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黑龙江双阳河流域土壤-作物-人体系统中硒元素及生态环境与人体健康评价

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摘要:【研究目的】典型黑土地分布区土壤硒自然禀赋、成因来源、生态环境效应及人体健康评价为健康地质研究焦点, 开展双阳河流域土壤-作物-人体系统中硒迁移与累积的定量研究, 对黑土地富硒农产品开发和地方病防治具有重要意义。【研究方法】测定成土母质、土壤、大气降尘、灌溉水、作物籽实和人发等多介质硒含量, 运用 GIS、SPSS 软件对测试结果统计分析及评价。【研究结果】划定富硒土地 46.4 km², 主要沿双阳河及其支流分布; 硒以人为成因为主, 主要来源于大气降尘补充, 输入通量为 0.21~1.47 kg/km²·a。土壤、大豆、水稻、玉米和人发硒含量均值为 0.305 mg/kg、0.0352 mg/kg、0.0287 mg/kg、0.0198 mg/kg 和 0.145 mg/kg; 作物对土壤硒的富集系数(0.120)远小于 1, 人发对作物硒的富集系数(4.834)大于 1, 揭示人发对硒具有较强的浓缩和放大作用, 硒在土壤-作物-人体系统中的迁移与积累呈“V”字型模式。【结论】土壤硒含量整体为足硒或富硒, 无硒潜在环境风险。大豆、水稻富硒占比为 28.4%、9.4%, 未发现硒超标情况, 无生物硒中毒风险。人发硒含量明显低于参考值 0.20~0.60 mg/kg, 人体处于缺硒状态, 随年龄增长呈递减趋势, 可能存在一定的健康风险, 应引起地方相关部门关注。

关键词: 硒; 土壤; 作物; 人发; 人体健康; 黑土地质量; 环境地质调查工程

创新点: 基于系统的生态地球化学数据, 运用统计分析学原理, 开展典型黑土区土壤-作物-人体系统中硒迁移与积累研究, 为富硒农产品开发和地方病防治提供科学依据。

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Geochemical characteristics and evaluation of ecological environment and human health of selenium in soil-crop-human system in Shuangyang River Basin, Heilongjiang

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Abstract: This paper is the result of ecological geological survey engineering.

[Objective] The natural endowment, genetic source, ecological environment effect and human health evaluation of soil selenium in typical black soil distribution areas are the research focus of healthy geology, a quantitative study on the migration and accumulation of selenium in the soil-crop-human system in the Shuangyang River Basin was carried out. The development of selenium-rich agricultural products in black soil and the prevention and control of endemic diseases are of great significance. **[Methods]** The selenium content of parent material, soil, atmospheric dust, irrigation water, crop seeds and human hair was determined, and the results were statistically analyzed and evaluated by GIS and SPSS software. **[Results]** The selenium-enriched land is 46.4 km², mainly distributed along the Shuangyang River and its tributaries; the selenium is largely caused by human beings, mainly from atmospheric dust supplementation with an input flux of 0.21–1.47 kg/km²·a. The mean selenium contents of soil, soybean, rice, corn and human hair are 0.305 mg/kg, 0.0352 mg/kg, 0.0287 mg/kg, 0.0198 mg/kg and 0.145 mg/kg. The enrichment coefficient of selenium from crops to soil (0.120) is much less than 1, and the enrichment coefficient of selenium from human hair to crops (4.834) is greater than 1, indicating that human hair has a strong concentration and amplification effect on selenium. The migration and accumulation of selenium in the soil-crop-human system shows a "V"-shaped pattern. **[Conclusions]** In general, the selenium content in the soil is sufficient or rich with no potential environmental risk. Soybean and rice are rich in selenium, accounting for 28.4% and 9.4%, with no selenium exceeding the standard and poisoning in organisms. The content of selenium in human hair is significantly lower than the reference value of 0.20–0.60 mg/kg, indicating that the human body is in a state of selenium deficiency, and the selenium content tends to decrease with age. There may be certain health risks, which should arouse the attention of local relevant departments.

Key words: selenium; soil; crops; human hair; human health; black soil quality; ecological geological survey engineering

Highlights: Migration and accumulation of selenium in soil-crop-human system of typical black soil area were studied based on systematic ecological geochemical data and statistical analysis principle, which provided scientific basis for the development of selenium-rich agricultural products and local disease control.

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1 引言

硒具有抗氧化、抗癌变、抗病毒等生理功能,是含硒蛋白的关键组成物质,参与人体一系列重要的生理代谢(Muller et al., 1969; Hakanson et al., 1980; Qin et al., 2013; Hosnedlova et al., 2017)。人体对硒具有一定的营养需求,缺硒会引起诸多疾病,如白肌病、桑椹状心脏病、克山病和大骨头病等(谭见安等, 1998; 曾昭华等, 1998, 2001; Ferreira et al., 2005; 刘发欣, 2007; Tan et al., 2012; Chen et al., 2012; 赵君等, 2021; Xiu et al., 2022); 同样,生活在

土壤硒含量高于 3.0 mg/kg 的区域,存在硒中毒的风险(程伯容等, 1980; 林克惠等 2009; 戴慧敏等, 2015; 迟凤琴等, 2016)。硒一般通过“土壤-作物-人体”或“土壤-作物-动物-人体”的食物链方式(李海华等, 2000; 胡省英等, 2003; 陈文德等, 2009; 梁帅等, 2021), 以及人体直接暴露在硒源区时人体皮肤接触和呼吸吸入的方式进入人体(廖自基, 1989; 李婷等, 2020)。饮食摄入是人体最主要的硒摄入方式, 90%的硒均来源于此(白志鹏等, 2002), 人体硒含量水平直接取决于食物硒含量, 间接取决于土壤和水中的硒含量。

本文基于“典型地区1:5万土地质量调查及成果转化应用研究”课题所获取的黑龙江省双阳河流域成土母质、表层土壤、作物籽实、灌溉水、大气干湿沉降物和入发等原始数据,探讨硒在土壤-作物-人体系统中的含量特征及生态环境与人体健康评价,为地方富硒农产品开发和地方病防治提供依据。

2 研究区概况

区域上位于新华夏系第二沉降带松嫩凹陷东北部,自白垩纪以来,区域地层处于东西向倾斜构造状态,自东向西,由下而上沉降有巨厚白垩系陆相碎屑岩层,包括厚度较大的古近—新近系下—中新统依安组含煤的泥页岩层(夹有数层粉、细砂岩)和古近—新

近系上新统孙吴组疏松的砂岩、砂砾岩、砂质泥岩等(图1)。受晚新近世构造运动和小兴安岭影响,高平原区(东北部)相对上升,低平原区(西南部)继续下降,第四系覆盖物由自东北向西南增厚,前第四系绝大部分被第四系覆盖,仅于高平原河谷强烈切割处出露(黑龙江省第四纪地质及环境,1990)。

双阳河流域地处黑龙江省齐齐哈尔市东南角,坐标47°17'~47°54'N、125°29'~126°31'E,属寒温带大陆性季风气候,夏短多雨,秋季早霜,四季温差较大,年均气温1.2℃、降水量488.2 mm、日照2730 h。地貌为小兴安岭松嫩平原漫岗丘陵地带,海拔高度260~280 m,地势总体北高南低,地表坡度2°~4°。为黑龙江省农作物品种积温区划第三积温带(2300°

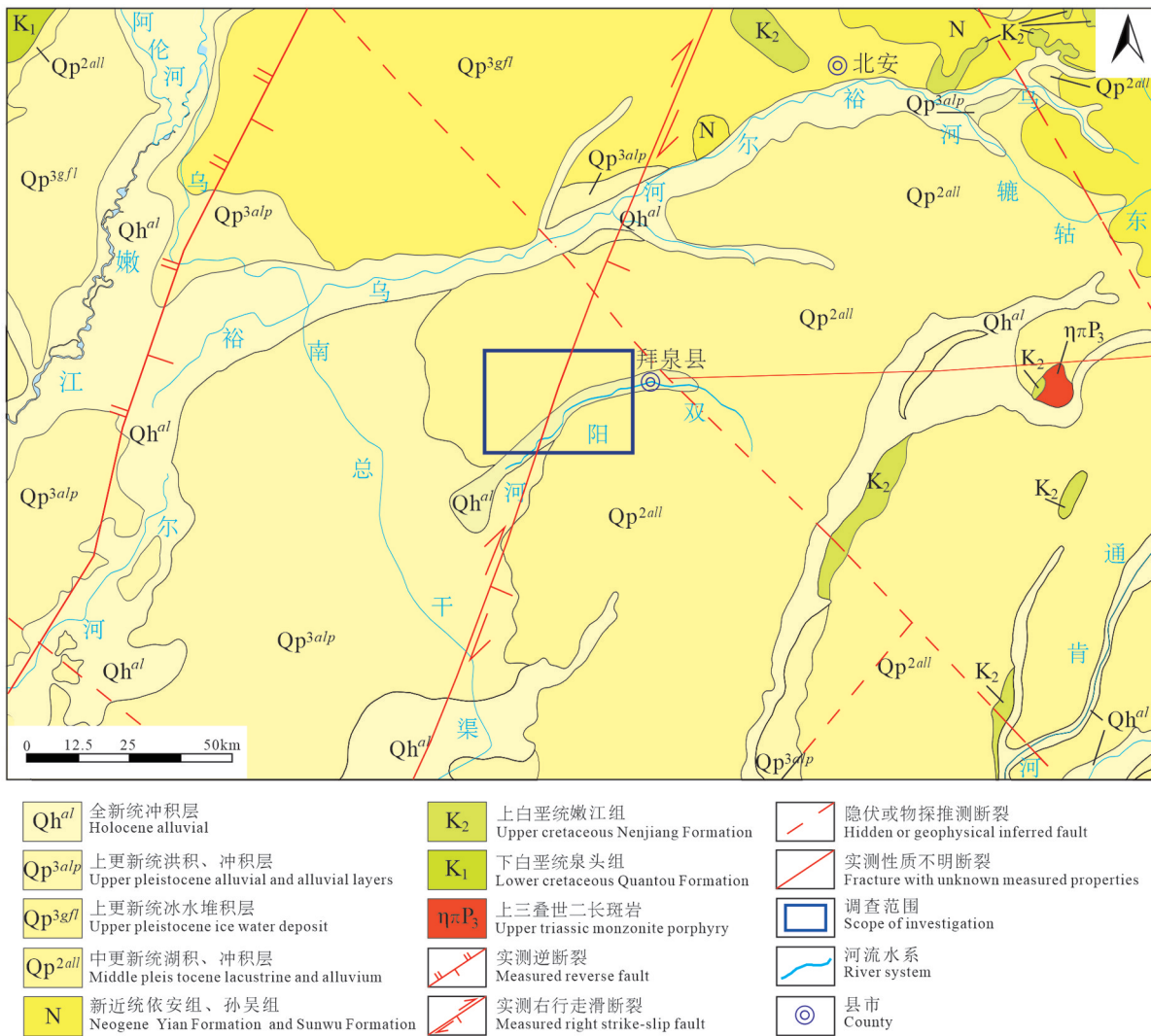


图1 研究区位置及区域地质简图
Fig.1 The location of the study area and a sketch map of the regional geology

~2500°),适于农耕,耕地类型主要为旱地,其次为水浇地。土壤类型主要为黑土(北部和东南部)、草甸土(中部)和黑钙土(西南部)。

3 材料与方法

3.1 样品采集与测试

3.1.1 样品采集

参照《土地质量地球化学评价》(DZ/T 0295—2016)规范,表层土壤样品采集选用网格法,密度4点/km²,深度20 cm,刻槽取样;样品原始重量大于1500 g,由3个子样组合而成,任意2个子坑间距大于30 m,子样坑呈三角形分布。成土母质样品按高硒区、过渡区、低硒区分类采集,深度1.6~2.0 m。土壤剖面样品采集按成土母质类型布置,深度1.6~2.0 m,密度1个/20 cm。作物及根系土样品采集依据表层土壤样品测试结果,采集富硒区大豆、水稻等作物籽实及同位置根系土样品。灌溉水样品采集主要沿双阳河自上游至下游依次采集,密度1点/3 km。大气降尘样品采集选用大气干湿沉降法,降尘桶口径28.3 cm,固定于距地面约5 m的高处,取样周期一年。人发采集对象主要为常年居住在当地的人群,采集距头皮2.5 cm之内的发样,重5~10 g。样品采集点位信息见图2和表1。

3.1.2 样品测试

样品测定了Se、pH、Corg等指标,测试单位为自然资源部沈阳地质矿产监督检测中心,主要采用电感耦合等离子体发射光谱法(ICP-OES)、原子荧光法(AFS)等测试方法。样品测试精密度、准确度和数据报出率等质量参数均符合《多目标地球化学调查规范》(DZ/T0258—2014)和《生态地球化学评价样品分析技术要求》(DD 2005-03),分析质量可靠,数据准确。

3.2 数据分析与研究方法

3.2.1 数据统计

文中描述性统计、方差计算等内容均是通过SPSS ver.22.0(IBM, New York, NY, USA)完成。土壤硒含量的空间分布图使用ArcGIS 10.2(ESRI, Redlands, CA, USA)绘制。其他统计图是通过Origin 2019(OriginLab, Northampton, USA)绘制。

3.2.2 大气干湿沉降

大气干湿沉降Se元素沉降通量($Q_{\text{大气}}$)的计算

公式为:

$$Q_{\text{大气}} = (C_{\text{干}} \times W_{\text{干}} + C_{\text{湿}} \times V_{\text{湿}}) / A \times 10 \quad (1)$$

式中: $Q_{\text{大气}}$ 为大气干湿沉降中Se元素沉降通量,kg/km²; $C_{\text{干}}$ 为干沉降中Se元素的含量,mg/kg; $W_{\text{干}}$ 为接尘桶中干沉降的质量,g; $C_{\text{湿}}$ 为湿沉降中Se元素的浓度,mg/L; $V_{\text{湿}}$ 为接尘桶中湿沉降的体积,mL; A 为接尘桶口面积,取490.86 cm²。

4 硒地球化学分析

4.1 土壤硒含量特征

表层土壤样品测试数据统计显示(表2),硒含量为0.13~2.12 mg/kg,变异系数0.243,为中等程度变异,反映硒在该区域分布不均匀,可能受到后期人为活动的影响(赵燕等,2021)。土壤硒(Se)平均含量为0.305 mg/kg,高于黑龙江土壤背景值0.147 mg/kg,是其2.07倍(中国土壤元素背景值,1990)。土壤有机碳(Corg)含量为0.86~6.50%,均值为2.40%;酸碱度(pH)为5.16~9.35,均值为6.96。

4.2 作物硒含量特征

作物籽实样品测试数据统计显示(表3),作物籽实硒含量为0.014~0.057 mg/kg,均值为0.03 mg/kg,不存在硒含量超标样品,变异系数为0.209。不同类型作物籽实的硒含量差异较大,大豆、水稻和玉米三种作物籽实硒含量最大值和最小值均依次降低,三者均值为0.0352 mg/kg、0.0287 mg/kg和0.0198 mg/kg,其中大豆变异系数为0.229,达中度变异。

4.3 人发硒含量特征

人发是人体硒元素的排泄器官之一,其硒含量实际上就是人体在一段时间里向头发排泄该元素的量,从而可反映出硒在体内的整体代谢状况。人发硒含量一般比血液、尿液的高,并且人发具有取材方便、不损伤人体、易于保存和运输等优点,被看作是一种理想的活体硒检测材料和环境污染指示器(秦俊法等,2003,2005)。人发样品测试数据统计显示(表4),硒含量为0.099~0.192 mg/kg,均值为0.145mg/kg。

5 硒生态环境与人体健康评价

5.1 富硒土壤成因来源

从土壤硒元素地球化学图3可以发现,硒含量高值区整体沿双阳河及其支流分布,分布形态与地

表1 地球化学样品类型及数量统计
Table 1 Types and quantity statistics of geochemical samples

样品种类	采集数量	备注
表层土壤	2767件	20cm
农作物及根系土	180套	作物籽实、20cm
成土母质	30件	1.6~2.0m
土壤剖面	85件	0.2~2.0m
灌溉水	21件	双阳河流域
大气干湿沉降	15件	双阳河流域
人发	32件	双阳河流域

大气干湿沉降是硒输入土壤的一种重要途径,是弥补农田生态系统硒输出的重要方式,研究表明每年大气圈接受天然和人为硒总量为 15×10^9 g(王子健等,1989;朴河春等,1995;秦海波等,2009)。15个干湿沉降桶全年接收点实测数据见表6,依据公示(1)计算

出大气干湿沉降硒输入通量为 $0.21 \sim 1.47 \text{ kg/km}^2 \cdot \text{a}$,揭示大气降尘为土壤硒的主要补充来源,与夏学齐等(2012)学者的研究结果一致。农田灌溉对于提高农作物的产量和品质具有重要的意义,也是硒元素进入土壤的驱动力之一;21件灌溉水样品硒(表6,部分样品测试结果低于检测限 0.0002 mg/L)年输入通量统计结果普遍较低,小于 0.002 kg/km^2 ,排除灌溉水是土壤硒元素的主要来源。因此,表层土壤硒富集主要来自大气降尘补充,为人类金属冶炼、火力发电、冬季取暖等生产活动中煤燃烧所致(陈冰如等,1989;张莹,2007;田贺忠等,2009),其次来源于成土母质,灌溉水提供的硒源较少。

5.2 土壤硒潜在环境风险及生态效应

参照谭见安等(1998)对全国范围内土壤硒含量与地方病的对比研究给出的土壤硒含量生态环

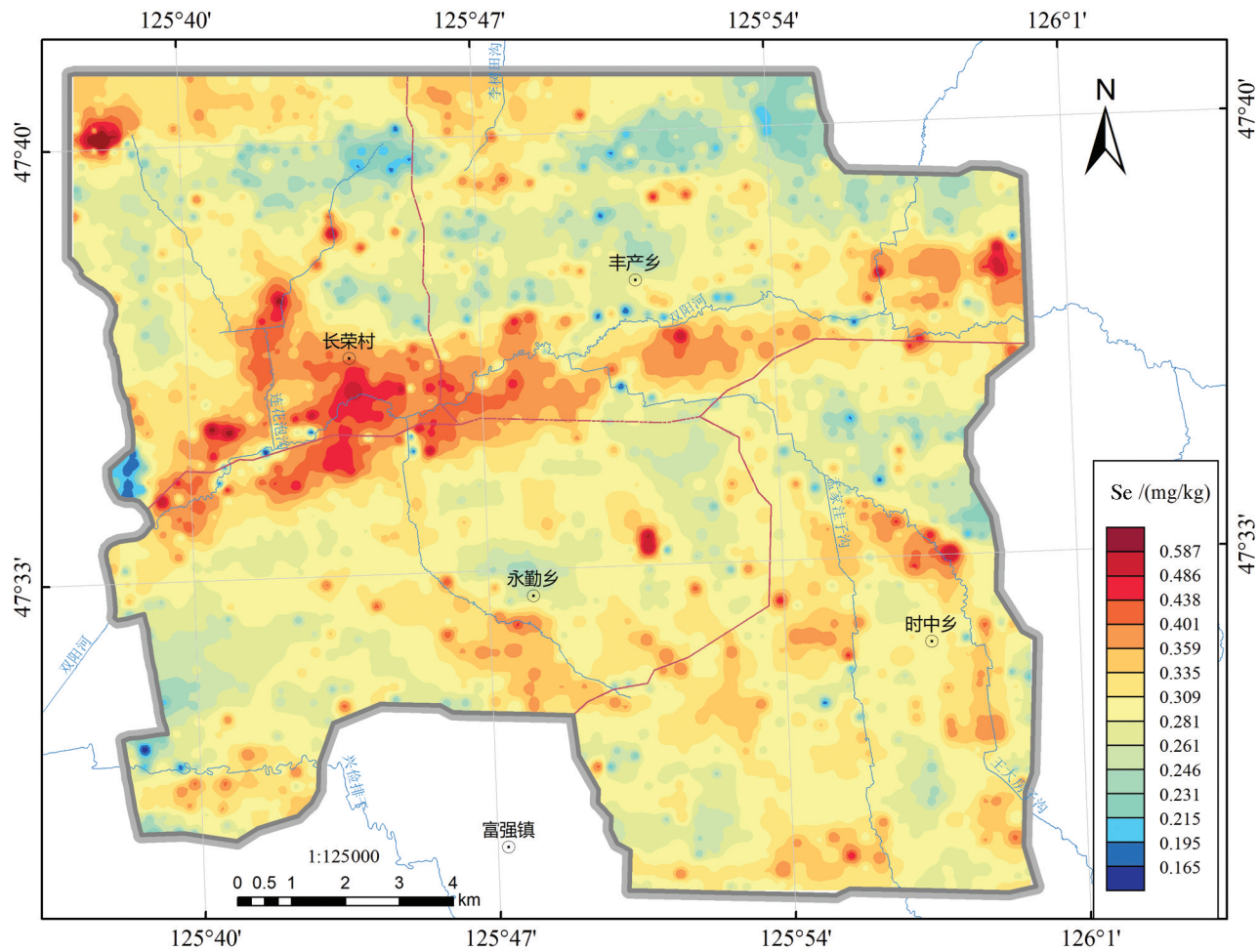


图3 双阳河流域表层土壤硒元素地球化学图

Fig.3 Geochemical map of selenium element in surface soil of Shuangyang River Basin

表2 表层土壤硒等主要指标含量统计

Table 2 Statistics of selenium elements in surface soil

元素	样本数	最大值	最小值	平均值	标准差	变异系数	黑龙江土壤背景值	富集系数
Se/(mg/kg)		2.12	0.13	0.305	0.074	0.243	0.147	2.07
Corg/%	2767	6.50	0.86	2.40	0.47	0.196	-	-
pH		9.35	5.16	6.96	0.92	0.132	-	-

表3 作物硒含量参数统计(mg/kg)

Table 3 Statistics of selenium in surface soil (mg/kg)

作物(样本数)	极小值	极大值	均值	变异系数	富集系数	富硒参考值	富硒数量	富硒占比
大豆(81)	0.024	0.057	0.035	0.229	14.08		23	28.4%
水稻(61)	0.02	0.056	0.03	0.200	11.48	0.04	3	9.4%
玉米(38)	0.014	0.031	0.02	0.150	7.92		0	0
共计(180)	0.014	0.057	0.03	0.209	-		-	-

表4 人发中硒含量特征统计(mg/kg)

Table 4 Statistical table of selenium content in human hair (mg/kg)

元素	样本数	最大值	最小值	平均值	标准差	变异系数	参考值
Se	32	0.192	0.099	0.145	0.025	0.172	0.20~0.60

表5 成土母质、土壤剖面硒含量统计(mg/kg)

Table 5 Statistics of selenium content in parent material and soil profile(mg/kg)

类型	样本数	最大值	最小值	平均值	标准差	变异系数
成土母质	30	0.107	0.027	0.065	0.020	0.299
土壤剖面	85	0.374	0.034	0.153	0.101	0.660

境风险监管界限值,划定土壤硒含量潜在环境风险红线为0.125 mg/kg,代表土壤硒缺乏;黄线为0.175 mg/kg,代表土壤硒存在潜在不足(表7)。2767件土壤测试结果统计显示,无红线以下样品,黄线以下样品为22件,占比0.80%;土壤硒含量平均水平相对较高,整体为足硒或富硒,无硒潜在环境风险,但空间上表现出较大的变异性,零星存在土壤硒潜在不足的地块;硒含量均低于土壤硒中毒阈值3.0 mg/kg,不存在土壤硒超标,无生物硒中毒风险。

作物从土壤中主动或被动吸收硒,受硒有效态量制约(刘红等,2000;梁帅等,2021)。大豆、水稻、玉米三种作物对硒元素的BCF_{作物/土壤}(BCF=作物籽实硒含量/根系土硒含量,根系土硒含量均值为0.25 mg/kg)分别为0.141、0.115、0.792,整体远小于1(BCF_{作物/土壤}=0.120),大豆、水稻、玉米硒富集程度依次降低。81件大豆样品中有23件样品硒含量>0.04 mg/kg,达到富硒食品标准,占比28.4%;32件水稻样

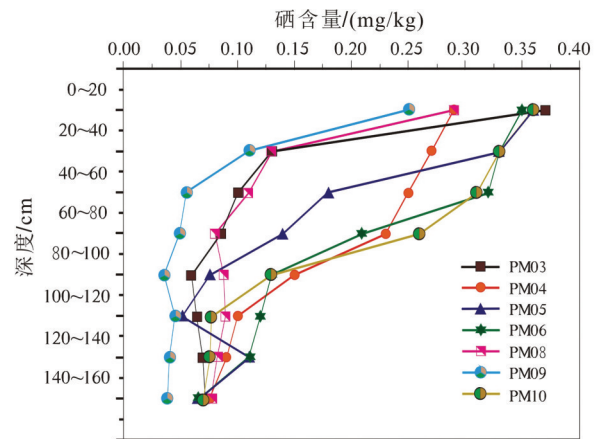


图4 土壤垂向剖面硒含量变化图

Fig.4 Changes of selenium content in soil vertical profile

表6 大气降尘、灌溉水硒含量参数统计

Table 6 Statistics of selenium content in atmospheric dustfall and irrigation water

类型	样本数	最大值	最小值	平均值	标准差	变异系数
大气降尘/(mg/kg)	15	0.63	0.17	0.24	0.157	0.64
灌溉水/(mg/L)	30	0.003	-	0.001	0.001	1

品中有3件样品硒含量>0.04 mg/kg,达到富硒食品标准,占比9.4%;玉米中未发现富硒样品。

人体摄入作物籽实及其产品后,微量元素又富集在人体不同的组织或器官中,通常其含量高于作物(M_i型),甚至高于土壤(M_j型);人发对作物的BCF_{人发/作物}大于1(CF_{人发/作物}=4.834),偶也有低于作物含量的情况出现(M_k型);硒元素沿土壤-作物-人

表7 土壤硒潜在环境风险划分标准
Table 7 Classification criteria for potential environmental risks of soil selenium

全量硒/(mg/kg)	硒效应	面积比例/%
<0.125	硒反应不足	0
0.125~0.175	硒潜在不足	0.80
0.175~0.4	足硒	90.89
0.4~3	富硒	8.31
>3	硒中毒	0

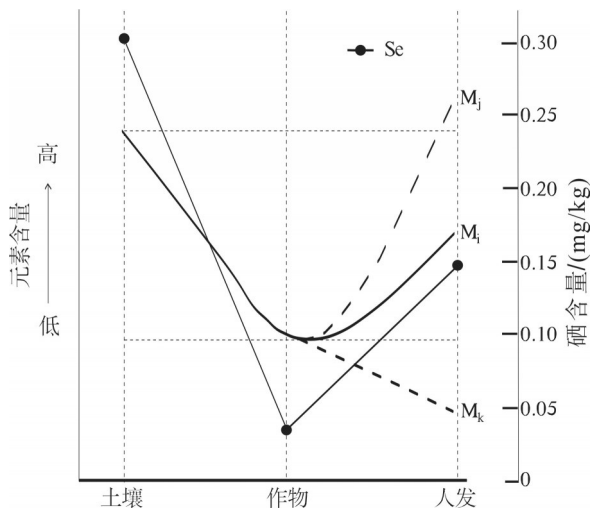


图5 硒在土壤—作物—人发系统中的迁移与积累模式
(据马建华等修改, 2014)

Fig.5 Migration and accumulation patterns of selenium in soil-crop-human hair system
(modified from Ma Jianhua et al., 2014)

发迁移路径的积累曲线整体呈“V字型”(图5),说明人发对硒具有较强的浓缩和放大作用。

5.3 人体健康评价

人发硒含量受性别、年龄、居住环境、饮食和生活习惯等诸多因素影响(秦俊法等, 2003),因此不同国家和地区人发硒参考值存在较大差别。研究区女性头发硒含量均高于男性(表8),可能与女性的生活习惯、体质和运动量有关,造成体内硒累积量高于男性(梁刚等, 2012)。人发硒含量均值为0.145 mg/kg,明显低于参考值范围0.20~0.60 mg/kg(中国微量元素科学研究会标准, 2007),反映采集样本整体处于缺硒状态,可能存在一定健康风险。

不同年龄组人发硒含量统计见表9,人发硒含量自8~20岁年龄段至>70岁年龄段均呈下降趋势,且人发硒含量与年龄段之间的相关系数为-0.408*

(*代表在0.05水平(双侧)上显著相关),呈显著负相关性,均说明人发中硒含量随着年龄的增长呈递减趋势,可能与人体的代谢水平有关(Trojanowski et al., 2010; Perumal et al., 2011),应引起地方相关部门关注。

6 结论

(1)划定富硒土壤面积46.4 km²,主要沿双阳河及其支流分布,成因以人为成因为主,主要来自大气降尘(输入通量为0.21~1.47 kg/km²·a)补充,为人类金属冶炼、火力发电、冬季取暖等生产活动中煤燃烧所致。

(2)土壤硒含量0.13~2.12 mg/kg,均值为0.305 mg/kg,是黑龙江土壤背景值2.07倍,变异系数为0.243,达到中等变异。大豆、水稻和玉米三种作物籽实硒含量均值分别为0.0352 mg/kg、0.0287 mg/kg和0.0198 mg/kg。人发硒含量为0.099~192 mg/kg,均值为0.145 mg/kg。作物对土壤硒的BCF_{作物/土壤}远小于1(BCF_{作物/土壤}=0.120),而人发对作物硒的BCF_{人发/作物}大于1(BCF_{人发/作物}=4.834),硒沿土壤-作物-人发迁移路径的积累曲线整体呈“V字型”,说明人发对硒具有较强的浓缩和放大作用。

(3)土壤硒含量整体为足硒或富硒,无硒潜在环境风险和生物硒中毒风险。作物籽实均未发现

表8 不同性别人发硒含量(mg/kg)

Table 8 The content of selenium in human hair of different genders (mg/kg)

性别	最大值	最小值	均值	标准差	变异系数
男(7)	0.183	0.099	0.145	0.023	0.159
女(25)	0.192	0.125	0.164	0.024	0.146
共计(32)	0.192	0.099	0.145	0.025	0.172

表9 不同年龄段人发硒含量(mg/kg)

Table 9 The content of selenium in human hair of different age groups (mg/kg)

年龄	样本	最小	最大	均值	标差	变异系数
8~20	8	0.122	0.192	0.157	0.022	0.140
21~30	1	0.114	0.114	0.114	-	-
41~50	3	0.122	0.159	0.141	0.019	0.135
51~60	8	0.099	0.183	0.140	0.028	0.200
61~70	8	0.105	0.184	0.136	0.024	0.177
>70	3	0.108	0.159	0.132	0.026	0.197

硒超标情况,大豆富硒占比28.4%,水稻富硒占比9.4%,玉米中未发现富硒样品。人发硒含量明显低于参考值0.20~0.60 mg/kg,人体处于缺硒状态,随年龄增长呈递减趋势,可能存在一定的健康风险,应引起地方相关部门关注。

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