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Emission Inventory Development for Spatiotemporal Release of Vanadium from Anthropogenic Sources in China

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Abstract. Anthropogenic activities contribute primarily to the toxic vanadium presence in the surface environment, but quantitative assessment of its emissions from anthropogenic sources to various environmental receptors is still lacking. This study has developed nationwide vanadium emission inventory in China during 2015-2019, covering five major anthropogenic sources, including coal combustion, stationary oil burning, transportation, industrial production, and waste handling. Cumulative emission flux modelling has shown that 211094 t, 3725 t, and 0.1 t of vanadium were discharged into atmosphere, soil and water during this period. Coal combustion and stationary source of oil burning are the largest vanadium contributors, accounting for 47.5% and 39.6% of emission inventory. Shandong, Liaoning, Hebei, Guangdong and Hunan are among the largest provincial emitters. Emissions pertinent to raw coal combustion mainly increase by 719 t and 316 t in the provinces of North China and Northwestern China, respectively. Vanadium output pertinent to steelmaking constitutes 88.2% emission in industrial production, and continued to increase in all regions. Emissions induced by vanadium mining shows remarkable spatial heterogeneity, with 66.1% output determined in Southwestern China. Emissions pertinent to raw coal and coke combustion was the main source of uncertainty for the inventory development, resulting in output uncertainty ranging from -47.5% to 63.7% and -49.4% to 53.7%.

1 Introduction

Vanadium is a transit metal ubiquitously found in the natural environment, including soil, water, atmosphere, and fauna system. The global reserve of vanadium exceeds 63 million tons (U.S. Geological Survey, 2023). China, Russia, South Africa and Brazil were the top four countries with the most abundant vanadium resource (Schlesinger et al., 2017), among which China harbors the most vanadium, totaling 9.5 million tons (Fu et al., 2023). Vanadium exists in four main types of mineral sources in the earth crust, including vanadium-titanomagnetite, stone coal, petroleum associated minerals and uvanite (Zhang et al., 2023). As the largest vanadium producer, Sichuan and Hebei province in China account for vast majority of vanadium output, with principal vanadium forms in vanadium-titanium deposits (Yu et al., 2015), and Shaanxi, Gansu, Hubei, Hunan province, mainly in vanadium containing stone coal (Yuan et al., 2022). Vanadium plays indispensable catalytical roles in modern industrial development, particularly in the manufacture of steel, ceramic, glasses, pigment products, as well as vanadium redoxflow batteries (Watt et al., 2018; Wang et al., 2023). The rising demand in vanadium production has led to intensive anthropogenic emission, which makes vanadium a re-emerging environmental problem and draws increasingly more attention from research entities (Watt et al., 2018).

Vanadium exposure can lead to potential adverse effects (Huang et al., 2015; Fei et al., 2023; He et al., 2023), which calls for stricter environmental regulation on vanadium level in the environmental medium. The mobilization of environmental vanadium can occur through natural pathways, i.e., weathering of vanadium bearing rocks (Hope et al., 2008; Shiller and Mao, 2000) and volcanic activities (Bortnikova et al., 2009), as well as wide ranges of anthropogenic activities, which has significantly surpassed the vanadium output induced by natural processes (Schlesinger et al., 2017). The inefficient vanadium utilization (i.e., 70%) during industrial processes results in discharging vanadium into the natural environment, including air (Wang et al., 2020a), soil (Zhang et al., 2020) and water (Wright et al., 2014). For example, in Panzhihua city, China, soil contains up to 4793 mg/kg of vanadium adjacent to the smelter plant (Zhang et al., 2019), while 228 ng/m^3 of vanadium is determined in aerosol samples collected from the same area (Wang et al., 2020a). Among all emission pathways, the cumulative atmospheric emission of vanadium made the largest contribution to environmental pollution, which has increased by 86% over the past few decades in China (Bai et al., 2021). In addition, the sum of atmospheric emission factor of vanadium bearing ore during mining, smelting and product making process is 43 kg/t (Anderson, 1973), which is significantly higher than that induced by coal burning (0.001 kg/t), petroleum consumption (0.06 kg/t), and steelmaking (0.0001 kg/t) (Anderson, 1973). However, there is an absence in the quantitative evaluation of vanadium flux derived from different anthropogenic sources to different environmental receptors, especially at the continental scale.

Emission inventories are developed to effectively elucidate the magnitude, spatial and temporal patterns of pollutant emission originated from different sectors of activities, which can serve as policy backbone to reduce the environmental impact of pollutants. The development of vanadium emission inventories is performed at global scales for anthropogenic sources (Pacyna et al., 2001). However, these inventories hardly capture the variation in vanadium emissions across different sectors, which usually are affected by ever-changing market demand and improvements in pollution control technologies, resulting in poor resolute inventory. To address the inadequacy in control and supervision of vanadium discharge, it is vital to establish the national emission inventory system, which comprehensively tracks the vanadium emission pattern at a spatiotemporal scale, providing insightful information for governing bodies to formulate solutions to reduce the environmental impact based on source, pattern and occurrence of vanadium emission. However, no effort has not been made with this regard.

The goal of this study is to establish the bottom-up inventories of vanadium emission from anthropogenic activities during the period from 2015 to 2019 in China. The following tasks were carried out to implement the analysis: (1) performing the modelling study to quantify the contribution of anthropogenic sources to environmental receptors; (2) determining the temporal and spatial pattern of anthropogenic vanadium release from different sectors of anthropogenic activities, in terms of years and regions (provinces); (3) conducting the uncertainty analysis for the inventory model.

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2 Methodologies

2.1 Anthropogenic sources for vanadium emission

The environmental impact was assessed utilizing the life cycle assessment (LCA) that accounted for five major anthropogenic sources leading to vanadium emission, which were consistent with previous studies (Bai et al., 2021), including (1) coal combustion, (2) stationary oil burning, (3) transportation, (4) industrial processes, and (5) waste disposal. The subgroups of each category were also displayed in full in Table 1 along with their presumed environmental receptors. Emission sources were assumed to release vanadium into the respective receptors through single route (i.e., airborne, solid waste, wastewater) (Hope, 2008). The presumed vanadium emission routes of each anthropogenic process were illustrated in details in the Supporting Information.

Table 1. Anthropogenic sources of vanadium emission and potential receptors.

Subgroups (acronym)	Sources	Potential receptors
Raw coal (RC)	Coal combustion	Air
Coke (CK)		Air
Crude oil (CO)	Stationary oil burning	Air
Gasoline (GO)		Air
Kerosene (KS)		Air
Diesel (DO)		Air
Fuel oil (FO)		Air
Gasoline (GO)	Transportation	Air
Diesel (DO)		Air
Fuel oil (FO)		Air
Steelmaking (SM)	Industrial processes	Air
Glass production (GM)		Air
Coal mining (MG)		Soil
Petroleum (OE)		Soil
Vanadium mining (VM)		Soil
Municipal waste incineration (INC)	Waste disposal	Air
Landfill (LF)		Soil
Solid waste disposal (SW)		Soil
Sludge disposal (SD)		Soil
Wastewater (WW)		Water

2.2 Computational method for establishing emission inventory

The decision tree was constructed to provide guideline in selecting the appropriate computational method during inventories development (Fig. S1). In general, the onsite measurements of emission parameters during anthropogenic processes were obtained to estimate the vanadium emission pertinent to consumption by industrial application, integrating mainly the emission

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factors of each procedure and activity levels (Gu et al., 2023). For datasets reflecting the industrial production, the following formula was used:

$EV = \sum j(EFj \times APj)$ (2-1)

Where EV was the vanadium emission in metric ton, EFj was the emission factor in kg vanadium emitted per ton of products; APj was the total production amount in corresponding industrial application.

Datasets obtained from regional or provincial statistic reflected the geographic variation in vanadium emission, for which the following computation was applied.

$EV = \sum i(EFi \times EPi)$ (2-2)

Where EV was the vanadium emission in metric ton, EFi was the regional (provincial) emission factor in kg vanadium emitted per metric ton of products; EPi typified the total production volume at specific geographic region.

Default values obtained from other literature were used due to insufficiency in data collection and data quality, which were employed in the last resort. Therefore,

$EV = EP \times AP$ (2-3)

EP was the default value for vanadium emission factor in vanadium emitted per metric ton； AP was the national output in metric ton.

The inventory was built based on five major categories of anthropogenic process, which comprised totally 21 sub-sources, across China's 31 provinces. The detailed algorithm of the bottom-up emission inventory was provided in full details in the Supporting Information, with raw datasets uploaded in data repository (Zhang, 2023).

2.3 Uncertainty analysis

The uncertainties in algorithm of emission inventory affected both data input and output of emission model. The model was formulated based on previous experience, with errors which failed to reflect the realistic condition. Moreover, the input parameters experienced constant changes by random chance. In this study, input data for quantification of the anthropogenic activities and emission factors were summarized from available literature sources. The uncertainty levels of these variables were evaluated by using the Bootstrapping simulation (Tian et al., 2011). The types of probability distribution of variables were determined (i.e., normal distribution, triangle distribution, Beta distribution, Gamma distribution etc.), followed by random sampling of the variables to obtain bootstrapped samples, for which the confidence intervals were calculated to delineate the uncertainties level of the datasets. The detailed calculation procedure was provided in Supporting Information. Furthermore, the uncertainty in the inventory output for vanadium emission was quantified by Monte Carlo simulation, which was performed in AuvToolPro2.0 following the protocol (Zheng et al., 2017).

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3. Results and discussion

3.1 Modelling the anthropogenic vanadium emission fate

The conceptual model for vanadium flux flow illustrated the relationship between anthropogenic emission sources and the cumulative vanadium in environmental receptors (Fig.1). During 2015-2019, the majority fraction of vanadium generated from anthropogenic processes was directly discharged into the atmosphere, leading to a total emitted vanadium content of 211094 t. Most of the atmospheric vanadium was derived in raw coal combustion, burning of gasoline and fuel oil in stationary setting, resulting in cumulative amounts of 91911 t, 31145 t and 21718 t, respectively. 3725 t of vanadium was directly deposited into the soil, which was predominantly attributed to industrial waste discharge and sludge disposal, totaling 1398 t and 1425 t, respectively. Wastewater treatment was the sole and direct source of vanadium discharge into water environment, and it only contributed 0.1 t of vanadium during 2015-2019. However, it was commonly recognized that soil served as the largest sink for vanadium (Cao et al., 2018; Zhang et al., 2019; Zhang et al., 2023) as vanadium was firstly discharged into atmosphere, and subsequently introduced into soil as result of wet or dry deposition (Wang et al., 2020b). In addition, the surface runoff and vertical percolation further transported vanadium enriched in surface soil into surface water (Zhang et al., 2023), or even groundwater aquifer (Fei et al., 2022).

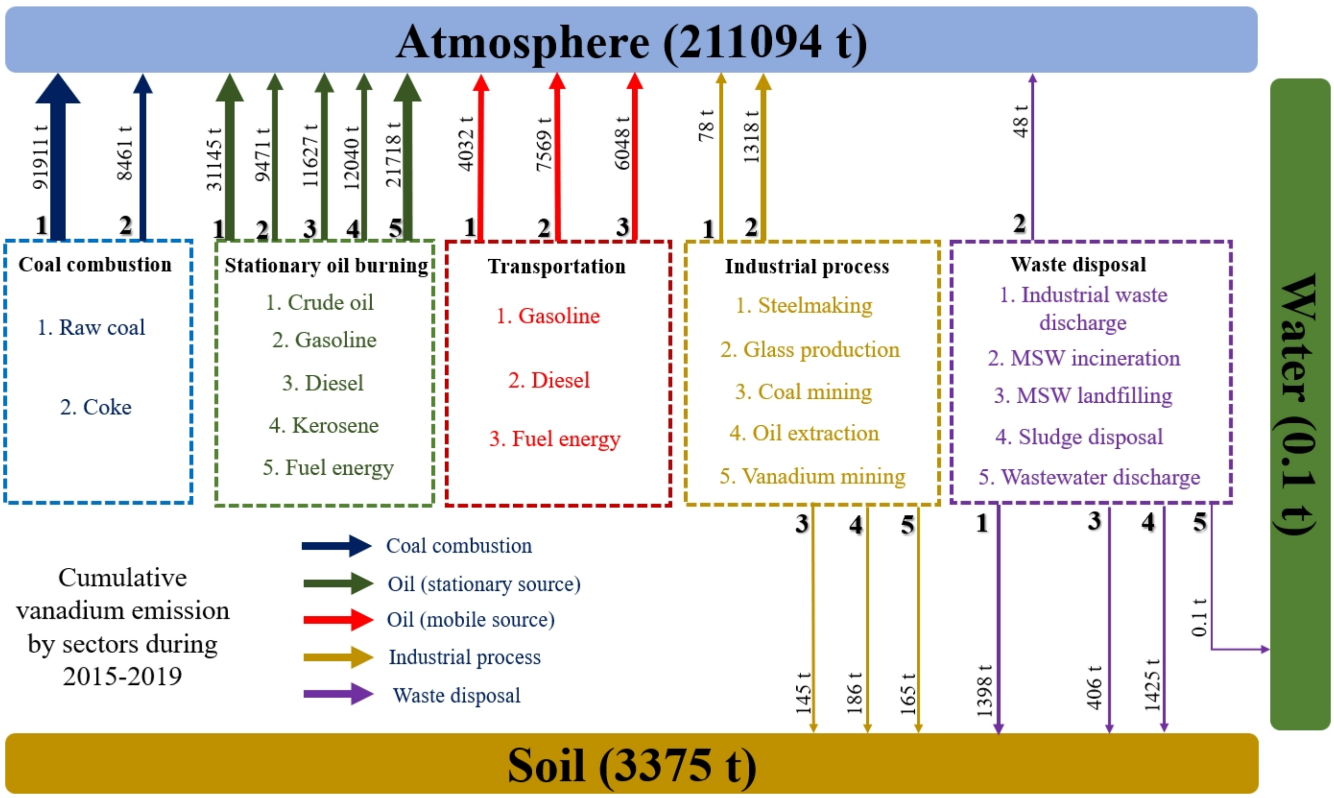

Figure 1: Environmental flux model for vanadium directly discharged from anthropogenic activities into environmental receptors. Vanadium flux data was calculated cumulatively during 2015-2019 from different anthropogenic sectors with vanadium emission,

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i.e., coal combustion, stationary source oil burning, transportation source, industrial processes and waste disposal, all of which were labeled by numbers. Lines with arrow head indicated the vanadium flux from different sources to respective receptors, with line thickness proportional to the magnitude of vanadium flux.

3.2 Temporal variation pattern of vanadium emission

During 2015-2019, the temporal trend of vanadium emission by contribution percentage of major anthropogenic sources was illustrated in Figure 2. The total vanadium emission increased slightly from 41389 t in 2015 to 44069 t in 2019 (Fig. 2a). Coal burning and stationary oil combustion made up of the major portion of vanadium emission inventory, accounting for 47.5% and 39.6% of cumulative emitted vanadium, respectively. Vanadium emission inventory also comprised input from transportation source, totaling 8.2%. The temporal variation was also consistent with global trend (William et al., 2017), where emission from both stationary and mobile source of oil combustion jointly constituted 50% of vanadium emission inventory, while coal burning led to 47.5% of total output.

Emission originated from consumption of raw coal accounted for 91.6% of total emission pertinent to coal combustion (Fig. S2), which exhibited a steady increasing trend from 18096 t to 18856 t during 2015-2019 (Fig. 2b). Vanadium emission pertinent to coke consumption made up of 8.4% of total coal induced emission, and it showed a slight decreasing trend from 1789.9 t to 1743.6 t. This trending pattern corroborated China's increasing efforts to enhance the energy efficiency of sintering and steelmaking processes (Xing et al., 2020). Vanadium emission pertinent to stationary source of oil burning was predominantly contributed by crude oil consumption (36.2%) (Fig. S2). The pronounced increase in vanadium emission was observed in consumption of crude oil, gasoline and kerosene, from 5882.7 t, 1754.3 t, 1993.9 t, to 6782.1 t, 1987.1 t, 2720.2 t, respectively (Fig. 2c). The rapid economic growth during this period has driven increased demand in oil refining activities (Zhang et al., 2019b). However, the vanadium emission pertinent to combustion of fuel oil decreased significantly, which was possibly because of successful implementation of emission trading scheme focused on energy-intensive heavy industries (Ouyang et al., 2020). As for transportation sector, consumption of gasoline (42.9%), diesel (34.3%) and fuel oil (22.8%) contributed considerably to vanadium emission (Fig. S2), all of which experienced emission growth from 691.1 t to 961.3 t, 1453.7 t to 1602.4 t, and 1000.5 t to 1469.5 t, respectively (Fig. 2d). Yearbook of Transportation and Automobile Industry suggested that increase in car purchase and logistic activities were the main driver for the elevated consumption of oil products, which raised the utilization of vanadium-based catalyzer for exhaust gas treatment (National Bureau of Statistics of China, 2020). Moreover, industrial processes and waste disposal also contributed minor portion of vanadium emission inventory, accounting for 3.28% and 1.52%, respectively (Fig. S2). Among industrial production activities, steelmaking process was the primary emitter (88.2%) (Fig. S2), showing a significant increasing trend from 1212 t to 1593 t during 2015-2019 (Fig. 2e). It was worth noting that despite of high vanadium concentrations detected around vanadium mining sites (Wang et al., 2020b), the mining related vanadium emission was marginal (1.9%), which was ascribed to limited activity level in total compared to widespread fossil fuel burning. Among sources of waste disposal, the handling of wastewater sludge (43.5%) and industrial solid waste (42.7%) were the primary contributors (Fig. S2). Vanadium emission pertinent to wastewater sludge decreased significantly from 322.8 t to 249.9 t during 2015-2019. The National Soil Act published in 2016 called for regulated sludge

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utilization, promoting the wastewater sludge recycling for construction material and biofuel extraction, instead of land application in order to avert the exacerbated soil pollution (Rohrbach et al., 2023). The disposal of industrial solid waste increased vanadium output from 249.1 t to 331.1 t (Fig. 2f), while vanadium emission pertinent to landfill and MSW incineration were relatively stable during 2015-2019. Lastly, negligible impact was incurred on vanadium emission inventory as result of direct municipal wastewater discharge. However, the intrinsic vanadium concentration in surface water was extremely low (< 10 ng/L) (Schlesinger et al., 2017), any input from anthropogenic source caused noticeable change in water quality.

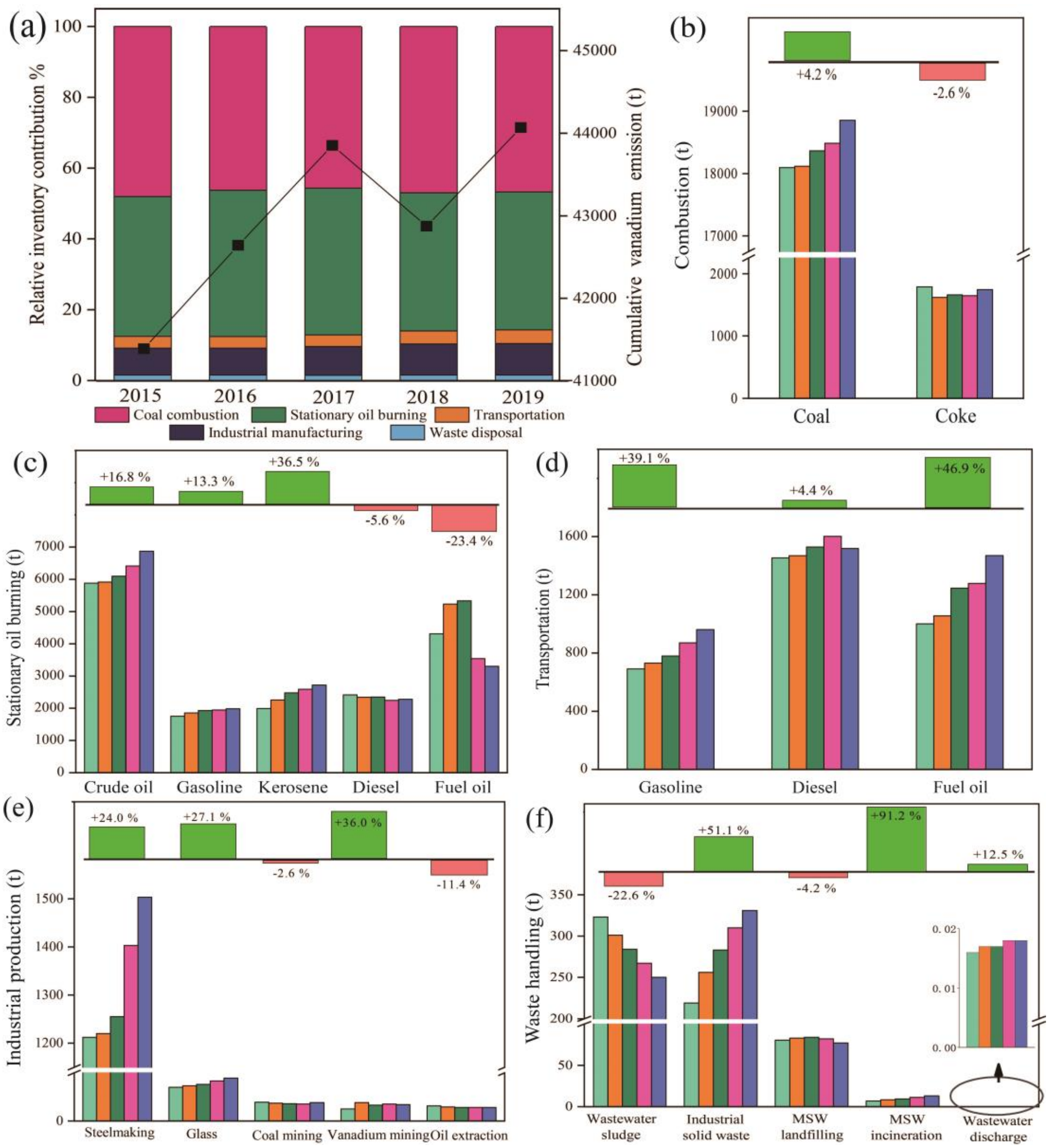

Figure 2. Historical trend of vanadium emission inventory during 2015-2019. (a) The relative contribution of yearly cumulative vanadium emitted from each major anthropogenic source was calculated, along with total emission amount; the yearly vanadium

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emission from subgroups of (b) raw coal combustion, (c) stationary oil burning, (d) transportation, (e) industrial production, and (f) waste handling.

3.3 Provincial level of vanadium emissions and spatiotemporal distribution pattern

To facilitate further investigation on variation patterns in anthropogenic contribution by various sources, we classified the 31 provinces into 7 geographic regions as follows: Eastern China (EC), Central China (CC), Northern China (NC), Northeastern China (NEC), Northwestern China (NWC), Southern China (SC) and Southwestern China (SWC). The vanadium emission inventories from different sources at provincial level over the period of 2015-2019 were presented in Table S1 – S18, and displayed in ArcGIS maps to reflect the cumulative (Fig. S3) and spatiotemporal distribution (Fig. 3). In general, a substantial amount of vanadium emission was found in northern, eastern coastal, southern and south western provinces, with Shandong, Liaoning, Hebei, Guangdong and Hunan among the largest emitters with cumulative vanadium discharge over 10000 t (Fig. S3a).

Henan, Hunan in CC, Shanxi, Inner Mongolia in NC, Shaanxi in NWC, Shandong in EC and Yunnan, Guizhou in SWC were the major contributor to vanadium emission (> 4500 t) from coal combustion (Fig. S3b). Major increase in raw coal induced emission was determined in NC (719 t) and NWC (361 t) (Fig. 3b). Coal was used as the main energy source to meet the needs of economic development throughout China, especially in NC and NWC where coal resources are abundant and relatively inexpensive (Tian et al., 2012). In comparison, vanadium emission decreased notably in SWC (-290 t) and CC (-269 t). Coal produced in SWC region was highly enriched with toxic arsenic (Wang et al., 2020b), which may invoke more restrictions on the production activity. There was also increased investment on greener energy such as natural gas (Wang, 2021) adopted by CC (e.g., Henan) to reduce their dependence on coal. The major vanadium emission (> 500 t) pertinent to coke usage was found in Hebei in NC, Hunan in CC, and Sichuan, Yunnan in SWC, where coking operations were concentrated. Large vanadium emission (> 5000 t) pertinent to stationary oil burning mainly occurred in coastal regions including Liaoning in NEC, Shandong, Shanghai in EC and Guangdong in SC (Fig. S3c). In particular, NEC (583 t) and EC (259 t) observed significant increase in vanadium emission induced by crude oil consumption (Fig. 3c). Robust economic development in these regions stimulated the industrial activities in various sectors including machine manufacturing, electronics, petrochemical, and seaport activities, leading to higher demand in oil consumptions. Direct vanadium emission from industrial production processes were much less, but exhibited a heterogeneous distribution pattern (Fig. S3d). The steel industry was the largest contributor to the industrial source of vanadium emission inventory. NC made the largest contribution, particularly in Hebei with 1587.2 t of cumulative emission due to heavily concentrated steelmaking operation. Moreover, vanadium emission in this sector remained an increasing trend in all regions during 2015-2019 (Fig. 3d), which reflected the robustness of steelmaking industries. In comparison, emission induced by glass products manufacturing were scattered in Hebei in NC, Shandong in EC, Hubei in CC, Sichuan in SWC and Guangdong in SC, with much smaller output. Fossil energy (coal and oil) exploration also made minor contribution to the emission inventory, mainly in NC (Shanxi, Inner Mongolia) and NWC (Xinjiang, Shaanxi). Similarly, vanadium mining activity played rather an insignificant role in overall emission inventory. However, SWC alone accounted

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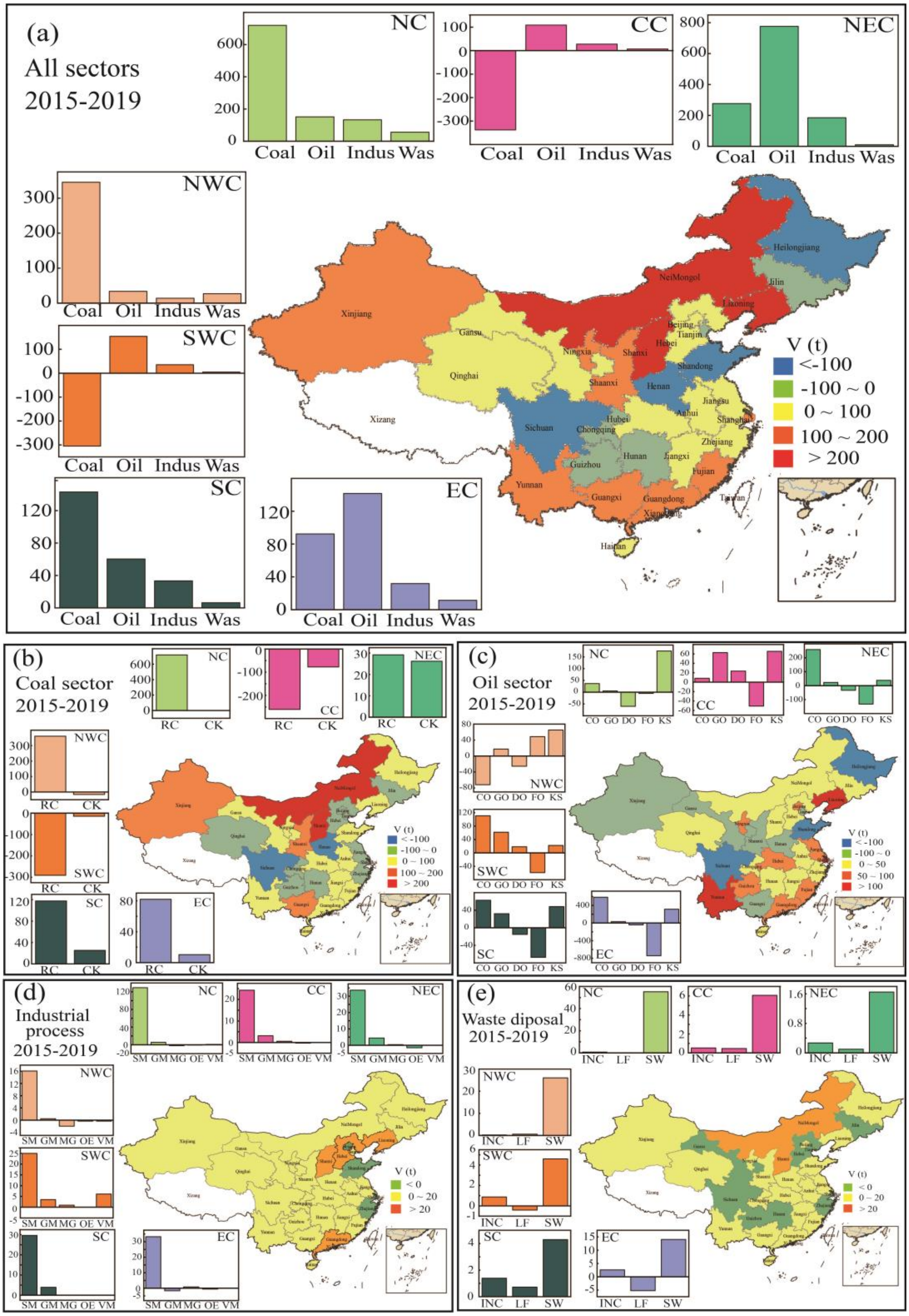

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Figure 3. Spatiotemporal variation of vanadium emission inventories at provincial and regional levels during 2015-2019. Temporal difference in emitted vanadium from (a) all sectors; (b) coal combustion; (c) stationary oil burning; (d) industrial process, and (e) waste disposal for each province and region between 2015 and 2019. The colors of shaded area corresponded with vanadium emission level in different provinces. The inlet box displayed the temporal variation in regional vanadium emission level during 2015-2019.

for 66.1% of emission pertinent to vanadium mining, which was mainly ascribed to intensive mining activities in the world's largest vanadium-titanium reserve, in Panzhihua city, Sichuan. Strong demand for vanadium production persisted, as SWC was the only region experiencing an increase in vanadium emissions related to vanadium mining. Therefore, further investigation into other sectors of the vanadium industry, such as vanadium smelting, vanadium product processing, would be crucial to improve the comprehensiveness of the inventory, as they may have become major industrial sources for vanadium emission. For waste disposal activities, Vanadium emission pertinent to industrial solid waste generation increased in all regions (Fig. 3e). Larger proportion of vanadium output (> 250 t) occurred in NC (Hebei, Inner Mongolia, Shanxi) (Fig. S3e), where both production and consumption of wide range of mineral resources were located, generating a massive amount of slag subjected to leaching processes (Li et al., 2013). For municipal waste, the temporal variation in vanadium emission was insignificant, which may be attributed to an increased recycling rate (Xiao et al., 2018), and new regulation that led to a smaller volume of trash subjected to disposal, such as the ban on imported waste (Song et al., 2021).

Among all regions, NWC was responsible for less proportion of overall vanadium emission. However, emission related to numbers of sources in NWC increased, including the consumption of raw coal, oil burnings, and industrial solid wastes (Fig. 3). A large number of enterprises with intensive energy footprint were relocated to NWC due to incentive policies in order to boost the economic development of western provinces and ease the environmental burden of eastern regions. This move consequently resulted in remarkable growth in vanadium emission over the study period, highlighting the need for more attention in the future regarding its environmental impact.

3.4 Uncertainty analysis of vanadium emission inventory

The successful establishment of inventory was affected by various factors including the authenticity, representativeness, accuracy of data, and adopted statistic methods. To determine the cause of uncertainty and the level of impact it incurred would provide valuable guidance for improving the quality of inventory building. The uncertainties of input data were firstly analyzeed by bootstrapping simulation. Table S19 provided the characterization for fitted probability distribution of bootstrapped samples and the range of uncertainties for emission factors and activities levels. With a few exceptions including raw coal consumption and vanadium content in coke, most samples after bootstrap simulation were normally distributed, suggesting an adequate fit to original samples that well represented the variability and uncertainties. As a result, vanadium release fraction for coke oven and chain grate boiler showed the greatest uncertainties, ranging from -105% to 108%, and -56% to 56%, respectively. The availability of these datasets was extremely scarce and of poor accessibility. In comparison, Activity levels in different sectors were well recorded either by reliable agencies or characterized thoroughly in scientific research, which led to less uncertainties.

Table S20 displayed the quantification results for the uncertainty of vanadium emissions determined by a 95% confidence interval. Among the model outputs, the uncertainties of vanadium emission pertinent to the combustion of raw coal and coke ranged from -47.5% to 63.7% and -49.4% to 53.7%, respectively, which can be considered the major source for uncertainties due to poorly characterized emission factors for pollution control devices and vanadium fraction in flue gas. On the other hand, the inventories for vanadium emission pertinent to oil burning and industrial processes were well established with less than ± 10% uncertainties, thanks to sufficient activity data as well as well-established emission factors with relatively lower distribution variability. Sensitivity analysis was carried out to determine the relative importance of subgroup inventory, applying Pearson correlation to measure the association between subgroup inventory and the total inventory. Vanadium emission pertinent to consumption of coal products (raw coal, coke), stationary sources (crude oil, gasoline, diesel, fuel oil), steelmaking process and MSW disposal (incineration and landfilling) significantly correlated with total vanadium emission (Fig. 4). Therefore, it would be crucial to focus future effort on reducing the uncertainties of corresponded vanadium emission categories to achieve improved vanadium emission inventory.

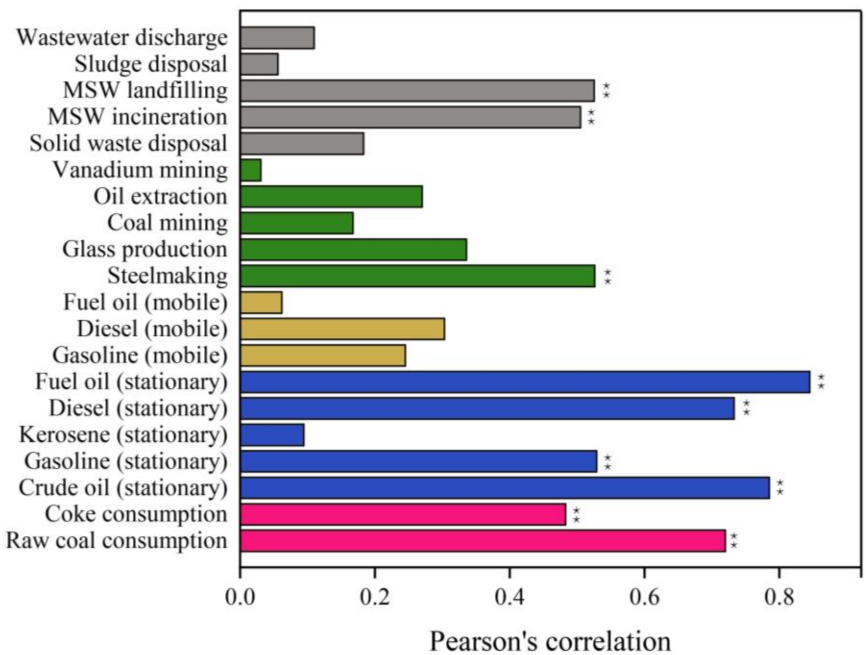

Figure 4. Sensitivity analysis of subgroup emission inventories. The types of anthropogenic sources were distinguished by the color of bars, with asterisk signs indicating the significant correlation (Pearson's method, $p < 0.05$).

To improve the emission inventory, the following measures need to be considered: Perform on-site study for the pollutant control device in raw coal and coke combustion sectors in order to improve the data quality of emission factors; Allocate more focus on vanadium industry (mining, smelting, product manufacturing) by conducting site-based data collection for more accurate inventory development; Improve the clarity of flux model by accounting the variability of emission routes from single anthropogenic source.

4. Conclusions

Our study examined the vanadium emission pattern to develop the China's vanadium emission inventory based on bottom-up approach, comprising five major categories and 20 affiliated sub-groups. Vanadium emission increased from 41389 t to 44069 t during 2015-2019, with the majority of vanadium discharged directly into the atmosphere. Coal combustion and stationary sources of oil burning accounted for 47.5% and 39.6% of overall emission inventory, respectively. Major vanadium emission occurred in northern, eastern coastal, southern and south western provinces, with Shandong, Liaoning, Hebei, Guangdong and Hunan emitting over 10000 t of vanadium cumulatively. Robust economic growth led to a temporal increase in vanadium emission induced by consumption of coal and oil during 2015-2019. The contribution of vanadium mining sector showed strong heterogeneity in spatial distribution, with 66.1% derived from Sichuan province. Emission pertinent to raw coal and coke combustion was the main sources of uncertainty for the inventory development, which can be attributed to poorly characterized emission and operation data for boiler operation.

Data availability

Datasets used for development of vanadium emission inventories were uploaded in Github and can be assessed through doi:10.5281/zenodo.10565660.

Author contributions

BZ reviewed and edited the paper; YX supervised the project. XH collected the data sets and performed analysis, with contribution from HZ, BJ and QL. HZ wrote the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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