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

Han Zhang[1,2], Baogang Zhang[2,*], Bo Jiang[1], Qimin Li[2], Xuewen Hu[2], Yi Xing[1,*]

[1] School of Energy and Environmental Engineering, University of Science and Technology Beijing, Beijing, 100083, China
[2] MOE Key Laboratory of Groundwater Circulation and Environmental Evolution, School of Water Resources and Environment, China University of Geosciences Beijing, Beijing 100083, China

Correspondence to: Baogang Zhang (baogangzhang@cugb.edu.cn) & Yi Xing (xingyi@ustb.edu.cn)

Abstract. Anthropogenic activities are the major contributor to vanadium pollution in surficial environment, but quantitative assessment of anthropogenic vanadium emission across environmental receptors remain scarce. This study develops nationwide vanadium emission inventory in China during 2015-2019, covering five major anthropogenic sources: coal combustion, stationary oil burning, transportation, industrial production, and waste handling. Cumulative flux model reveals emissions of 211095 t, 3725 t, and 0.1 t of vanadium into atmosphere, soil and water during 2015-2019, respectively, highlighting the atmosphere as the primary vanadium receptor. Coal combustion (46.7%) and stationary oil burning (40.1%) are the largest vanadium contributors. Emission pattern is mainly shaped by regional activity levels, with Shandong, Liaoning, Hebei, and Guangdong contributing 35.8% of national inventories. Shanxi and Inner Mongolia, designated as energy producing bases, dominate coal combustion related emission. Beijing-Tianjin-Hebei achieves significant emission reductions due to coal restriction policies and adoption of cleaner energy and advanced end-of-pipe system. In economically developed coastal regions, coal consumption has decreased, while oil consumption makes greater contribution due to industrial modernization. Moreover, steelmaking operation accounts for 88.2% emission in industrial production, and continue to increase nationwide. Emissions induced by vanadium mining shows remarkable spatial heterogeneity, with 66.1% output in Southwestern China. Inventory uncertainties are driven by insufficiently resolved activity data, poorly characterized emission factors, and variability in vanadium content in raw materials, particularly in coal combustion, transportation, and waste disposal sectors. These findings underscore atmosphere as the main vanadium receptor and identify the primary contributors, which is helpful to develop targeted mitigation strategies.

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, totalling 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 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 redox-flow 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 various 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 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 environmental mediums (Wang et al., 2020a; Zhang et al., 2020; 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 vanadium levels in local aerosol samples are as high as 228 ng/m^3 (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), significantly higher than that for coal burning (0.001 kg/t), petroleum consumption (0.06 kg/t), and steelmaking (0.0001 kg/t) (Anderson, 1973). However, the quantitative evaluation of vanadium flux derived from various anthropogenic sources to environmental receptors remain lacking, especially at the continental scale. Moreover, although China have promulgated the Air Pollution Prevention and Control Action Plan (Clean Air Act) since 2014 to reduce industrial emissions (Zheng et al., 2018), its specific impact on vanadium emission trends has yet to be thoroughly explored (Zheng et al., 2018).

Emission inventories are essential tools to elucidate the magnitude, spatial and temporal patterns of pollutant emission from different various sectors, serving as a foundation for policies aimed at reducing environmental impact. 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, influenced by ever-changing market demand and improvements in pollution control technologies, resulting in low-resolution 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 sector and regional specific solutions for mitigating the impact 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 in China for the period of 2015 – 2019, following the introduction of "Clean Air Act". The study encompasses three primary tasks: (1) performing the modelling study to quantify the contribution of anthropogenic sources to environmental receptors; (2) evaluating the temporal and spatial pattern of anthropogenic vanadium release from different sectors across years and regions (provinces); (3) conducting the uncertainty analysis to assess the reliability of the inventory model. This comprehensive approach aims to provide a detailed understanding of vanadium emissions, supporting efforts to mitigate their environmental impacts.

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 (Table 1), which was 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
Oil extraction (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

For each emission source, activity data at provincial level were retrieved from yearbooks of China's industrial activities, including coal (Appendix SS-A), energy (Appendix SS-B), industrial and resource production (Appendix SS-C), transportation (Appendix SS-D), and waste disposal (Appendix SS-E). Additional statistics on socioeconomic information, government policies, and global warming potentials were also collected from published data of National Statistics Bureau (Appendix SS-F, SS-G, SS-H). All appendix can be accessed in Zenodo depository (Zhang, 2024). For transportation derived emission, the yearly volume of national oil consumption was applied due to lack of provincial data. All dynamic emission factors were retrieved from previous reported values, and assumed consistent across all provinces. All activity datasets were collected during the period of 2015-2019, which may reflect the effects of air pollution mitigation practices implemented since the introduction of "Clean Air Act".

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

$$EV = \sum j(EFj \times APj) \quad (2\text{-}1)$$

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 amounts of industrial products (or wastes) in metric ton.

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

$$EV = \sum i(EFi \times EPi) \quad (2\text{-}2)$$

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,

$$EV = EP \times AP \quad (2\text{-}3)$$

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 inventories was provided in full details in the Supporting Information, with activity level data and dynamic emission factors 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 with 10000 times iterations. For sensitivity study, the linear relationship between input parameters and emission levels were evaluated using Pearson rank correlation coefficient, ranging from -1 to 1. Any input parameter with the absolute coefficient values closer to 1 indicate strong correlation, which may suggest the source of uncertainties (Liu et al., 2021). All analysis computations were performed in AuvToolPro2.0 following the protocol (Zheng et al., 2017).

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, anthropogenic processes mobilized a total volume of 214820.1 t of vanadium into the environment, with a yearly average of 42964 t. Based on a global inventory developed during 2015-2016, 730000 t of vanadium was released into environment due to anthropogenic activity (Schlesinger et al., 2017). As a major energy consumer and industrial advanced nation, China's yearly emission is estimated to comprise 6% of global emission output based on comparison with the global result. Among different receptors, 98.3% vanadium generated from anthropogenic processes was directly discharged into the atmosphere, leading to a total emitted vanadium volume of 211095 t. Most atmospheric vanadium was derived from coal combustion, stationary oil burning, and transportation, corresponding to a relative contribution of 46.7%, 40.1%, and 8.2%, respectively. Globally, 82.2% of vanadium was directly released into atmosphere during 2015-2016 (Schlesinger et al., 2017), with 54.6% and 24.6% of emission from combustion of oil products and coal, respectively. 3725 t of vanadium (1.7%) was directly deposited into China soil, which was predominantly attributed to industrial solid waste discharge and sludge disposal, totalling 1398 t and 1425 t, respectively. In comparison, global inventory assumed all mining and industrial production as the major source of emission flux to the soil (Schlesinger et al., 2017), with a relative contribution of 18.5%. 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. Both China and global inventory highlighted the dominance of atmospheric flux. However, while coal combustion was the predominant source for atmospheric emission in China, combustion of oil products weighed more importantly for global inventory. There was also a marked difference in estimated emission flux to soil, with a greater soil flux due to global industrial and mining activities. In contrast, the presented inventory showed a much smaller proportion of vanadium flux to soil receptor, because it was assumed that the majority of emission, from the large contributors such as steelmaking and glass production, was released into atmosphere. The development of global inventory employed mainly the available data of industrial activities in the United States, which lacked sufficient data resolution for developing nations. However, coal combustion played more important roles in China's energy sector than petroleum. The contrast could also be attributed to variation in vanadium releasing fraction, emission reducing technology, vanadium content in raw materials in different regions, as well as assumptions made during inventory development. To improve the inventory, it is essential to perform local level investigation to increase the spatial and temporal specificity.

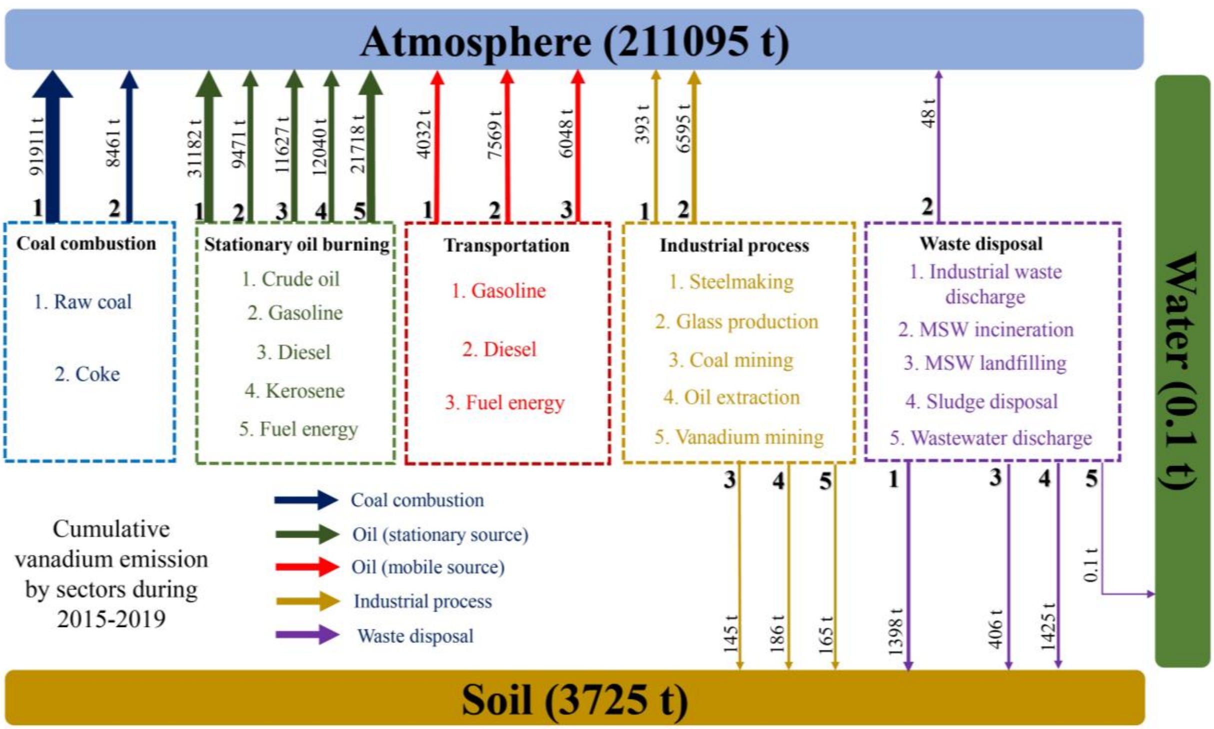

Figure 1: Environmental flux model for vanadium directly discharged from anthropogenic activities into environmental receptors. Vanadium flux data was calculated cumulatively during 2015-2019 from different anthropogenic sectors with vanadium emission, i.e., coal combustion, stationary source oil burning, 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). Emission originated from raw coal combustion accounted for 91.6% of subgroup inventory pertinent to coal consumption (Fig. S2), which exhibited a steady increasing trend from 18096 t to 18856 t during 2015-2019 (Fig. 2b), corresponding to a 2% decrease in relative contribution to the total emission inventory. Globally, the annual vanadium emission from coal combustion in 2016 was estimated to be only one fourth of the output in the previous decades (Schlinger et al., 2017). This reduction was largely due to the emission reduction measures implemented in developed nations, such as the United States and Western Europe since 1980s (Environmental Health & Engineering, Inc., 2011; Arienzo et al., 2021). Unlike the developed nations, China remained heavily reliant on coal, accounting for 51% of world annual coal consumption (Wang et al., 2020d). This dependency was further compounded by the continued expansion of installed power generation capacity (Wang et al., 2020e). Additionally, the average vanadium concentrations in China's raw coal were higher than the global average levels (Bartoňová et al., 2023), resulting in a disproportionately large contribution to global vanadium output and led to more challenges associated with domestic emission. Vanadium emission pertinent to coke consumption made up of 8.4% of total coal induced emission, and it showed a decreasing trend from 1789.9 t to 1743.6 t, corresponding to a decrease in relative contribution by 8.5%. This downward trending corroborated China's increasing efforts to enhance the energy efficiency of energy sectors, sintering and steelmaking processes under the "Clean Air Act" (Zheng et al., 2018; 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), which led to an increase in relative contribution to total inventory by 9.7%, 6.4% and 28%, respectively. Compared with inventory derived from global oil consumption, the annual vanadium output in China accounted for smaller proportion (~6.1%) of global vanadium emission in 2016 (Schlesinger et al., 2017), which has also increased by 2.5 folds since 2000 (Monakhov et al., 2004). The rapid economic growth during this period has driven up the oil refining activities (Zhang et al., 2019b). However, the vanadium emission pertinent to fuel oil combustion decreased significantly, which was possibly because of successful implementation of emission trading scheme focused on energy-intensive heavy industries (Ouyang et al., 2020). The reduction in fuel oil derived emission also aligned with the overall decline in global demand (International Energy Agency, 2016), likely attributed to the substitution of residual oil with use of natural gas in refinery activities (Visschedijk et al., 2013). This trend suggested that regulations in various countries prioritized limiting emissions from vanadium rich heavy oil.

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), along with change in relative contribution to the inventory by 30.6%, -2.0%, and 37.9%. Yearbook of Transportation and Automobile Industry 2015-2019 recorded the significant growth in total volumes of

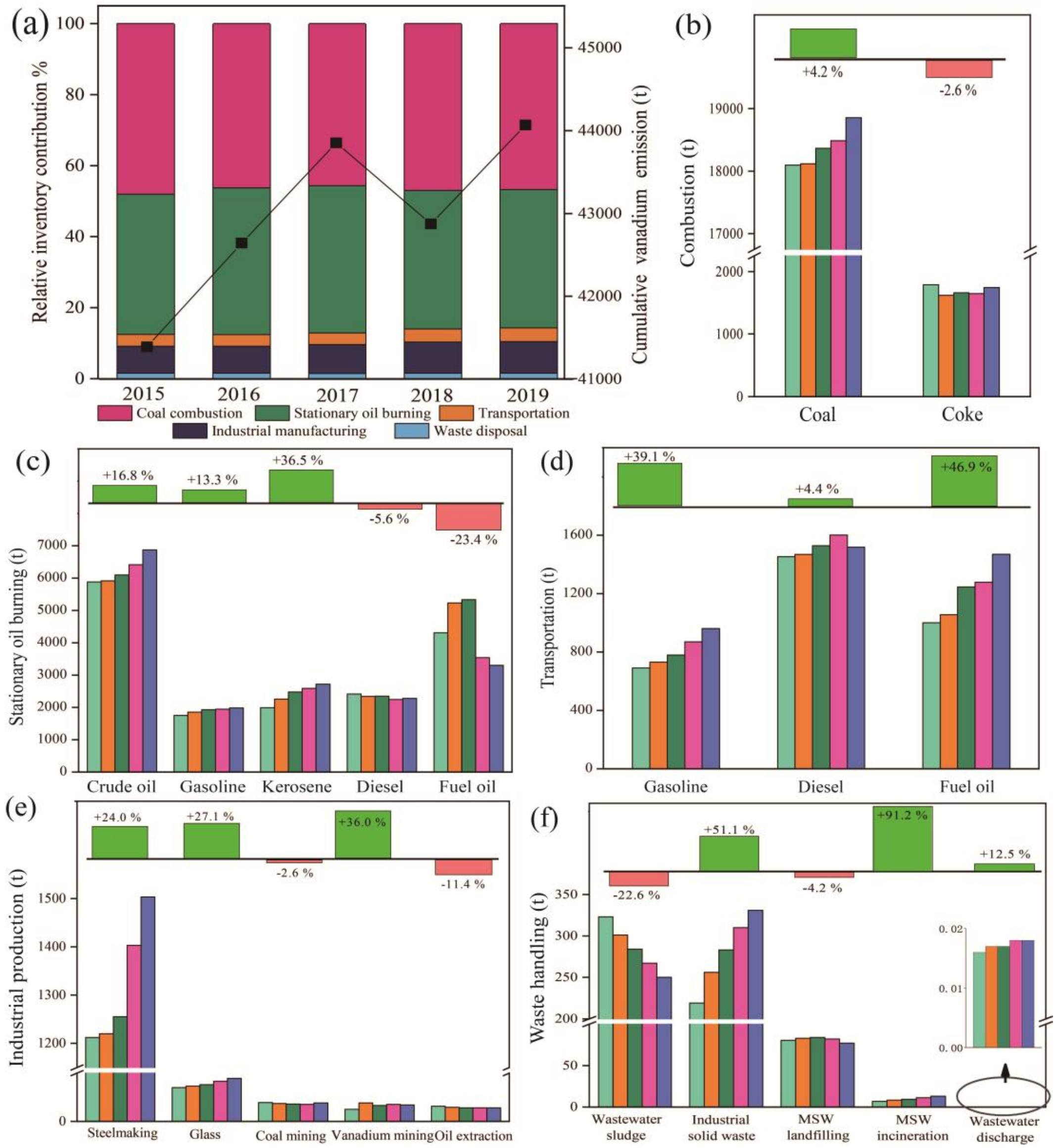

Figure 2. Historical trend of vanadium emission inventory during 2015-2019. (a) The relative contribution of yearly cumulative vanadium emitted from each major anthropogenic source was calculated, along with total emission amount; the yearly vanadium emission from subgroups of (b) raw coal combustion, (c) stationary oil burning, (d) transportation, (e) industrial production, and (f) waste handling.

on and off-road transportation, which may be the leading cause of major raise in vanadium emission associated with gasoline consumption. For heavy duty vehicles, sulfur emission remained nearly unchanged despite increased logistic activities (Zheng et al., 2018). The trend can be attributed to the strict regulation of vehicle diesel standard (GB-19146) introduced after 2013, which in turn reduced the demand in vanadium-based catalyzer (Bai et al., 2021). In maritime transportation, fuel oil was primarily utilized by shipping vessels. Although global vanadium emissions from maritime transportation decreased due to sulfur restrictions (IMO MARPOL Annex VI) imposed by International Maritime Organization in the early 21st century (Arienzo et al., 2021), China's inventory showed a progressive increase in vanadium output. This suggested that the expanded shipping activities has offset the effects of stricter sulfur emission controls.

In other sectors, 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 significantly increasing trend from 1212 t to 1593 t during 2015-2019 (Fig. 2e), with a 16.5% rise in relative contribution. In the recent decade, China undertook strong action to reduce the overcapacity in steel industry, resulting in a slow production growth (Zhu et al., 2022). The implementation of ultralow emission transformation (ULET) markedly reduced emission of PM2.5 and other pollutants (Tang et al., 2020). However, vanadium emission continued to rise, making up of 85% consumption in industrial production sector (ResearchInChina, 2018). The new standard for high strength rebar required more vanadium for alloying process (Polyak, 2019). In contrast, emission from mining and oil exploration in China did not show significant growth, consisting with the global trend (Schlesinger et al., 2017). The relative contribution to China emission inventory from coal mining (2.5%) and oil extraction (2.2%) were significantly lower than global inventory, where oil exploration and mining activity accounted for 56% and 18% of emission output, respectively (Schlesinger et al., 2017). Such contrast could be explained by higher level of data resolution in China's inventory, capturing a more detailed scope of mining and oil extraction activities, whereas the global inventory used more generalized data and assumptions to fill the data gap, leading to a broader, less precise estimates. For vanadium ore mining, the overall vanadium production remained stable in major producing powers such as Russia and South Africa (Polyak, 2015; Polyak, 2017; Polyak, 2019). However, the newly adopted environmental policy placed a ban on importation of vanadium slags in China (Polyak, 2019), which may stimulate the demand for domestic vanadium production. Among waste disposal sectors, 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, with remarkable drop in relative contribution by 27.3%. The National Soil Act published in 2016 regulated sludge 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). Vanadium emission pertinent to landfill and MSW incineration were relatively stable during 2015-2019. The disposal of industrial solid waste increased vanadium output from 249.1 t to 331.1 t (Fig. 2f), incurring a significant increase in relative contribution by 41.9%. Recycling rate of steel slags in China was at only 20-30%, compared to a much higher utilization rate of 97% in developed nations (Yang et al., 2024). 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.

The large-scale installation of air pollution control devices (APCDs) commenced since 2004 (Wang et al., 2020e). After the introduction of "Clean Air Act", the energy and industrial manufacturing sectors underwent a decreasing trend in pollutant emission (e.g., SO_2, NO_x) (Zheng et al., 2018), and transportation emission (e.g., NO_x, CO, NMVOC) remained relatively steady. Despite these achievements, the overall activity levels continued to rise, along with vanadium output. The implementation of ultra-low emission (ULE) standards mandated the retrofitting of power plants with more efficient air pollutant control devices (APCDs), driving widespread technological upgrade. According to APCDs statistics (Appendix: SS-A-3), electrostatic precipitators have remained prevalent during study period due to the cost effectiveness. In comparison, a remarkable increase in application rates of baghouse filters and wet flue gas desulfurization was observed compared to previous report (Liu et al., 2015). Moreover, catalytic reduction systems for NO_x removal were increasingly incorporated into newly commissioned facilities (Liu et al., 2019). However, catalytic reduction process often involved vanadium-based catalyst, potentially contributing to an increase in atmospheric emission. Despite advance in APCD's implementation, the growing activity level driven by elevated energy consumption and demand for vanadium in various sectors may offset these improvements, underscoring the complex interplay between technological progress and industrial expansion.

3.3 Provincial level of vanadium emissions and spatiotemporal distribution pattern

To facilitate further investigation of the 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 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. Shandong, Liaoning, Hebei, Guangdong, and Hunan were among the largest provincial emitters with cumulative vanadium discharge over 10000 t (Fig. S3a), accounting for over 35.8% of total national emission in combined.

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 (> 4500A 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 NC, Shanxi (420 t) and Inner Mongolia (358 t) experienced the largest increases in coal derived emission. According to Energy Development Strategy Actional Plan 2014-2020, both provinces were designated as major bases of large-scale thermal power generation, contributing significantly to the national energy supply network through long-distance electricity transport. Both provinces have faced economic challenges with one of the lowest GDP growth and income rises in China (Appendix SS-F), making energy substitution unlikely. In contrast, Beijing and its surrounding areas have achieved substantial reductions in coal use through Regional Collaboration on Air Pollution Control, aiming to reduce 25% of PM2.5 concentrations in Beijing-Tianjin-Hebei by phasing out small coal-fired boilers and switching to natural gas (Yan et al., 2018). Additionally, according to statistics (Appendix SS-A3), NC has one of the highest application rates of APCDs including the advanced baghouse filters, which has the highest capture efficiency for fine particles. Vanadium emission

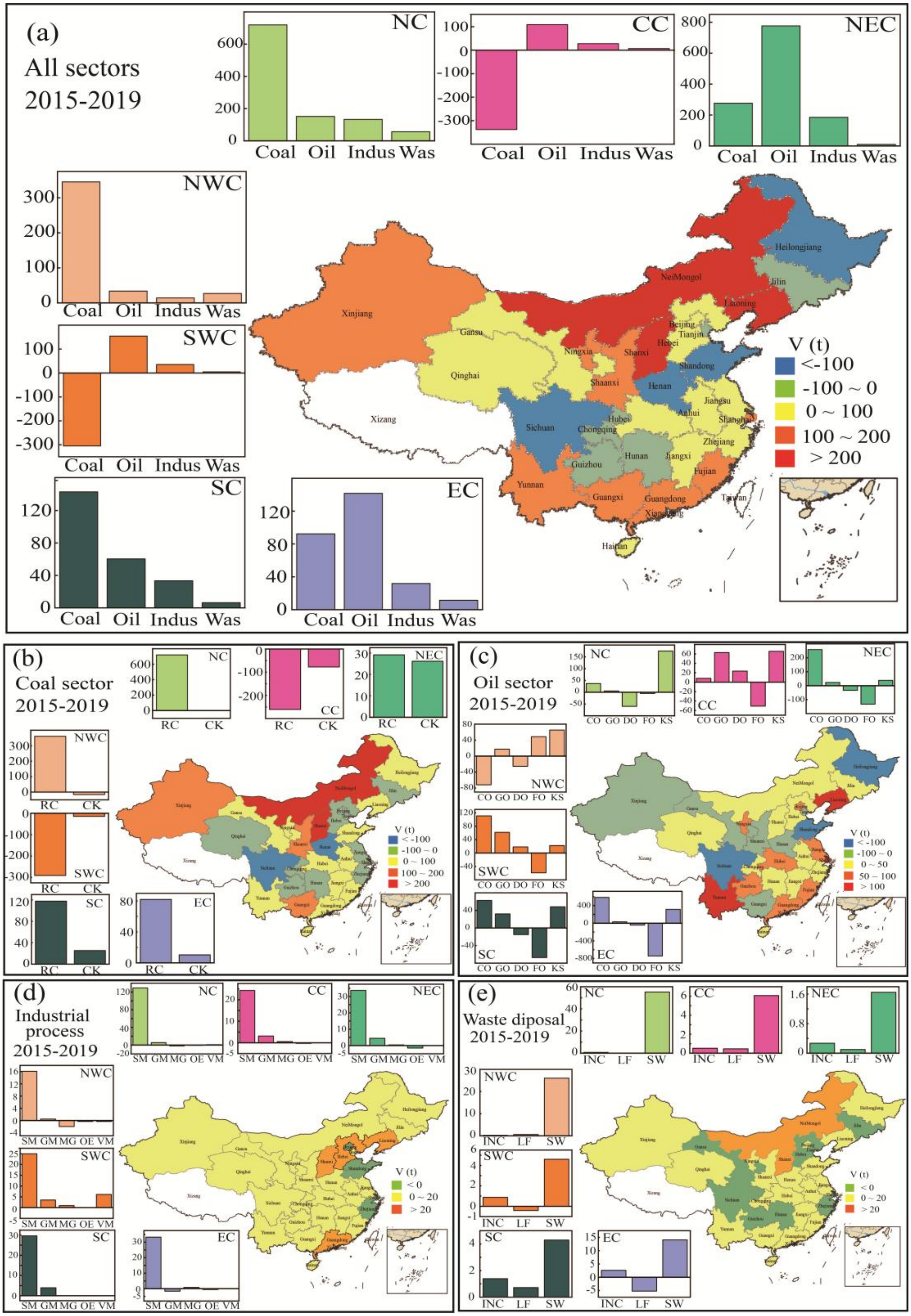
(a)
All sectors
2015-2019
NC
CC
NEC
NWC
SWC
SC
EC
Coal Oil Indus Was
V (t)
<-100
-100 ~ 0
0 ~ 100
100 ~ 200
> 200
Xinjiang
Xizang
Qinghai
Gansu
NeiMongol
Heilongjiang
Jilin
Liaoning
Beijing
Tianjin
Hebei
Shanxi
Ningxia
Shaanxi
Shandong
Henan
Jiangsu
Anhui
Shanghai
Hubei
Sichuan
Chongqing
Zhejiang
Guizhou
Hunan
Jiangxi
Fujian
Yunnan
Guangxi
Guangdong
Taiwan
Hainan
(b)
Coal sector
2015-2019
RC CK
V (t)
< -100
-100 ~ 0
0 ~ 100
100 ~ 200
> 200
(c)
Oil sector
2015-2019
CO GO DO FO KS
V (t)
< -100
-100 ~ 0
0 ~ 50
50 ~ 100
> 100
(d)
Industrial
process
2015-2019
SM GM MG OE VM
V (t)
< 0
0 ~ 20
> 20
(e)
Waste diposal
2015-2019
INC LF SW
V (t)
< 0
0 ~ 20
> 20

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.

decreased notably in SWC (-290 t) and CC (-269 t), with Sichuan, Guizhou, Chongqing, and Henan, Hubei exhibiting significant decline in coal consumption (Appendix SS-A5). Coal produced in SWC region was highly enriched with toxic arsenic (Wang et al., 2020b), which may invoke more restrictions on local consumption. There was also increased investment on greener energy such as natural gas adopted by CC (e.g., Henan) to reduce their dependence on coal (Wang, 2021). SWC region significantly reduced coal consumption while maintained a steady growth of power generation (Appendix SS-H1 and H2). This pattern coincided with increased investment on shale gas supply, particularly in Sichuan province with one of the most natural gas consumptions in China. Similarly, in delta area (Shanghai-Jiangsu-Zhejiang) of Yangtze River, the most economically developed region with the highest GDP and income per capita (Appendix SS-F), a significant decline in emission related to coal burning was observed, which aligned with a rapid increase in natural gas consumption (Appendix SS-H3). 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). Provinces like Shandong and Guangdong, as economic powerhouses with populations exceeding 100 million, benefited from seaport advantages, enabling large-scale oil imports to fuel industrial and economic growth, ultimately leading to increased vanadium emissions. Direct vanadium emission from industrial production processes were much less, but exhibited a heterogeneous distribution pattern (Fig. S3d). The steel industry was the primary source of emission inventory related to industrial production, with the largest emission from Hebei province (1587.2 t) due to concentrated steel making operations. In comparison, small emissions induced by glass production were scattered in Hebei in NC, Shandong in EC, Hubei in CC, Sichuan in SWC and Guangdong in SC. Fossil energy (coal and oil) extraction also made minor contribution to the emission inventory, mainly in NC (Shanxi, Inner Mongolia) and NWC (Xinjiang, Shaanxi). For vanadium ore mining, SWC had the world's largest vanadium-titanium reserve in Sichuan, which alone accounted for 66.1% of emission, with persistent growth in production level. Therefore, further investigation into vanadium industry, such as smelting and product processing, would be crucial to improve the comprehensiveness of the inventory, as they may have become major industrial sources for vanadium emission. For industrial waste disposal, vanadium emission pertinent to industrial solid waste disposal increased in all regions (Fig. 3e). Larger proportion of vanadium output (> 250 t) occurred in NC (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). However, the emission in Hebei experienced a decreasing trend. Since the introduction of the Steel Industry Adjustment and Upgrading Plan (2016), the elimination of inefficient production processes decreased the generation of slag and dusts, the consolidation of industry into larger enterprise further encouraged the recycling and re-utilization of solid waste, such as converting slag into construction materials or recovering vanadium. 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). The National Main Functional Zone Plan (2010) explicitly designated the western region of China as a base for energy resource development and heavy industry. Local governments in the western region have introduced investment incentives to attract energy-intensive enterprises to establish operations, 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.

The policy changes usually imposed more direct impact on coal consumption. In coastal region with strong economy and public awareness, the enforcement of environmental policies has become very urgent because wealthier populations tend to be more concerned with environmental well-being, and these regions have more financial and technological resources for implementing environmental policies. For example, coastal region made huge investment on liquified natural gas terminals and processing facilities, which can support the steady growth in natural gas usage. However, for poorer provinces, there is less incentive to prioritize environmental concerns, as the immediate focus is typically on supporting economic development. These regions may rely on cheaper energy sources, such as coal, to fuel industrial growth and meet the energy demands of expanding economies. The development of vanadium emission inventory at provincial level will serve as key basis to policy making, which would warrant more targeted efforts on major provincial emitters for different emission source.

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. Identifying the sources of uncertainty and assessing their impacts provide valuable insights for improving the inventory quality. The uncertainties of input data were firstly evaluated 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. Most samples after bootstrap simulation were normally distributed, suggesting an adequate fit to original samples that well represented the variability and uncertainties. Table S20 displayed the quantification results for the uncertainty of vanadium emissions determined by a 95% confidence interval. Among the model outputs, the inventories for vanadium emission pertinent to oil burning and industrial processes were well established with less than ± 10% uncertainties. However, emission pertinent of coal combustion, transportation, and wastewater sludge exhibited great level of uncertainties. Pearson’s correlation coefficients were further computed to measure the linear relationship between input parameters and emission inventories. For subgroup inventories with high level of uncertainties, transportation related oil consumption and wastewater sludge generation were significantly correlated ($p < 0.05$) with emission levels (Figure 4A&4B). Both sectors lacked activity level data at provincial level, resulting in imprecise or insufficient data resolution for inventory development. For coal combustion, vanadium release fraction of typical boilers exhibited significant correlation ($p < 0.05$) with emission level. Some release fraction data were retrieved from old studies from other countries, which may not well-represent the overall status in China. Vanadium content in raw coal and coke was another influential factor ($p < 0.05$), as its high variability may introduce significant uncertainty in the emissions calculation. The application rates of electrostatic precipitator, bag filter, and wet method of flue gas desulfurization showed stronger correlation with emission inventory, which may serve as potential source of uncertainties. It was also challenging to access the temporal and spatial data on technology penetration for APCDs. The data profile assumed that application rates remained unchanged during the study period, and extrapolation was made for old-fashioned technologies (e.g., electrostatic precipitator) based on previous report (Liu et al., 2011). To reduce the uncertainty level, more focus should be allocated to improve the data resolution of transportation related activity levels. It is also vital to perform on-site measurement for domestic facility process in raw coal and coke combustion sectors in order to improve the data quality of emission factors. For example, previous study employed continuous emission monitoring system to provide real-time tracking on emission for computation of emission factor (Tang et al., 2020).

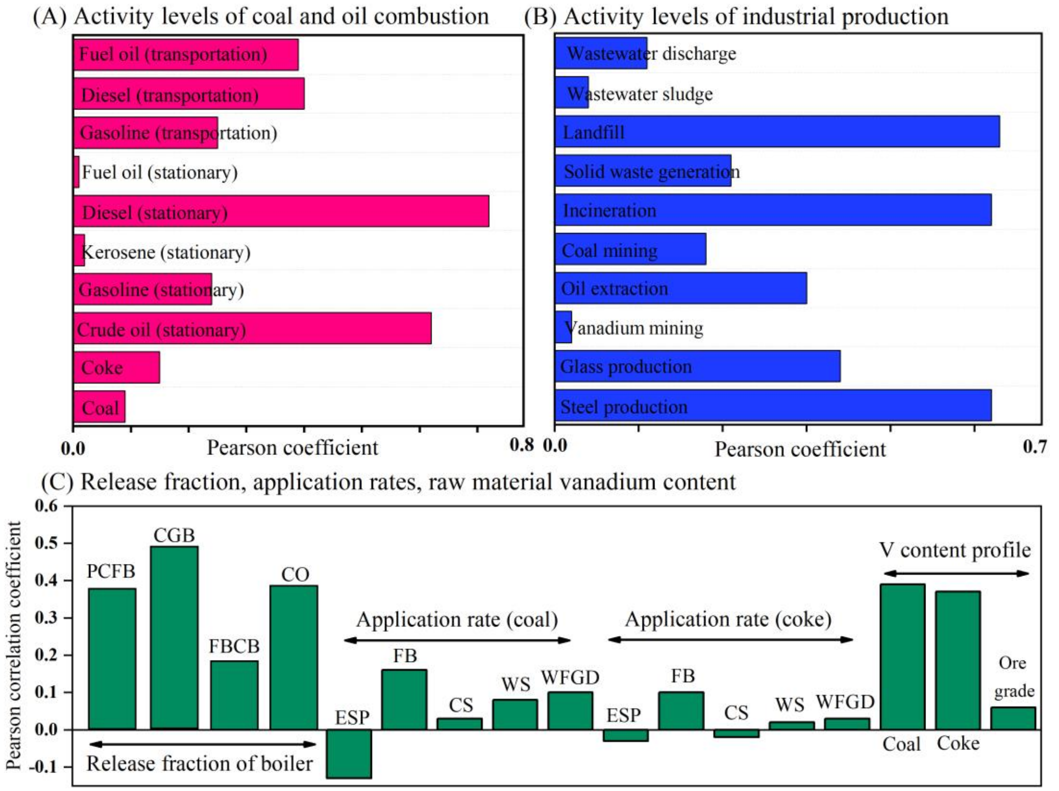

Figure 4. Sensitivity analysis of input parameters using Pearson rank correlation coefficient. The strength and direction of linear relationship between subgroup emission and input parameters related to (A) consumption level, (B) production level, and (C) other emission factors, were measured using Pearson correlation method (abbreviation was used as following: ESP – electrostatic precipitator, FB – baghouse filter, CS – cyclone separator, WS – wet scrubber, WFGD = wet method flue gas desulfurization, PCFB – pulverized coal fired boiler, CGB – chain-grate boiler, FBCB – fluidized bed combustion, CO – coke oven).

3.5 Compared with previous domestic study

In a previous study on historical trend of vanadium emission in China (Bai et al., 2021), the emission from coal combustion significantly declined following a peak in 2007, with emission levels in 2010s lower than this study. This contrast may largely stem from differences in collection and handling of APCDs data. According to China Electricity Council, major technological upgrades since 2010s led to a sharp increase in implementation of baghouse filter and wet method desulfurization. However, we assumed that their annual installation rates have stabled in recent years. Notably, the study on vanadium removal efficiencies of baghouse filter (79.0 ± 17.9%) was extremely scarce and may be seriously undermined compared to conventional electrostatic precipitators (89.7 ± 13.7%). In the previous study, the relative contribution of both stationary and mobile source showed a decreasing trend until 2017. In comparison, oil consumption data in this inventory showed a persistently increasing trend in all sectors except for fuel oil and diesel. For transportation emission, our study utilized the amount of fuel consumption reported by national statistics bureau, whereas vehicle counts were integrated into the emission calculation in previous study. It should be noted that the substantial rise in electrical and natural gas-powered vehicle after 2015 may affect the emission calculation. Therefore, difference in data source may contribute to the discrepancy between inventories. Moreover, both studies agreed on significant increase in emission pertinent to industrial production and solid waste disposal, highlighting the contribution of steel and glass production to the rise of vanadium output. However, the present study also underscored the importance of solid waste disposal, which were neglected in the previous study.

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 46.7% and 40.1% 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. This inventory highlighted the significant role of regional economic development and energy demand in shaping vanadium emission patterns. Advanced policies in economically developed areas reduced emissions by enabling cleaner energy transitions, while energy-dependent regions showed persistent coal-related emissions. The analysis revealed critical uncertainties in emissions from transportation, coal combustion, and sludge disposal, largely due to insufficiently resolved activity data and poorly characterized emission factors. These uncertainties underscored the need for region-specific data collection and on-site measurements, particularly for vanadium release fractions and APCD efficiencies. Overall, this study provides an essential framework for tracking vanadium emissions, identifying hotspots, and guiding policy interventions. Future efforts should focus on integrating seasonal and higher-resolution datasets to further refine the inventory and address emerging challenges in emission control and mitigation strategies.

Data availability

Datasets used for development of vanadium emission inventories were uploaded in Github and can be assessed through 10.5281/zenodo.14467726.

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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