% 分位数分别为 0.68 × 10⁻⁶ 和 49.96 × 10⁻⁶. 采用蒙特卡洛模拟的分布及累积分布如图 3 所示.

2.3 NDMA 导致的肝癌 DALYs 风险

将肝癌发病率中值代入疾病模型, 计算年龄别 DALYs, 结果如图 4 所示. 总的死亡损失和失能损

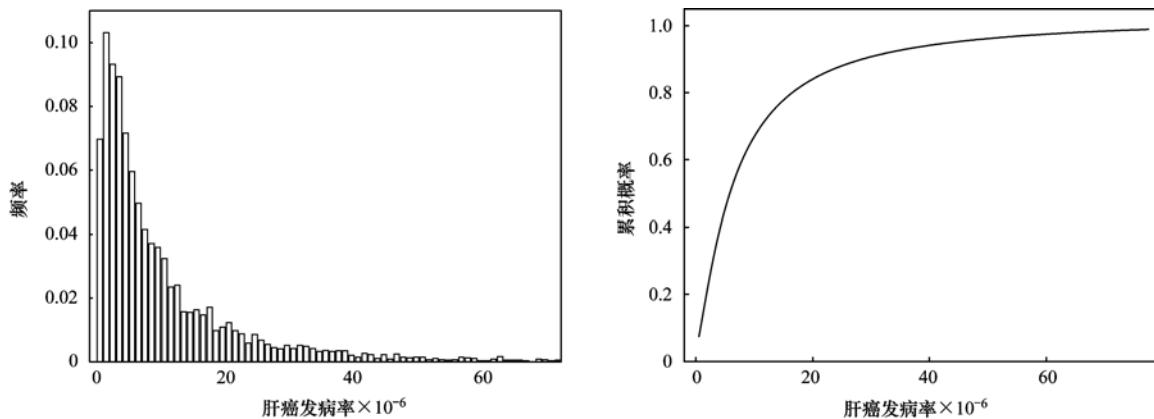
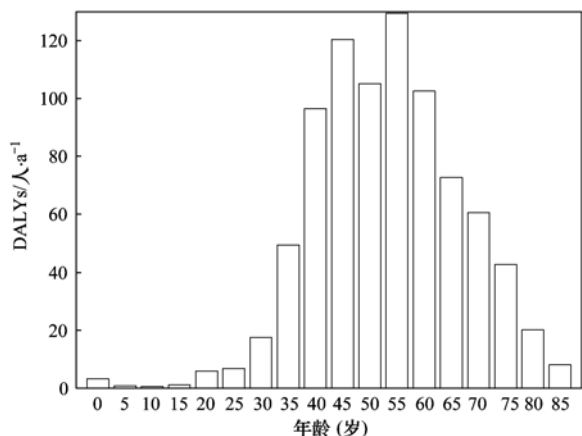


图3 NDMA 导致的肝癌发病率的分布及其累积分布

Fig. 3 Distribution plot and cumulative probability distribution of incidence of liver cancer caused by NDMA

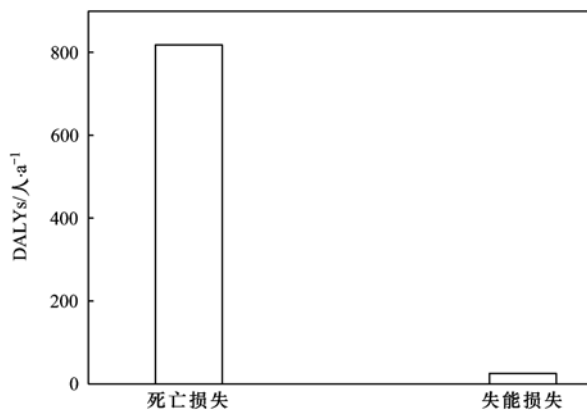
失的对比如图 5 所示. 将各年龄组的 DALYs 损失相加并除以总人口数可以获得人均 DALYs 损失.



基于肝癌发病率中值

图 4 年龄别疾病负担

Fig. 4 Age-specific DALYs losts



基于肝癌发病率中值

图 5 死亡损失和失能损失对比

Fig. 5 Comparison of YLLs and YLDs

NDMA 导致的肝癌总疾病负担为 844.15 (100.88, 7 411.94) 人·a⁻¹, 人均疾病负担为 6.27 × 10⁻⁷ (7.49 × 10⁻⁸, 5.50 × 10⁻⁶) 人·a⁻¹, 其中死亡损失为 818.31 (97.79, 7 185.04) 人·a⁻¹, 占 96.9%, 失能损失为 25.84 (3.09, 226.90) 人·a⁻¹, 占 3.1% (括号中的数值表示其 5th 分位数和 95th 分位数). 肝癌死亡率高, 而治愈率不足 5%^[33], 因此死亡损失明显大于失能损失. 疾病负担最大的年龄组为 55~60 岁 (129.40 人·a⁻¹), 其次是 45~50 岁 (120.44 人·a⁻¹), 中年和老年的疾病负担较高, 这部分人群是饮水摄入 NDMA 造成健康风险的高危人群, 应特别注意防范. WHO 的《饮用水水质准则》中规定疾病负担的参考水平为 10⁻⁶ 人·a⁻¹^[37], 而 NDMA 导致的肝癌人均 DALYs 损失没有超过该参考水平. 据 2010 年全球疾病负担报道, 我国肝癌

人均 DALYs 损失为 659.7 × 10⁻⁵ 人·a⁻¹, 因此本研究估计的 NDMA 造成的 DALYs 损失不足其万分之一.

3 讨论

我国饮水中 NDMA 浓度的中值和均值不高, 导致的肝癌人均 DALYs 损失也处于较低的水平. 实际上, 亚硝胺的主要来源是腌制食品和烟草等非饮用水^[37]. 我国内蒙巴盟河套地区的调查显示, 当地腌肉和酸菜中的亚硝胺含量可达 80 000 ng·kg⁻¹, 是水中亚硝胺平均浓度的几千倍^[38]. 另外, 在火腿中检测出的亚硝胺类化合物平均含量达到 8 500 ng·kg⁻¹, 咸鱼及其制品普遍在 3 000 ng·kg⁻¹ 以上^[37,39]. 所以 NDMA 在不同途径的摄入风险仍需进一步研究. 虽然我国集中供水的 NDMA 健康风险整体水平较低, 但局部地区水厂的 NDMA 浓度较高, 最高的甚至达到了 100 ng 以上^[7], 将可能造成 1.48 × 10⁻⁵ 的肝癌发病率以及 1.63 × 10⁻⁶ 人·a⁻¹ 的人均 DALYs 损失, 超过了 WHO 规定的疾病负担参考水平, 须引起关注.

我国至今还没有提出关于 NDMA 的饮用水水质安全标准. WHO 表示, 在一个不吸烟的家庭中, 饮用水暴露途径的慢性每日摄入量为 0.000 3 ~ 0.001 μg·kg⁻¹, 而食物途径为 0.004 3 ~ 0.011 μg·kg⁻¹, 饮水摄入量在总摄入量中所占比例低于 10%^[40], 并利用动物实验数据的 TD₀₅ (Tumorigenic Dose 05, 引起 5% 癌变的剂量) 在癌症发病率为 10⁻⁵ 水平下计算, 推荐 NDMA 的饮用水安全值为 100 ng·L⁻¹^[41], 仅考虑了危害的响应级别. 加拿大安大略省及美国加州政府环境健康危害评估处 (OEHHA) 基于癌症发病率为 10⁻⁶ 的水平, 分别制订了 10 ng·L⁻¹ 的 NDMA 安全浓度, 考虑了当前非常低的检测限及饮用水处理去除 NDMA 的效果, 所以标准更为严格^[42,43]. 我国在具体制定本国标准时还需结合自己的国情. WHO 饮用水水质准则中指出, 10⁻⁶ 人·a⁻¹ 的风险参考水平基本等同于 10⁻⁵ 的终身癌症发病率^[44]. 以 WHO 所推荐的风险参考水平 (DALYs, 10⁻⁶ 人·a⁻¹) 来制定我国饮用水中 NDMA 浓度安全基准, 应为 6.12 ng·L⁻¹. 所以从降低健康风险的角度可以首先考虑把 6 ng·L⁻¹ 设置为 NDMA 水质安全标准的下限. 但是若仅依此制定标准则会导致约 30% 水厂超标.

消毒是保障水质安全必不可少的环节^[45], 饮用水中的 NDMA 主要是在氯消毒的过程中产生的.

NDMA 的去除技术有紫外照射、臭氧氧化、高级氧化、生物降解、物理吸附、反渗透膜等^[46],其中紫外照射是目前应用最广泛的方法,去除率达 95% 以上,但紫外线对污水中多数的 NDMA 前体物基本没有去除效果,即在后续用氯消毒过程中仍然会有 NDMA 重新生成的可能性^[47],而且费用过高,去除 90% 的 NDMA 所需的 UV 剂量是灭活病毒和细菌所需剂量的 10 倍,因此紫外照射法去除 NDMA 可行但不经济^[45]. 臭氧去除的主要问题是效率太低, $7.7 \text{ mg} \cdot \text{L}^{-1}$ 的臭氧对 NDMA 的氧化率小于 25%,同时臭氧和高级氧化技术的去除率都受到羟基自由基的限制^[45]. 生物降解的机制尚不明确,且容易受到复杂环境因素的影响,去除效果很不稳定^[48]. 碳质活性炭是相对最佳的吸附剂,但吸附能力易受进水 NDMA 浓度的影响,当 NDMA 浓度大于 $50 \text{ ng} \cdot \text{L}^{-1}$ 时吸附能力最强^[48]. 反渗透膜的去除率为 56% 到 70%,但在实际运行过程中的膜污染会使去除率下降^[46]. 所以现阶段无法通过 NDMA 的去除技术直接大幅降低其浓度,达到较为严格的水质标准. 由图 6 可知,随着 NDMA 浓度标准的放宽,水厂超标率不断下降,但癌症风险不断增加. 当 NDMA 的浓度标准达到 $40 \text{ ng} \cdot \text{L}^{-1}$ 时,大约可以使 95% 以上的水厂达标,保证了正常的饮用水消毒处理,这种情况造成的人均 DALYs 损失为 6.52×10^{-6} ,与 WHO 的疾病负担参考水平(10^{-6})比较,致癌风险没有达到十万分之一的变化. 因此考虑到我国的水处理技术水平及经济条件限制,可以把 $40 \text{ ng} \cdot \text{L}^{-1}$ 作为 NDMA 水质安全标准的上限.

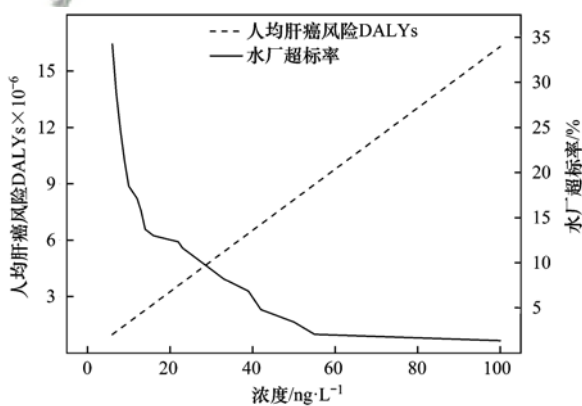


图 6 水厂超标率及人均肝癌风险

Fig. 6 Percentage of water treatment plants over standard and averaged liver cancer risk caused by concentration of NDMA

综上所述,认为 NDMA 的饮用水安全标准设为 $6 \sim 40 \text{ ng} \cdot \text{L}^{-1}$ 范围内更符合我国国情,一方面可以控制 NDMA 的浓度,尽可能减少健康风险,另一方

面也能够保证饮用水消毒处理的效果. 未来是否将 NDMA 等亚硝胺类物质纳入水质标准,以及如何制定标准是一个极具争议的话题,还须进行多领域、多学科的研究论证.

4 结论

我国集中供水的 NDMA 浓度整体不高,仅有局部地区的水厂浓度很高. 饮水途径摄入 NDMA 造成的肝癌发病率为 5.69×10^{-6} ,人均 DALYs 损失为 $6.27 \times 10^{-7} \text{ 人} \cdot \text{a}^{-1}$,整体风险水平不高且符合相关风险参考水平. 针对 NDMA 潜在的健康威胁,结合我国的水处理技术水平及经济发展状况,认为未来 NDMA 的水质标准设在 $6 \sim 40 \text{ ng} \cdot \text{L}^{-1}$ 范围内较为合理. 今后还须进一步进行相关研究,以控制 NDMA,保障我国饮用水安全.

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