

Article

Comprehensive Analysis of External Dependency in Terms of Material Criticality by Employing Total Material Requirement: Sulfuric Acid Production in Japan as a Case Study

Shoki Kosai ^{1,*} , Seiji Hashimoto ², Kazuyo Matsubae ³, Benjamin McLellan ⁴  and Eiji Yamasue ¹ 

¹ Department of Mechanical Engineering, College of Science and Engineering, Ritsumeikan University, Shiga 525-8577, Japan; yamasue@fc.ritsumei.ac.jp

² Department of Environmental Systems Engineering, College of Science and Engineering, Ritsumeikan University, Shiga 525-8577, Japan; shashimo@fc.ritsumei.ac.jp

³ Graduate School of Environmental Studies, Tohoku University, Sendai 980-8577, Japan; matsubae@m.tains.tohoku.ac.jp

⁴ Graduate School of Energy Science, Kyoto University, Kyoto 606-8501, Japan; b-mclellan@energy.kyoto-u.ac.jp

* Correspondence: kosai0203@gmail.com; Tel.: +81-80-5308-2146

Received: 14 January 2018; Accepted: 7 March 2018; Published: 12 March 2018

Abstract: Securing stable material supply is of paramount importance since materials are fundamental to the economy and human well-being. The domestic production to consumption ratio has conventionally been utilized as a main index for external dependency in terms of material supply security and its criticality. However, the conventional approach confines its application to raw materials, which potentially risks reaching short-sighted conclusions in the policy-making process. Beyond the conventional analysis of external dependency, the development of a more applicable approach for every material is of paramount importance to consider the diversification of external dependency more comprehensively and to aid the analysis of overall material criticality. As such, this paper establishes a new methodology for analyzing external dependency related to every material and product by using the concept of total material requirement (TMR). Applying the methodology, the external dependency of sulfuric acid production in Japan is evaluated as a case study. Previously unexamined elements associated with external dependency in the conventional approach are revealed by this new comprehensive approach. The new approach may be of use to policymakers in designing more sophisticated and well-grounded material securement policy.

Keywords: external dependency; total material requirement; country concentration; supply risk; criticality; mining

1. Introduction

In recent decades, the acceleration of industrialization in emerging and developing nations and the rapid expansion of the world's population have dramatically affected the global material landscape [1]. Global material consumption increased by approximately 60% from 1990 to 2009, exceeding 70 Gt per year [2]. On the basis of historical trends, material demand is expected to continue to increase in the future [3]. Securing a stable material supply is of paramount importance, since materials are fundamental to the economy and human well-being, and their utilization and production have important impacts on the environment.

Raw materials have been widely highlighted in the criticality assessment for securing material utilization. Given that the nature of raw material criticality is equivocal, multi-dimensional indices have been used for its quantitative evaluation. Among the well-known dimensional combinations are the two axes of criticality assessment, comprising supply risk and vulnerability to supply disruption [4–6]. There are several research reports additionally including the environmental impact to formulate the three axes [7–10]. Meanwhile, another promising dimensional combination is developed, comprising supply risk and economic importance [11–14]. Despite the concept ambiguity, there seems to be an agreement that raw material criticality assessment is highly associated with the security of material supply. Other authors have also adopted the security of material supply as a key component in their own individual frameworks of criticality assessment [15–17]. In fact, the interest in quantitative analysis on the security of raw material supply has been growing significantly as a research topic. Rosenau-Tornow et al. quantitatively evaluated supply risk by proposing indicators for market assessment [18]. Dewulf et al. integrated both regulatory and social aspects in the discussion of supply disruption risk [19]. Moss et al. developed the concept of security of supply based on both market factors and political factors to deploy energy technologies [20]. Achzet and Helbig reviewed research papers on supply risk and identified the related 20 indicators [21].

For the evaluation of raw material supply risk, the severe dependency on foreign countries for raw material production is particularly focused on as a key factor in most research reports [21]. Most industrialized economies are critically reliant on continuous supplies of raw material imported from overseas due to scarce domestic deposits [22], which has increasingly shifted the balance between the supply and demand of raw materials [18,23]. Under the current landscape of raw material supply, for some materials, mining production is significantly concentrated in a small number of nations, and in some cases the vulnerability of political stability is seen as a critical concern [24]. Furthermore, regulation of mining and production may be tightened due to the social, economic, and ecological issues in the raw material mining nations and areas [25]. These factors potentially lead to the threat of supply disruption. In fact, the country concentration in production with the inclusion of a political instability index has been the most frequently selected indicator among the various proposed indicators [8,11,18,26].

In addition to the supply-side concentration of raw material producers, the demand-side risk of dependency on imports of various raw materials has been also considered. Several countries and regions such as the United States, Japan, and the EU [15,27,28] have undertaken evaluations of their raw material dependency. The analysis of external dependency of a given country is essential to understanding the mechanism of security of material supply more practically.

The basic concept of evaluating external dependency is developed based on the calculation of raw material production and consumption, particularly associated with mining, in the importing countries. The interest of research has reached the stage of applying raw material criticality assessment to the analysis of complex products such as cars [29]; however, this approach cannot be simply applied to down-stream materials and commodities. At every step of the supply chain for manufacturing materials and products from mining outputs, a significant amount of other materials and energy is utilized in the intermediate processes. There is a high probability that these materials and energy and even the processes themselves will be supplied or undertaken by third-party countries. Given that these overall processes are inevitable for the production of commodities, the external dependency arising from the intermediate processing has to be taken into account in the same way as the country concentration associated with mining production. Different from the conventional analysis of external dependency, the development of a more encompassing approach for every material is of paramount importance to consider the diversification of external dependency more comprehensively and to aid the analysis of overall material supply security.

An appropriate index needs to be chosen for the evaluation of material circumstances and supply security. There are several reported indices such as the concentration of carbon dioxide or sulfur dioxide emissions for material supply, as well as the aforementioned production quantity [30]. In this paper,

total material requirement (TMR) is proposed as a more encompassing material intensity associated with mining [31,32]. TMR involves the evaluation of the hidden flows arising from non-economic activities as well as direct and indirect inputs arising from economic activities. TMR has been assessed at every material level, including phosphorus [33–35], steel [36], ethylene [37], aluminum [38], platinum [39] and fossil fuels [40]. Every material, even that which requires complicated processes, can be disaggregated by introducing the concept of TMR [30]. This TMR assessment at every material level makes it possible to trace back origins of other materials and energy utilized in the intermediate processes. The origins associated with material production include both the mining nations and those that contribute to indirect material requirements and hidden flows to process raw materials. These characteristics of TMR assessment enable it to be applied effectively to the concept of external country dependency for every material.

It must be mentioned that the idea of accounting for all involved materials through tracing back manufacture processes has been already developed as material flow analysis (MFA). One of the major streams is economy-wide MFA (EW-MFA) to monitor the material utilization at the national level. The concept of TMR has been employed in EW-MFA to synchronically evaluate international comparison of material consumption in the course of economic development [41]. Low level of disaggregation of material category including biomass, fossil fuels, metals, and non-metals is assessed in EW-MFA on a TMR basis as a top-down approach [42,43]. Meanwhile, TMR has been scarcely utilized in MFA on a product level as a bottom-up approach.

In the raw material criticality assessment, environmental impacts have been also widely considered [44,45] on the basis of life cycle impact assessments (LCIA) [46,47]. Finnveden et al. summarize three areas of protection in LCIA, comprising human health, natural environment and natural resources [48]. These main components in LCIA are quantified in the form of ReCiPe endpoints (one of LCIA methods) [49], which are utilized as integrated indicators of environmental impacts in the raw material criticality assessment by evaluating the inevitable consequences of environmental system [8]. Particularly, given that mining sites are vitally related to external dependency in raw material criticality assessment as mentioned above, environmental impacts arising from mining activities have to be carefully taken into account as follows. First, mining activities require energy input, causing various environmental issues such as global warming resulting from carbon emissions [50]. Second, vice-generative mine waste consisting of waste rocks, tailings and slags releases suspended particulates into the environment [51] and causes chemical toxicity issues due to oxidation [52]. Third, mining activities change the land and alter the ecological system through deforestation, pathogen introduction and biodiversity mitigation [53]. These environmental consequences from mining activities overlap with several components of ReCiPe midpoints; climate change, water use, fossil resource scarcity, fine particular matter formation, photochemical oxidation formation, human toxicity and land use.

In contrast to environmental indicators evaluating the consequences of a system, the potential magnitude of the cause has been scarcely taken into account. The only reported indicator in criticality assessment for evaluating the magnitude of the cause is carbon emissions [54]. In addition, the particular elementary flows newly described in ecoinvent are not fully evaluated from the viewpoint of mining activities in current LCIA methods [55]. Given that the degree of the aforementioned environmental impacts arising from mining activities are determined by the magnitude of ore grades and depth [56,57], the concept of TMR can be employed as a new indicator for evaluating environmental impacts arising from mining activities.

As mentioned above, the production and import quantities have been conventionally employed as the main index for the discussion of external dependency. However, the conventional approach confines its application to raw material, which potentially risks reaching a short-sighted conclusion in the policy making process. In contrast, the proposed comprehensive approach based on the concept of TMR can be applied to every material and product. In addition, it is expected that the utilization of this new approach would assist in revealing the hidden elements associated with external dependency

in the conventional approach. Given that the security of material supply is a driving force for material securement policy, this approach may be of use to policymakers in designing well-grounded policy narratives. Furthermore, it hopefully provides an informative evaluation of material criticality and environmental impacts arising from individual mining activities. As such, the objective of this paper is to establish a methodology for evaluating external dependency applied to every material and product based on TMR, beyond the conventional approach applied to only raw materials.

This paper is structured as follows: Section 2 establishes the methodology for evaluating country concentration. Subsequently, a case study is conducted based on the developed methodology in Section 3. Finally, Section 4 concludes this paper.

2. Materials and Methods

The development of a framework for the analysis of external dependency applied to every material and product based on TMR is presented in this section.

It must be noted that there are three factors which are required in this analysis: The material and product quantity (kg, L, m³), the material and product quantity based on the TMR (kg-TMR), and the specific TMR for material and product (kg-TMR/kg, kg-TMR/L, kg-TMR/m³). These factors are referred to as quantity, quantity on a TMR basis, and specific TMR, respectively, in this paper. In order to calculate the external dependency based on the concept of TMR, both quantity on a TMR basis and specific TMR for assessed materials and products have to be calculated. The relationship between these three factors is expressed in the following equation.

$$(\text{quantity on a TMR basis}) = (\text{specific TMR}) \times (\text{quantity}) \quad (1)$$

To begin with, the basic process flow for a product with the inclusion of material inputs is generalized. This flow is given in Figure 1. Input materials are required to process a product and are described in the form of I_j , where j represents the number of input materials for the process ($j = 1, 2, \dots, J$). Through the process, several products are obtained. These output products are described in the form of M_k , where k represents the number of output products for the process ($k = 1, 2, \dots, K$).

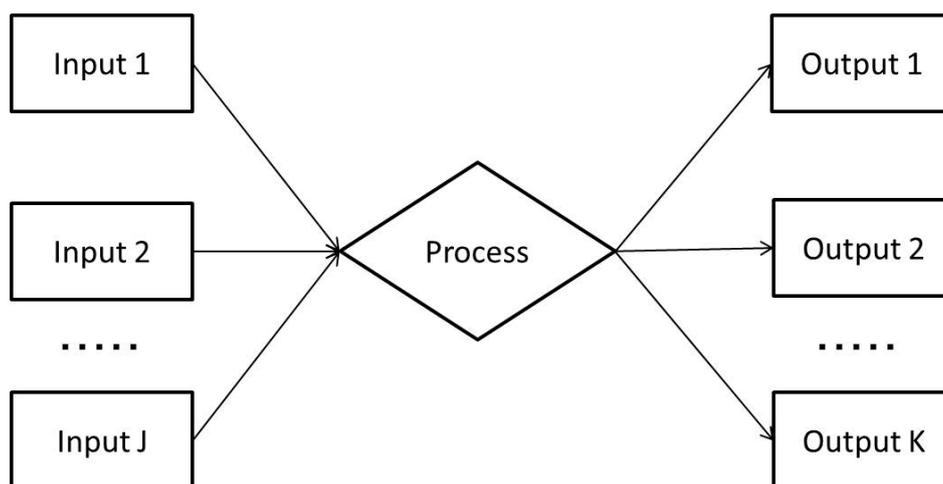


Figure 1. Basic flow of product processing.

Based on the construction of the generic process flow, the calculation of every product's TMR will be initiated. In the case where the specific TMR of the assessed-specified product has already been reported, the quantity on a TMR basis can easily be calculated with Equation (1). On the other hand, in the case where the TMR of the assessed-specified product has not yet been reported, the quantity

on a TMR basis has to be directly computed. The calculation method for quantity on a TMR basis is illustrated below. Firstly, the total quantity of both input materials and output products is defined as I and M , expressed in the following equation.

$$I = \sum_{j=1} I_j \quad (2)$$

$$M = \sum_{k=1} M_k \quad (3)$$

Subsequently, the specific TMR of each input material (I_j) is described in the form of S_j , and the total quantity on a TMR basis of input materials (T) is computed in the following equation.

$$T = \sum_{j=1} (S_j \times I_j) \quad (4)$$

In the case where only one product is generated through the process, the specific TMR of this product can be obtained by dividing the computed T by the quantity of this product. In the case of multiple products generated through the process, an appropriate allocation of computed total quantity on a TMR basis to each product obtained should be applied to determine the specific TMR. Proposing various allocation methods in life cycle assessment [58], this study applies economic allocation, considering the respective monetary values of products since the material value would be a major incentive for mining activities. The unit price of each product is described in the form of P_k and the total price of obtained products is computed using the following equation.

$$\text{Total Price of obtained products} = \sum_{k=1} (M_k \times P_k) \quad (5)$$

The allocation rate for each product is described in the form of r_k , which is determined based on the total price of obtained products as follows.

$$r_k = \frac{M_k \times P_k}{\sum_{k=1} (M_k \times P_k)} \quad (6)$$

It must be noted that the quantity to be allocated is computed by subtracting the total quantity of products from the total quantity on a TMR basis of input materials. The quantity to be allocated to product k is expressed in the form of T'_k , which is obtained in the following equation.

$$T'_k = (T - M) \times r_k \quad (7)$$

$$= \left(\sum_{j=1} (S_j \times I_j) - \sum_{k=1} M_k \right) \times \frac{M_k \times P_k}{\sum_{k=1} (M_k \times P_k)} \quad (8)$$

Finally, the quantity on a TMR basis of the product k described as T_k is computed in the following equation.

$$T_k = T'_k + M_k \quad (9)$$

$$= \left(\sum_{j=1} (S_j \times I_j) - \sum_{k=1} M_k \right) \times \frac{M_k \times P_k}{\sum_{k=1} (M_k \times P_k)} + M_k \quad (10)$$

After obtaining the quantity on a TMR basis of each product, the analysis of external dependency is then addressed.

The required data are the quantity of product and material produced by country of origin. Based on the data collection, the quantity on a TMR basis of product by country c described as $T_{k,c}$ can be computed using the following equation.

$$T_{k,c} = \left(\sum_{j=1} (S_j \times I_{j,c}) - \sum_{k=1} M_{k,c} \right) \times \frac{M_{k,c} \times P_k}{\sum_{k=1} (M_{k,c} \times P_k)} + M_{k,c} \quad (11)$$

The quantity on a TMR basis by associated countries ($T_{k,c}$) corresponds to the breakdown of quantity on a TMR basis of the product (T_k), expressed in the following equation.

$$T_k = \sum_{c=1} T_{k,c} \quad (12)$$

The explanation of both subscripts and abbreviations is summarized in Table 1.

Table 1. Summary of subscripts and abbreviations.

Content	Subscript/Abbreviation	Unit
Number of input materials	j	-
Number of output products	k	-
Producing country	c	-
Quantity of input material	I	kg, L, m ³
Quantity of output product	M	kg, L, m ³
Quantity on a TMR basis	T	kg-TMR
Specific TMR	S	kg-TMR/kg, kg-TMR/L, kg-TMR/m ³
Unit price of product	P	\$/kg
Allocation rate	r	-

3. Results and Discussion

This research paper focuses on sulfuric acid manufactured in Japan as a case study analyzing external dependency based on the developed methodology.

Japan is well-known as a resource-poor country, with critical dependency on resource imports [59]. The strategy of supply risk mitigation due to this high external dependency for materials has been seen as a critical issue in Japan [60]. As such, Japan is considered to be an appropriate country for this analysis.

In addition, there are several available techniques for producing sulfuric acid such as sulfur combustion, metal sulfide roasting and smelting, pyrite roasting, sulfuric acid regeneration, metal sulfate roasting, as well as combustion of H₂S or other sulfur-containing gases [61]. In particular, the utilization of by-product SO₂ gas from the smelting process of non-ferrous metals such as copper, hereafter referred to MSRS, contributes to approximately 80% of sulfuric acid production in Japan, while the rest of sulfuric acid production is covered by the utilization of sulfur by-product from the desulfurization process of crude oil, hereafter referred to SC [62]. On the other hand, MSRS and SC contribute approximately to 30% and 60%, respectively, of global sulfuric acid production [63], which presents the opposite trend in Japan. In addition, although sulfuric acid can be also produced from natural gas through hydrogen sulfide, Japan cannot rely on this process since only liquefied natural gas is imported. Although sulfuric acid is not apparently related to ores, there is a significant interaction with smelting non-ferrous metals that have been analyzed from the viewpoint of criticality. As such, sulfuric acid production in Japan is considerably unique and is associated with mineral criticality.

It should be noted that the case study excludes secondary resources (recycling). In addition, the analyzed year is 2007.

3.1. Electricity Generation

Before analyzing the manufacturing process of sulfuric acid, the specific TMR of electricity has to be individually computed. This is because electricity is a major component of determining the external dependency of sulfuric acid, utilized for direct input in the process of sulfuric acid production as well as for the operation of machinery.

Electricity was generated from many sources in 2007 in Japan, comprising oil, coal, natural gas, hydro, and nuclear. As its share was merely 1%, the electricity generated by non-hydropower renewables is ignored for simplicity in this study.

The data for electricity generation by source were obtained from The Federation of Electric Power Company in Japan [64]. The data on imported oil, natural gas, and coal by country were obtained from UN Comtrade [65]. In addition, the data on the specific TMR of electricity generation by oil, natural gas, coal, and nuclear were obtained from NIMS-EMS [66]. Following this report, it is assumed that the TMR of electricity generation by hydro is zero, and the operation of pumped storage hydro is ignored for simplicity. The data are shown in Table 2. The specific TMR for each of energy sources is first multiplied by the corresponding share in the energy mix and then summed (called additive aggregation method [67]) to derive the specific TMR of electricity (1.89 kg-TMR/kWh).

Table 2. TMR of electricity generation by energy source.

Energy Source	Share in Energy Mix (%)	Specific TMR (kg-TMR/kWh)
Oil	13	1.738
Natural gas	28	0.310
Coal	25	4.761
Nuclear	26	0.454
Hydro	8	0

Based on the electricity generation per year by source, its quantity on a TMR basis by source can be calculated. The share of electricity generation source in both quantity and quantity on a TMR basis is shown in Figure 2. The development of a well-diversified electricity grid mix has been considered in Japan, especially since the oil shock in the 1970s. In an attempt to be less reliant on foreign imports of oil and to reduce GHG emissions, nuclear power was developed significantly. The increase in the share of nuclear power generation delivered an improvement of diversification in the electricity mix. Meanwhile, diversification on a TMR basis is critically jeopardized compared to the case of the share in electricity generation. The process of generating electricity by using coal requires much greater material inputs, which contributes to the low diversification on a TMR basis.

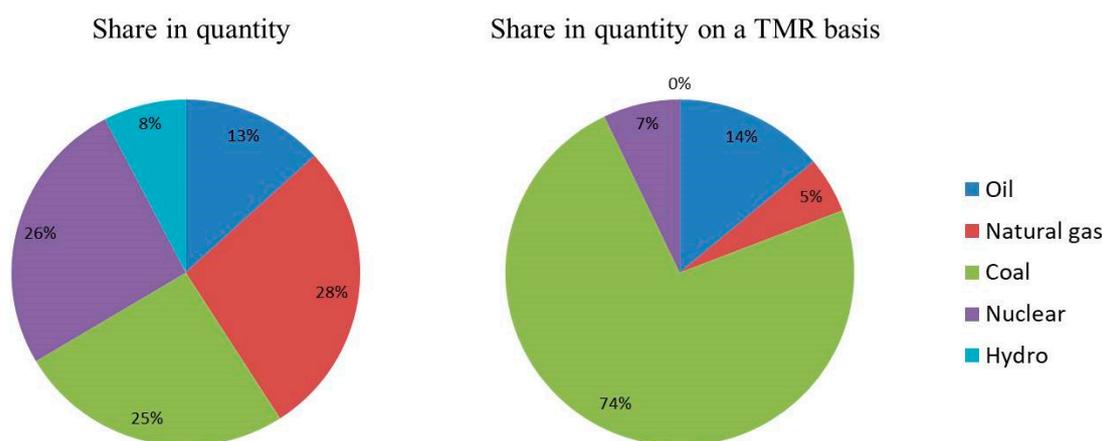


Figure 2. Share of electricity generation by source in both quantity and quantity on a TMR basis.

Subsequently, the share by country associated with electricity generation in Japan in both electricity generation and electricity on a TMR basis is computed. The results are shown in Table 3. Even though Australia covers one quarter of Japan’s external dependency, there are 18 countries each associated with at least 1% of electricity generation in Japan. On the other hand, only 12 countries contribute to more than 1% of electricity generation on a TMR basis, and Australia covers approximately 50% of Japan’s external dependency. Diversification is therefore more critical on a TMR basis.

Table 3. Share by country associated with electricity generation in Japan in both quantity and quantity on a TMR basis.

Electricity Generation			Quantity on a TMR Basis		
Ranking	Country	Share (%)	Ranking	Country	Share (%)
1	Australia	25.9	1	Australia	46.9
2	Indonesia	9.36	2	Indonesia	14.1
3	Canada	8.58	3	China	6.88
4	Japan	7.68	4	Canada	6.10
5	United Arab Emirates	5.91	5	Russian Federation	4.96
6	Saudi Arabia	5.05	6	Saudi Arabia	4.01
7	Qatar	4.71	7	United Arab Emirates	3.72
8	Malaysia	4.48	8	Qatar	2.00
9	Namibia	3.94	9	Iran, Islamic Republic	1.53
10	Niger	3.93	10	Kuwait	1.18

In addition, corresponding to the dominant share of coal on a TMR base, several countries from where Japan imports coal cannot be simply ignored in terms of external dependency. Both China and the Russian Federation are not ranked in the top 10 countries associated with electricity generation in Japan by quantity. In contrast, China is ranked 3rd, while the Russian Federation is ranked 5th on a TMR basis. It must be noted that imports from both China and the Russian Federation might be vulnerable. There are two attributes particularly associated with security of supply in the Worldwide Governance Indicators [68]: “Political Stability and Absence of Violence” and “Regulatory Quality” [69]. According to these two indicators, the risk of supply disruption in both China and Russian Federation may be considered higher than the worldwide average. The Japanese government has to consider the relationship with the countries listed in external dependency for both quantity and TMR in order to strengthen electricity supply security.

Several countries which have not been identified in the conventional approach are revealed by taking into account the total material requirement for electricity generation. Based on the computation of electricity generation on a TMR basis, the manufacturing process of sulfuric acid will be analyzed in the following section.

3.2. Manufacturing Processes of Sulfuric Acid Production in Japan

Metal sulfide roasting and smelting and sulfur combustion are utilized as a method of manufacturing sulfuric acid in Japan. In this section, sulfuric acid produced from both approaches is assessed. The data on quantities for every component were obtained from the Japan Environmental Management Association for Industry (JEMAI) [70].

3.2.1. Metal Sulfide Roasting and Smelting (MSRS)

In MSRS, sulfuric acid is obtained as a by-product when crude copper is processed from ore. Copper ore is considered the main metal sulfide for sulfuric acid production. Considering the process of obtaining crude copper as a targeted material, the flow of sulfuric acid production by MSRS is presented in Figure 3.

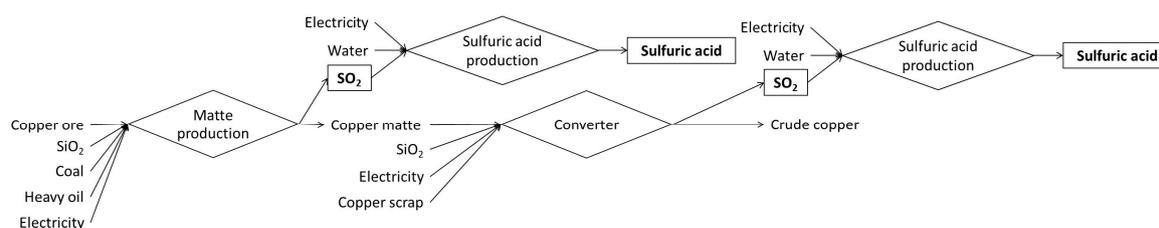


Figure 3. Flow of sulfuric acid production by MSRS.

Copper matte is obtained from processing mined copper ores with a number of other input streams, such as SiO₂, coal, heavy oil, and electricity used as input in the matte production process. The obtained copper matte is then converted into crude copper with SiO₂, coal, oxide, electricity, and copper scrap as secondary sources. SO₂ is generated as a by-product from both matte production and conversion to be utilized for sulfuric acid production.

The data of specific TMR for copper ore [71], coal and heavy oil [66], and SiO₂ [66] are available. The TMR of electricity is computed in Section 3.1. However, the specific TMR of SO₂, copper matte, and crude copper has not been reported previously. Both quantity on a TMR basis and specific TMR for copper matte and crude copper are calculated considering allocation based on a monetary unit. The prices of SO₂, copper matte, and crude copper have been assumed to be 200 \$/t, 3600 \$/t and 7200 \$/t, respectively, based on JEMAI [70].

Utilizing these, the specific TMR of sulfuric acid arising from the process of both matte production and conversion can be computed. The overall specific TMR of sulfuric acid based on MSRS is obtained by the following equation.

$$S_{sulfuric\ acid_{MSRS}} = \frac{\sum_{k=1}^2 T_{sulfuric\ acid_{MSRS}, k}}{\sum_{k=1}^2 M_{sulfuric\ acid_{MSRS}, k}} \quad (13)$$

where k is the MSRS process ($k = 1$: matte production; $k = 2$: conversion).

The results are shown in Table 4.

Table 4. Quantity on a TMR basis, and specific TMR for sulfuric acid based on both matte production and conversion, and TMR for overall sulfuric acid under MSRS.

MSRS Process	Quantity (kg)	Quantity on a TMR Basis (kg-TMR)	Specific TMR (kg-TMR/kg)
Sulfuric acid based on matte production	1.66	17.1	10.3
Sulfuric acid based on conversion	1.00	7.03	7.01
Sulfuric acid by the MSRS	-	-	9.05

The data for the country share associated with every input material and product are required for the assessment. SiO₂, other oxides and water used as inputs are entirely obtained in Japan, while many foreign countries are involved in the production of copper ore, coal, heavy oil, and electricity [70]. The data on imported copper ore, coal, and heavy oil by country were obtained from UN Comtrade [65]. The quantity on a TMR basis for electricity by country is obtained in Section 3.1. By integrating the country share of every component, the share by country associated with sulfuric acid by the MSRS in Japan on a TMR basis is computed.

3.2.2. Sulfur Combustion (SC)

The method of sulfur combustion, hereafter referred to SC, uses the sulfur from the oil refining process. Sulfur is generated through the process of oil refining from heavy oil with electricity and steam inputs. Crude oil is also required for oil import transportation. Other than sulfur, naphtha, liquefied petroleum gas (LPG), gasoline, kerosene, diesel, and heavy oil are products. Subsequently, the combustion of obtained sulfur is carried out to produce sulfuric acid in a secondary process. The flow of producing sulfuric acid through the SC is shown in Figure 4.

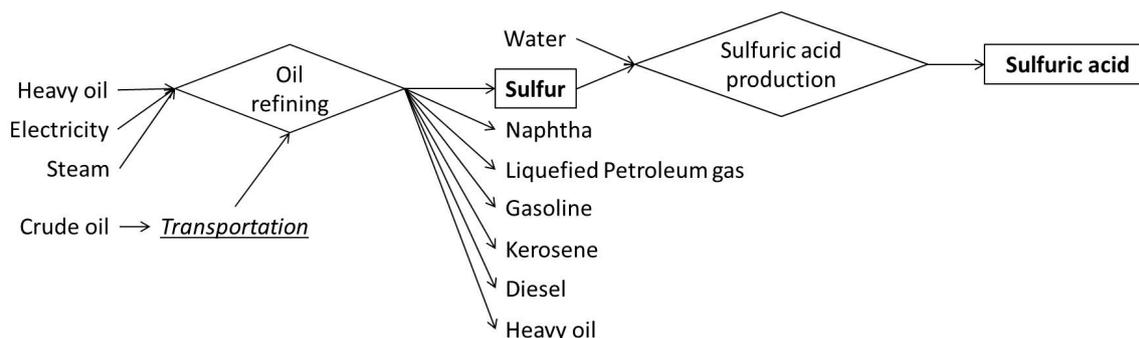


Figure 4. The flow of sulfuric acid production by the SC.

The specific TMR data of heavy oil and crude oil were obtained from NIMS-EMS Material Data [66]. The specific TMR data of steam were obtained from the Wuppertal Institute [72]. The specific TMR of electricity is computed in Section 3.1. In addition, the average distance for oil import transportation is assumed to be 10,800 km [73]. The data on required fuel for transportation were obtained from the Wuppertal Institute [72]. Both quantity on a TMR basis and specific TMR for the obtained sulfur are calculated based on the price of sulfur, naphtha, LPG, gasoline, kerosene, diesel, and heavy oil [70]. The calculation of specific TMR of sulfuric acid is then undertaken. The specific TMR of water is assumed to be 1 kg/kg. Here, 0.328 kg of obtained sulfur and 0.672 kg of water is required to produce 1 kg of sulfuric acid. As such, both quantity on a TMR basis and specific TMR for sulfur and sulfuric acid can be obtained. The results are given in Table 5.

Table 5. Quantity on a TMR basis, and TMR for sulfur and sulfuric acid under SC.

SC Process	Quantity (kg)	Quantity on A TMR Basis (kg-TMR)	Specific TMR (kg-TMR/kg)
Sulfur by SC	0.195	0.406	2.08
Sulfuric acid by SC	1.00	1.36	1.36

Pertaining to external dependency, steam and water are completely domestic, while many foreign countries are involved in the production of heavy oil, crude oil, electricity, sulfur and sulfuric oxide. The data on imported copper ore, coal, and heavy oil by country were obtained from UN Comtrade [65]. Quantity on a TMR basis of electricity by country is calculated in Section 3.1. Based on the country share data, quantity on a TMR basis by country can be obtained for both sulfur and sulfuric oxide.

3.2.3. Overall Sulfuric Acid

TMR of sulfuric acid based on both MSRS and SC was computed. TMR of overall sulfuric acid can be obtained by integrating these main two flows. As mentioned above, the share of MSRS and SC in Japan is 80% and 20% respectively [62]. The specific TMR of overall sulfuric acid can be calculated via the following equation.

$$S_{sulfuric\ acid} = S_{sulfuric\ acid_{MSRS}} \times 0.8 + S_{sulfuric\ acid_{SC}} \times 0.2 \tag{14}$$

Based on this evaluation, the specific TMR of overall sulfuric acid in Japan is 7.56 kg-TMR/kg. The country share on a TMR basis can also be obtained. The results are shown in Figure 5. Chile contributes to 38% of external dependency in Japan, followed by Indonesia (13%), Peru (13%), Australia (9%), and Canada (7%). Due to the high share of MSRS, it can be observed that the result is highly linked to the crude copper share by country in Japan. The established approach can reveal the external dependency for any material and product which has been previously difficult to quantify based on the conventional approach.

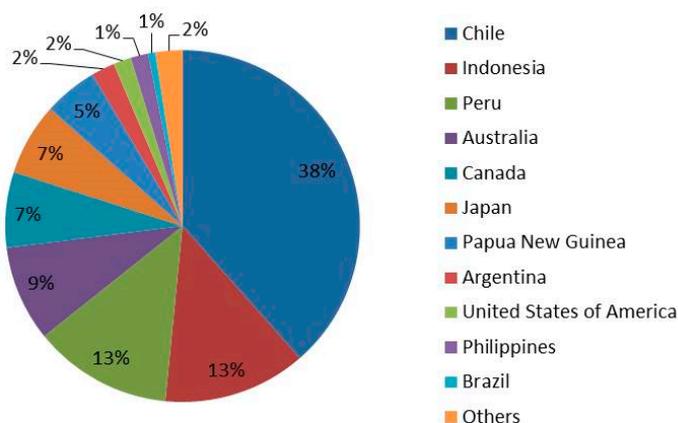


Figure 5. The share by country associated with sulfuric acid in Japan in quantity on a TMR basis.

3.2.4. Sensitivity Analysis

As presented in Section 3.2.4, the specific TMR of overall sulfuric acid was computed based on the established methodology by employing economic allocation. It must be noted that the monetary value potentially fluctuated depending on the shift of the material landscape. In order to identify the influence of the uncertainties of monetary value on the determination of the specific TMR of overall sulfuric acid, this section conducts a sensitivity analysis. Each monetary value for copper matte, crude copper, SO₂, naphtha, LPG, gasoline, kerosene, diesel, heavy oil, sulfur is changed by between −30% and 30% to observe the transition of respectively computed specific TMR of overall sulfuric acid production. For the proportion of MSRS to SC, in addition to the trend in Japan, the world trend is also assessed.

The result of the sensitivity analysis is given in Figure 6. In Japan, the SC-related elements including naphtha, LPG, gasoline, kerosene, diesel, heavy oil, and sulfur hardly affect the specific TMR of overall sulfuric acid due to less weight of its share and less value of specific TMR on a SC basis. On the other hand, the MSRS-related elements including copper matte, crude copper and SO₂ are more sensitive. Particularly, the increasing monetary value of SO₂ causes the increase in the specific TMR of overall sulfuric acid, while for copper matte and crude copper, it shows the opposite tendency. In the case of the world where the share of SC is much greater than MSRS, the sensitivity trend overlaps with the case in Japan. It is discovered that the uncertainties of monetary value for SC-related elements scarcely affect the specific TMR of overall sulfuric acid regardless of the share between MSRS and SC.

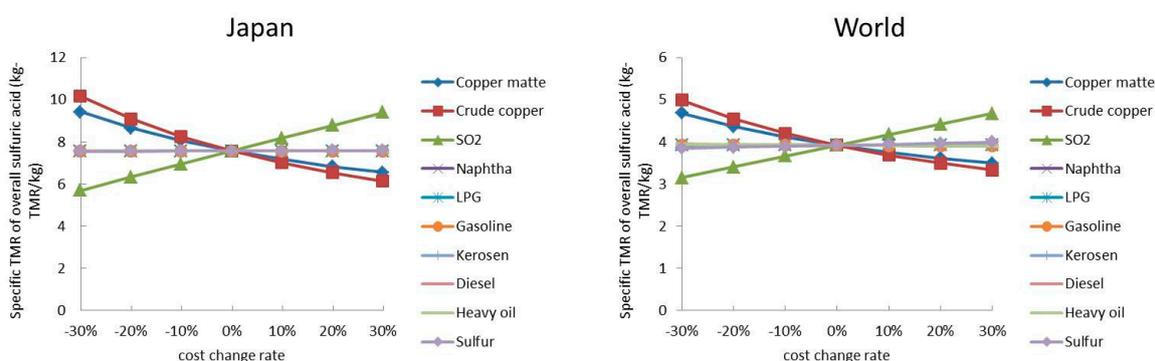


Figure 6. Sensitivity analysis of specific TMR of overall sulfuric acid.

3.3. Discussion

The external dependency of sulfuric acid in Japan in 2007 was analyzed as a case study. Through the case study, it is observed that the established approach can be applied to various materials and

products. In addition, the hidden elements associated with external dependency in the conventional approach can be revealed by this comprehensive approach based on the concept of TMR. In this section, the specific TMR and external dependency of sulfuric acid in Japan are diachronically monitored 2001–2014 corresponding to the transit of compositions. Based on the diachronic analysis, the parametric analysis of both environmental impacts and external dependency associated with sulfuric acid production in Japan on a TMR basis is conducted under various conditions of both MSRS and SC.

The sulfuric acid production share in Japan has been changed as presented in Figure 7 based on [74]. While the share of MSRS was 68% in 2001, it has constantly increased to reach 82% in 2014. Given that there are many non-ferrous smelting factories including copper, zinc and lead in Japan, the share of MSRS is greater than that of SC. Particularly, the copper production in Japan has increased in the last decade. The promotion of solar photovoltaic systems corresponding to the strategy of feed in electric tariff has increased the demand of electrical wire containing a large amount of copper for its generation equipment. In addition, the pervasive hybrid cars and miniaturization of electric appliances contribute to the demand of copper alloy products.

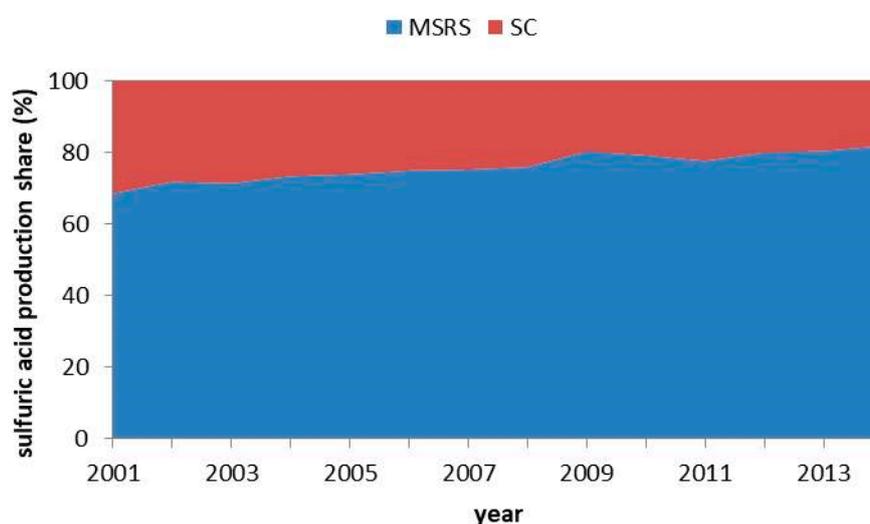


Figure 7. Sulfuric acid production share in Japan in 2001–2014.

Due to the transition of sulfuric acid production share in 2001–2014, the specific TMR of sulfuric acid in Japan has changed. The result is presented in Figure 8. The specific TMR of sulfuric acid increased from 6.63 kg-TMR/kg in 2001 to 7.65 kg-TMR/kg in 2014, since the specific TMR from MSRS is considerably greater than that from SC every year. Given that the specific TMR indicates the magnitude of mining activities as mentioned in Section 1, the environmental impacts arising from mining activities associated with sulfuric acid production in Japan have increased in recent decades.

The share by country associated with sulfuric acid in Japan on a TMR basis in 2001–2014 is also obtained, given in Figure 9. A significant shift of the country share of sulfuric acid production in Japan is not observed in recent decades. Since MSRS is dominant in Japan, the origin of copper supply is primarily listed. The country share on a TMR basis not only indicates the external dependency of sulfuric acid production in Japan but delivers the environmental and social implications. Particularly, TMR can be specifically utilized for the evaluation of localized environmental impact in comparison with the non-localized impact such as the concentration of carbon dioxide. This is because the spatial sphere of environmental impacts from mining is mostly local [75]. Furthermore, these localized environmental impacts result in the conflict of land use by the local community [76]. The greater magnitude of mining activities increasingly induces social issues, including aesthetic impacts and psychological resistance [53]. As such, the analysis of external dependency on the TMR basis could

quantitatively reveal environmental and social responsibilities which the manufacturing nation has to take for associated countries.

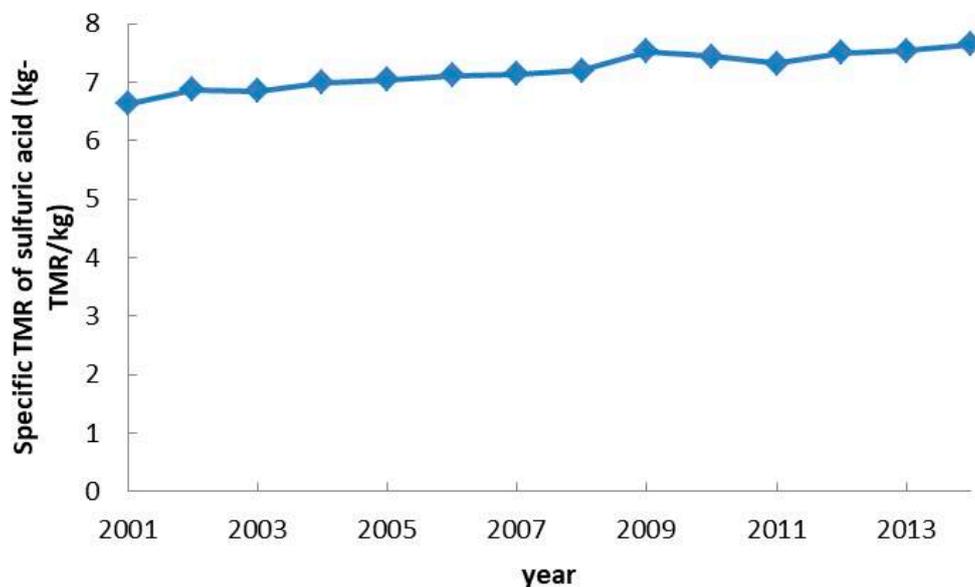


Figure 8. Specific TMR of sulfuric acid in Japan in 2001–2014.

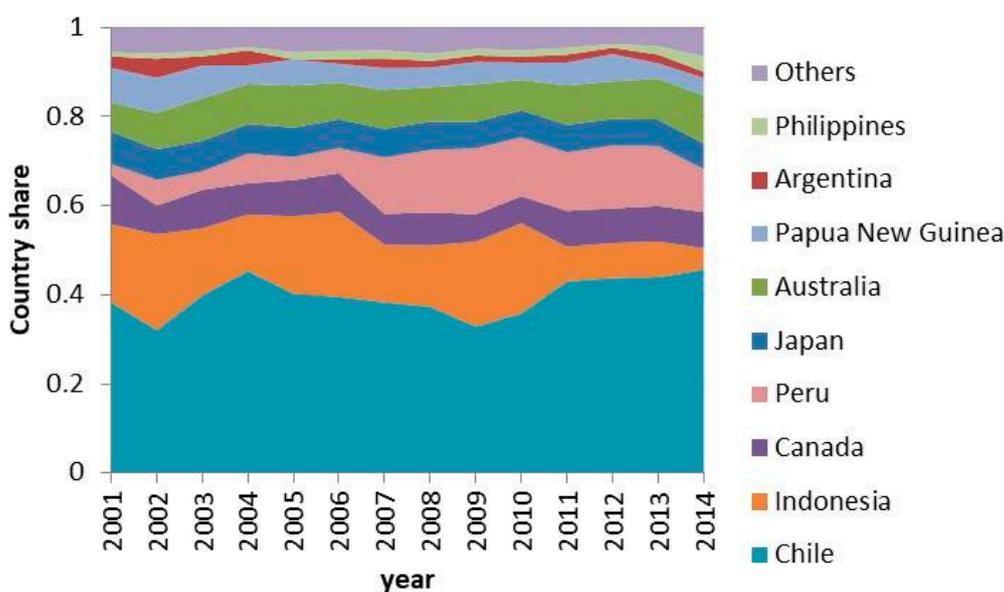


Figure 9. The share by country associated with sulfuric acid in Japan on a TMR basis in 2001–2014.

Subsequently, based on Figure 9, the share of Japan on a TMR basis is focused on to express self-sufficiency of sulfuric acid production in Japan, since self-sufficiency is a major indicator for evaluating external dependency. The result is presented in Figure 10. Given that self-sufficiency is determined by the calculation of specific TMR, it is correlated with the trend of specific TMR of sulfuric acid given in Figure 8. Self-sufficiency of sulfuric acid production in Japan on a TMR basis constantly decreased from 7.26% in 2001 to 5.76% in 2014. This trend indicates that the increasingly use of MSRS decreases self-sufficiency on a TMR basis. Furthermore, it can be said that the self-sufficiency of sulfuric production in Japan on a TMR basis is considerably low. It has been widely considered that there is little vulnerability of external dependency on sulfuric acid supply on a mere quantity basis. This is because domestic sulfuric acid production is much greater than the amount of import, and self-sufficiency on

a quantity basis is nearly 100%. In fact, 6,980,000 tons of sulfuric acid were domestically produced in Japan, while only 413 tons were imported from foreign countries such as Taiwan in 2010 [77]. The conducted case study provides the possibility of vulnerable self-sufficiency on a TMR basis of a certain product which is superficially secured from the perspective of external dependency.

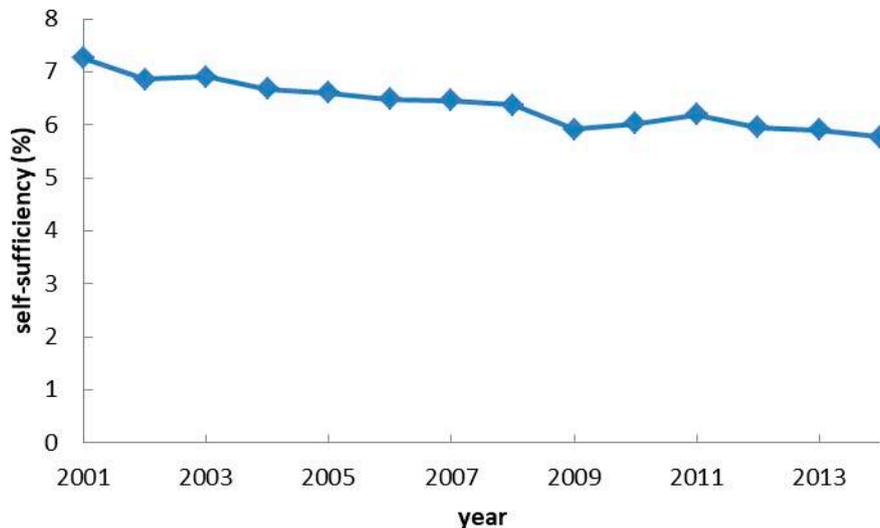


Figure 10. Self-sufficiency of sulfuric acid on a TMR basis in Japan in 2001–2014.

Finally, the parametric analysis of both environmental impacts and external dependency associated with sulfuric acid production in Japan on a TMR basis is conducted under various irconditions of both MSRS and SC. As mentioned above, the specific TMR of sulfuric acid represents environmental impacts, while external dependency is evaluated from self-sufficiency and political risk diversification aspects. In addition to self-sufficiency, political risk diversification can be also employed for the measurement of external dependency. The estimation of political risk diversification is focused on the risks of interruption of components used for sulfuric acid production. A geopolitical factor as a potential risk of disruption is assigned to each foreign supply origin. The Herfindahl-Hirschman Index (HHI) is widely accepted as an index for quantifying diversification of external dependency [8,18]. The Worldwide Governance Indicators target six dimensions of governance, and two dimensions—“Political Stability and Absence of Violence” and “Regulatory Quality”—are highly associated with security of supply [69]. The HHI is calculated by squaring the share of the product by country and summing the resulting number. A higher value of HHI corresponds to lower diversity. HHI is used to examine political risk diversification in this study. Here, the calculation of HHI excludes the domestic production component because it is separately evaluated as self-sufficiency. The political risk diversification is presented in the following equation.

$$\text{Political risk diversification} = \sum p_{ic} t_{ic}^2 \quad (15)$$

where p_{ic} is the political risk of foreign country c in year i , t_{ic} is the share of foreign country c in sulfuric acid production on a TMR basis in Japan in year i

The quantitative scale of external dependency is 0–1. A higher self-sufficiency and lower country concentration correspond to less external dependency, while a greater specific TMR corresponds to more environmental impacts. The rate of MSRS is changed from 0 to 1. The parametric analysis is conducted based on the data in 2007. Then, self-sufficiency, political risk diversification and specific TMR of sulfuric acid in Japan on a TMR basis are computed under the various combinations between MSRS and SC. The result is shown in Figure 11.

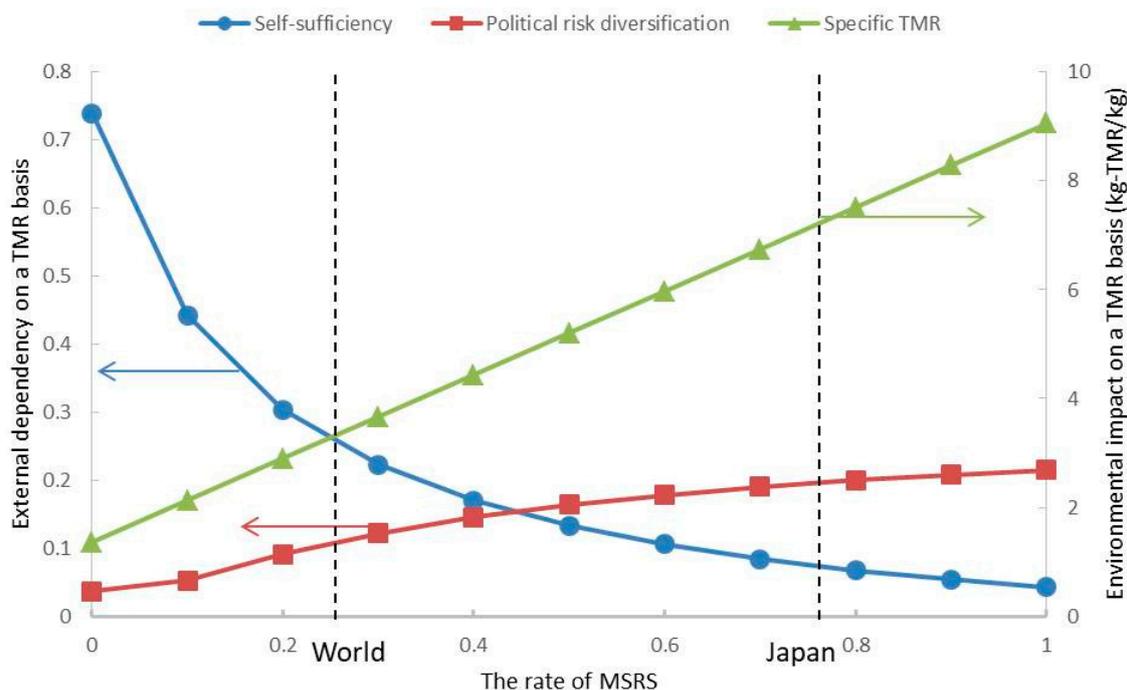


Figure 11. External dependency and environmental impacts of sulfuric acid in Japan on a TMR basis under the various combinations of both MSRS and SC.

Self-sufficiency is exponentially increased with a decreasing MSRS rate. In the case of 100% of SC, self-sufficiency reaches 73.9% on a TMR basis, while in the case of 100% of MSRS, it is only 4.2% on a TMR basis. Subsequently, by increasing the rate of MSRS, political risk diversification worsens, from 0.037 under 0% of MSRS to 0.215 under 100% of MSRS. In the research on raw material criticality, Rosenau-Tornow et al. considered a country concentration of greater than 0.15 to be critical [18]. According to their defined threshold, sulfuric acid produced by more than 50% of MSRS is categorized in the critical material. As explained in the diachronic analysis, the specific TMR of sulfuric acid is linearly increased when increasing the rate of MSRS. The specific TMR of sulfuric acid in Japan is 1.36 kg-TMR/kg under 0% of MSRS, while it is 9.05 kg-TMR/kg under 100% of MSRS.

In summary, when increasing the rate of MSRS, both environmental impacts arising from mining activities and external dependency associated with sulfuric acid production increase. As the scatter line indicates the rate of MSRS in Japan and the world, compared to the world trend, the current landscape of sulfuric acid production in Japan does not provide an environmentally benign manufacturing process and does not secure the supply of sulfuric acid from the viewpoint of external dependency.

There might be several potential policy implications in Japan for mitigating external dependency for sulfuric production. First is the encouragement of copper recycling to reduce the relative share of MS. Waste electrical and electronic equipment comprising large amount of copper have been recycled with the drying method at smelters, which requires energy and cost for delivery from the urban area to the site [78]. By introducing the wet method which enables recycling at the consumption site, a well-balanced centralized and distributed copper recycling system will be developed to mitigate the reliance on copper ore. Second is the stabilization of the monetary value for MSRS-related elements including copper matte, crude copper and SO_2 . In the last decades, the monetary value of copper has significantly fluctuated due to expansion of demand in developing countries around 2005, the financial crisis of 2007–2008, demand recovery by monetary easing around 2010, consumption decline by stopping monetary easing in the US around 2013, and constant increase in infrastructure investment in China around 2016. The possible collapse of the bubble economy in China may cause a significant drop of the copper monetary value, which potentially influences MSRS-related elements. Given the increase

in the specific TMR of overall sulfuric acid production with decreasing monetary value of both copper matte and crude copper pointed out through the sensitivity analysis, the expected financial crisis may deteriorate external dependency on sulfuric acid production.

The production of sulfuric acid in the future has to be carefully dealt with. The increase in global population requires much more food production. Higher food production accelerates the consumption of fertilizer. Given that most sulfuric acid is utilized for fertilizer, the demand of sulfuric acid is highly expected to exponentially increase. In addition, sulfuric acid is utilized instead of gravity concentration to increase the grade of non-ferrous metal including nickel. Subsequently, solvent extraction and electrowinning (SX/EW) are widely utilized for smelting lower-grade non-ferrous ores, which require a large amount of sulfuric acid [79]. Particularly in China, double the amount of sulfuric acid has been produced in the last decade. Considering the potential expansion of copper demand and the transition from fossil fuels to renewables, the global share of MSRS would potentially increase.

4. Conclusions

This paper has established a novel methodology for analysis of external dependency in terms of material criticality. This developed methodology can be utilized for external dependency analysis of any material or product. The concept of TMR has been applied as a more generic index beyond the conventional approach that uses only raw material supply. Following the established methodology, the external dependency of sulfuric acid in Japan was evaluated as a case study. In the case study, self-sufficiency and political risk diversification on a TMR basis are assessed as indicators of external dependency, while specific TMR is computed to quantify the environmental impacts arising from mining activities associated with sulfuric acid.

It should be mentioned that the hidden elements associated with external dependency in the conventional approach can be revealed by the established approach based on the concept of TMR. This approach could even be useful to identify the impacts of materials which contribute to only a small composition of a given product. Additionally, the developed algorithm to conduct the comprehensive analysis on the external dependency of every material can be readily implemented in any country confronting the issue of material securement and can be applied to material criticality assessment.

The security of material supply arising from external dependency is a driving force for material securement policy. This new approach may be of use to policymakers in designing a more sophisticated and well-grounded material securement policy.

Acknowledgments: The authors would like to thank Yamamoto from the sulfuric acid association in Japan for providing meaningful information. This study was partly supported by research funds from the Technology Foundation and the Research Institute of Science and Technology of the Society of Japan Science and Technology Agency (JST-RISTEX) as a research program in the study of Science, Technology and Innovation Policy, by KAKENHI Grants 15H02862 and 26281056, and by the Environment Research and Technology Development Fund (S-16, 3K163001) of the Ministry of the Environment, Japan.

Author Contributions: S.K., S.H., K.M., B.M., E.Y. conceived and designed the experiments; S.K., S.H., K.M., B.M., E.Y. performed the experiments; S.K., E.Y. analyzed the data; S.K., B.M., E.Y. wrote the paper.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Organization for Economic Co-operation and Development (OECD). *Material Resources, Productivity and the Environment*; OECD: Paris, France, 2013. Available online: http://www.oecd.org/greengrowth/MATERIAL%20RESOURCES,%20PRODUCTIVITY%20AND%20THE%20ENVIRONMENT_key%20findings.pdf (accessed on 25 April 2017).
2. Lutter, S.; Lieber, M.; Giljum, S. *Global Material Flow Database*; Technical Report, Version 2015.1; Vienna University of Economics and Business: Vienna, Austria, 2015. Available online: http://www.materialflows.net/fileadmin/docs/materialflows.net/WU_MFA_Technical_report_2015.1_final.pdf (accessed on 25 April 2017).

3. Organization for Economic Co-operation and Development (OECD). *OECD Environmental Outlook to 2050*; OECD: Paris, France, 2011. Available online: <http://www.oecd.org/environment/indicators-modelling-outlooks/49082173.pdf> (accessed on 25 April 2017).
4. Roelich, K.; Dawson, D.A.; Purnell, P.; Knoeri, C.; Revell, R.; Busch, J.; Steinberger, J.K. Assessing the dynamic material criticality of infrastructure transitions: A case of low carbon electricity. *Appl. Energy* **2014**, *123*, 378–386. [[CrossRef](#)]
5. Erdmann, L.; Graedel, T.E. Criticality of non-fuel minerals: A review of major approaches and analyses. *Environ. Sci. Technol.* **2011**, *45*, 7620–7630. [[CrossRef](#)] [[PubMed](#)]
6. Glöser, S.; Espinoza, L.T.; Grandenberger, C.; Faulstich, M. Raw material criticality in the context of classic risk assessment. *Resour. Policy* **2015**, *44*, 35–46. [[CrossRef](#)]
7. Nassar, N.T.; Barr, R.; Browning, M.; Diao, Z.; Friedlander, E.; Harper, E.M.; Henly, C.; Kavlak, G.; Kwatra, S.; Jun, C.; et al. Criticality of the geological copper family. *Environ. Sci. Technol.* **2012**, *46*, 1071–1078. [[CrossRef](#)] [[PubMed](#)]
8. Graedel, T.E.; Barr, R.; Chandler, C.; Chase, T.; Choi, J.; Christoffersen, L.; Friedlander, E.; Henly, C.; Jun, C.; Nassar, N.T.; et al. Methodology of metal criticality determination. *Environ. Sci. Technol.* **2012**, *46*, 1063–1070. [[CrossRef](#)] [[PubMed](#)]
9. Graedel, T.E.; Nuss, P. Employing considerations of criticality in product design. *JOM* **2014**, *66*, 2360–2366. [[CrossRef](#)]
10. Nassar, N.T.; Du, X.; Graedel, T.E. Criticality of the rare earth elements. *J. Ind. Ecol.* **2015**, *19*, 1044–1054. [[CrossRef](#)]
11. European Commission. *Critical Raw Materials for the EU, Brussel*; European Commission: Brussel, Belgium, 2010. Available online: <http://www.euromines.org/files/what-we-do/sustainable-development-issues/2010-report-critical-raw-materials-eu.pdf> (accessed on 25 April 2017).
12. European Commission. *Tracking the Challenges in Commodity Markets and on Raw Materials, COM*; European Commission: Brussel, Belgium, 2011. Available online: http://www.insg.org/presents/Mr_Anciaux_Apr11.pdf (accessed on 25 April 2017).
13. Blengini, G.A.; Nuss, P.; Dewulf, J.; Nita, V.; Peirò, L.T.; Vidal-Legaz, B.; Latunussa, C.; Mancini, L.; Blagoeva, D.; Pennington, D.; et al. EU methodology for critical raw materials assessment: Policy needs and proposed solutions for incremental improvements. *Resour. Policy* **2017**, *53*, 12–19. [[CrossRef](#)]
14. Malinauskiene, M.; Kliopova, I.; Hugi, C.; Staniškis, J.K. Geostrategic supply risk and economic importance as drivers for implementation of industrial ecology measures in a nitrogen fertilizer production company. *J. Ind. Ecol.* **2017**. [[CrossRef](#)]
15. Hatayama, H.; Tahara, K. Criticality assessment of metals for Japan’s resource strategy. *Mater. Trans.* **2015**, *56*, 229–235. [[CrossRef](#)]
16. Goe, M.; Gaustad, G. Identifying critical materials for photovoltaics in the US: A multi-metric approach. *Appl. Energy* **2014**, *123*, 387–396. [[CrossRef](#)]
17. New Energy and Industrial Technology Development Organization (NEDO). *Trend Report of Development in Materials for Substitution of Scarce Metals*; Report No. 08007835-0 08007838-0; Shinko Research Co. Ltd.: Tokyo, Japan, 2009.
18. Rosenau-Tornow, D.; Buchholz, P.; Riemann, A.; Wagner, M. Assessing the long-term supply risks for raw materials—A combined evaluation of past and future trends. *Resour. Policy* **2009**, *34*, 161–175. [[CrossRef](#)]
19. Dewulf, J.; Blengini, G.A.; Pennington, D.; Nuss, P.; Nassar, N.T. Criticality on the international scene: Quo vadis? *Resour. Policy* **2016**, *50*, 169–176. [[CrossRef](#)]
20. Moss, R.L.; Tzimas, E.; Kara, H.; Willis, P.; Kooroshy, J. The potential risks from metals bottlenecks to the deployment of Strategic Energy Technology. *Energy Policy* **2013**, *55*, 556–564. [[CrossRef](#)]
21. Achzet, B.; Helbig, C. How to evaluate raw material supply risk—An overview. *Resource Policy* **2013**, *38*, 435–447. [[CrossRef](#)]
22. Behrens, A.; Giljum, S.; Kovanda, J.; Niza, S. The material basis of the global economy: Worldwide patterns of natural resource extraction and their implications for sustainable resource use policies. *Ecol. Econ.* **2007**, *64*, 444–453. [[CrossRef](#)]
23. Henckens, M.L.C.M.; Driessen, P.P.J.; Ryngaert, C.; Worrell, E. The set-up of international agreement on the conservation and sustainable use of geologically scarce mineral resources. *Resour. Policy* **2016**, *49*, 92–101. [[CrossRef](#)]

24. Le Billion, P. The political ecology of war: Natural resources and armed conflicts. *Political Geogr.* **2001**, *20*, 561–584. [[CrossRef](#)]
25. Bleischwitz, R.; Dittrich, M.; Pierducca, C. Coltan from Central Africa, international trade and implications for any certification. *Resour. Policy* **2012**, *37*, 19–29. [[CrossRef](#)]
26. Hollins, O. *Material Security—Ensuring Resource Availability for the UK Economy*; C-Tech Innovation: Capenhurst, UK, 2008. Available online: http://www.oakdenehollins.com/pdf/material_security.pdf (accessed on 16 May 2017).
27. U.S. National Research Council. *Minerals, Critical Minerals, and the U.S. Economy*; National Academic Sciences: Washington, DC, USA, 2008. Available online: http://trade.ec.europa.eu/doclib/docs/2008/october/tradoc_140822.pdf (accessed on 25 April 2017).
28. Thomason, J.S.; Atwell, R.J.; Bajraktari, Y.; Bell, J.B.; Barnett, D.S.; Karvonides, N.S.J.; Niles, M.F.; Schwartz, E.L. *From national Defense Stockpile (NDS) to Strategic Materials Security Program (SMSP): Evidence and Analytic Support*; IDA Paper P-4593; Institute for Defense Analyses: Alexandria, VA, USA, 2010; Volume 1.
29. Henßler, M.; Bach, V.; Berger, M.; Finkbeiner, M.; Ruhland, K. Resource efficiency assessment-comparing a plug-in hybrid with a conventional combustion engine. *Resources* **2016**, *5*, 5. [[CrossRef](#)]
30. Yamasue, E.; Minamino, R.; Tanikawa, H.; Daigo, I.; Okumura, H.; Ishihara, K.; Brunner, P.H. Quality Evaluation of Steel, Aluminum, and Road Material Recycled from End-of-Life Urban Building in Japan in Terms of Total Material Requirement. *J. Ind. Ecol.* **2013**, *17*, 555–565. [[CrossRef](#)]
31. Adriaanse, A.; Bringezu, S.; Hammond, A.; Moriguchi, Y.; Rodenburg, R.D.; Schütz, H. *Resources Flows: The Material Basis of Industrial Economies*; World Resources Institute: Washington, DC, USA, 1997.
32. Eurostat; Statistical Office of the European Communities. *Economywide Material Flow Accounts and Derived Indicators (Edition 2000). A Methodological Guide*; European Communities: Luxembourg, 2000.
33. Kalmykova, Y.; Plme, U.; Fedje, K.K.; Yu, S. Total material requirement assessment of phosphorous sources from phosphate ore and urban sinks: Sewage sludge and MSW incineration fly ash. *Int. J. Environ. Res.* **2015**, *9*, 561–566.
34. Yamasue, E.; Matsubae, K.; Nakajima, K.; Hshimoto, S.; Nagasaka, T. Using total material requirement to evaluate the potential for recyclability of phosphorous in steelmaking dephosphorization slag. *J. Ind. Ecol.* **2013**, *17*, 722–730. [[CrossRef](#)]
35. Matsubae-Yokoyama, K.; Kubo, H.; Nakajima, K.; Nagasaka, T. A material flow analysis of phosphorus in Japan: The iron and steel industry as a major phosphorous source. *J. Ind. Ecol.* **2009**, *13*, 687–705. [[CrossRef](#)]
36. Yamasue, E.; Matsubae, K.; Nakajima, K.; Daigo, I.; Ishihara, K.N. Total material requirement of scrap steel from end-of-life vehicle. *J. Iron Steel Inst. Jpn.* **2014**, *100*, 778–787. [[CrossRef](#)]
37. Nuss, P.; Gardner, K.H.; Bringezu, S. Environmental implications and costs of municipal solid waste-derived ethylene. *J. Ind. Ecol.* **2013**, *17*, 912–925. [[CrossRef](#)]
38. Carruth, M.A.; Allwood, J.M.; Moynihan, M.C. The technical potential for reducing metal requirements through lightweight product design. *Resour. Conserv. Recycl.* **2011**, *57*, 48–60. [[CrossRef](#)]
39. Saurat, M.; Bringezu, S. Platinum group metal flows of Europe, part 1: Global supply, use in industry, and shifting of environmental impacts. *J. Ind. Ecol.* **2008**, *12*, 754–767. [[CrossRef](#)]
40. Dai, J.; Chen, B. Materials flows analysis of fossil fuels in China during 2000–2007. *Procedia Environ. Sci.* **2010**, *2*, 1818–1926. [[CrossRef](#)]
41. Bringezu, S.; Schütz, H.; Steger, S.; Baudisch, J. International comparison of resource use and its relation to economic growth: The development of total material requirement, direct material inputs and hidden flows and the structure of TMR. *Ecol. Econ.* **2004**, *52*, 97–124. [[CrossRef](#)]
42. Garmendia, E.; Urkidi, L.; Arto, I.; Barcena, I.; Bermejo, R.; Hoyos, D.; Lago, R. Tracing the impacts of a northern open economy on the global environment. *Ecol. Econ.* **2016**, *126*, 169–181. [[CrossRef](#)]
43. Arto, I. Using total material requirement to reduce the global environmental burden. *J. Ind. Ecol.* **2009**, *13*, 775–790. [[CrossRef](#)]
44. Graedel, T.E.; Reck, B.K. Six years of criticality assessments: what have we learned so far? *J. Ind. Ecol.* **2015**, *20*, 692–699. [[CrossRef](#)]
45. Harper, E.M.; Kavlak, G.; Burmeister, L.; Eckelman, M.J.; Erbis, S.; Espinoza, V.S.; Nuss, P.; Graedel, T.E. Criticality of the geological zinc, tin, and lead family. *J. Ind. Ecol.* **2015**, *19*, 628–644. [[CrossRef](#)]

46. Kolotzek, C.; Helbig, C.; Thorenz, A.; Reller, A.; Tuma, A. A company-oriented model for the assessment of raw material supply risks, environmental impact and social implications. *J. Clean. Prod.* **2018**, *176*, 566–580. [[CrossRef](#)]
47. Bach, V.; Berger, M.; Henssler, M.; Kirchner, M.; Leiser, S.; Mohr, L.; Rother, E.; Ruhland, K.; Schneider, L.; Tikana, L.; et al. Integrated method to assess resource efficiency—ESSENZ. *J. Clean. Prod.* **2016**, *137*, 118–130. [[CrossRef](#)]
48. Finnveden, G.; Hauschild, M.Z.; Ekvall, T.; Guinée, J.; Heijungs, R.; Hellweg, S.; Koehler, A.; Pennington, D.; Suh, S. Recent developments in life cycle assessment. *J. Environ. Manag.* **2009**, *91*, 1–21. [[CrossRef](#)] [[PubMed](#)]
49. Huijbregts, M.A.J.; Steinmann, Z.J.N.; Elshout, P.M.F.; Stam, G.; Verones, F.; Vieira, M.; Zijp, M. ReCiPe2016: A harmonised life cycle impact assessment method at midpoint and endpoint level. *Int. J. Life Cycle Assess.* **2017**, *22*, 138–147. [[CrossRef](#)]
50. Mudd, G.M. Global trends and environmental issues in nickel mining: Sulfides versus laterites. *Ore Geol. Rev.* **2010**, *38*, 9–26. [[CrossRef](#)]
51. Prior, T.; Giurco, D.; Mudd, G.M.; Mason, L.; Behrisch, J. Resource depletion, peak minerals and the implications for sustainable resource management. *Glob. Environ. Chang.* **2012**, *22*, 577–587. [[CrossRef](#)]
52. Mudd, G.M. Gold mining in Australia: Linking historical trends and environmental and resources sustainability. *Environ. Sci. Policy* **2007**, *10*, 629–644. [[CrossRef](#)]
53. Bridge, G. Contested terrain: Mining and the environment. *Annu. Rev. Environ. Resour.* **2004**, *29*, 205–259. [[CrossRef](#)]
54. Morley, N.; Eartherley, D. *Material Security: Ensuring Resource Availability to the UK Economy*; Oakedene Hollins; C-Tech Innovation Ltd.: Chester, UK, 2008.
55. Hischer, R.; Weidema, B.; Althaus, H.; Bauer, C.; Doka, G.; Dones, R.; Frischknecht, R.; Hellweg, S.; Humbert, S.; Jungbluth, N.; et al. Implementation of Life Cycle Impact Assessment Methods. 2007. Available online: http://esu-services.ch/fileadmin/download/publicLCI/03_LCIA-Implementation.pdf (accessed on 16 October 2017).
56. Northey, S.; Mohr, S.; Mudd, G.M.; Weng, Z.; Giurco, D. Modeling future copper ore grade decline based on a detailed assessment of copper resources and mining. *Resour. Conserv. Recycl.* **2014**, *83*, 190–201. [[CrossRef](#)]
57. Norgate, T.; Haque, N. Energy and greenhouse gas impacts of mining and mineral processing operations. *J. Clean. Prod.* **2010**, *18*, 266–274. [[CrossRef](#)]
58. De Vries, M.; de Boer, I.J.M. Comparing environmental impacts for livestock products: A review of life cycle assessments. *Livest. Sci.* **2010**, *128*, 1–11. [[CrossRef](#)]
59. Hatayama, H.; Tahara, K. Evaluating the sufficiency of Japan’s mineral resource entitlements for supply risk mitigation. *Resour. Policy* **2015**, *44*, 72–80. [[CrossRef](#)]
60. Prime Minister of Japan and His Cabinet. *Resource Securement Strategy*; Prime Minister of Japan and His Cabinet: Tokyo, Japan, 2012. Available online: <http://www.kantei.go.jp/jp/singi/package/dai15/sankou01.pdf> (accessed on 27 April 2017).
61. Ashar, N.G.; Golwalkar, K.R. *A Practical Guide to the Manufacture of Sulfuric Acid, Oleums, and Sulfonating Agents*; Springer: Berlin, Germany, 2013. Available online: <http://www.springer.com/978-3-319-02041-9> (accessed on 13 May 2017).
62. Merchant Research and Consulting Ltd. *Sulfuric Acid Market in Japan: 2017–2021 Review*; Merchant Research and Consulting Ltd.: Birmingham, UK, 2017.
63. Messick, D.L. World Sulphur Outlook. 2012. Available online: http://www.firt.org/sites/default/files/DonMessick_Sulphur_Outlook.pdf (accessed on 7 September 2017).
64. The Federation of Electric Power Company in Japan. *Graphical Filp-chart of Nuclear & Energy Related Topics 2011*; The Federation of Electric Power Company in Japan: Tokyo, Japan, 2011. Available online: http://www.fepc.or.jp/library/pamphlet/zumenshu/pdf/all_english.pdf (accessed on 9 May 2017).
65. UN Comtrade. *Mineral Fuels, Oils, Distillation Products*; UN Comtrade: New York, NY, USA, 2016. Available online: <https://comtrade.un.org/> (accessed on 8 April 2017).
66. Nakajima, K.; Ijima, K.; Halada, K. *Estimation of Total Material Requirement: Energy Resources and Industrial Materials*; NIMS-EMS Material Data for the Environment, No. 10; National Institute for Materials Science: Tsukuba, Japan, 2006. Available online: <http://www.nims.go.jp/genso/0ej00700000039eq-att/0ej00700000039of.pdf> (accessed on 11 May 2017).

67. BAng, W.; Choong, W.L.; Ng, T.S. Energy security: Definitions, dimensions and indexes. *Renew. Sustain. Energy Rev.* **2015**, *42*, 1077–1093.
68. The World Bank Group. Worldwide Governance Indicators. 2017. Available online: <http://info.worldbank.org/governance/wgi/index.aspx#home> (accessed on 10 May 2017).
69. Lefevre, N. Measuring the energy security implications of fossil fuel resource concentration. *Energy Policy* **2010**, *38*, 1635–1644. [[CrossRef](#)]
70. Japan Environmental Management Association for Industry. JEMAI-LCA Pro. 2005. Available online: <http://www.jemai.or.jp/> (accessed on 12 May 2017).
71. Halada, K.; Iijima, K.; Natagiri, N.; Okura, T. An approximate estimation of total material requirement of metals. *J. Japan Inst. Met. Mater.* **2001**, *65*, 564–670. [[CrossRef](#)]
72. Wuppertal Institut for Climate, Environment and Energy. *Material Intensity of Materials, Fuels, Transport Services, Food*; Wuppertal Institut: Wuppertal, Germany, 2011.
73. Uchiyama, Y.; Yamamoto, H. *Energy Analysis on Power Generation Plants*; Economic Research Center, Rep. No. Y90015; Central Research Institute of Electric Power Industry: Tokyo, Japan, 1991.
74. The Sulfxuric Acid Association in Japan. Sulfuric Acid Demand and Supply. 2017. Available online: <http://www.ryusan-kyokai.org/> (accessed on 20 July 2017). (In Japanese)
75. Franks, D.; Brereton, D.; Moran, C.J. Managing the cumulative impacts of coal mining on regional communities and environments in Australia. *Impact Assess. Proj. Apprais.* **2010**, *28*, 299–312. [[CrossRef](#)]
76. Brereton, D.; Forbes, P. Monitoring the impact of mining on local communities: A Hunter Valley case study. In Proceedings of the Minerals Council of Australia Inaugural Sustainable Development Conference, Melbourne, Australia, 26–28 October 2004.
77. Ministry of Finance. Trade Statistics of Japan. 2017. Available online: http://www.customs.go.jp/toukei/info/index_e.htm (accessed on 11 July 2017).
78. Ogawa, M.; Kato, M.; Majima, M.; Awazu, T.; Yata, H.; Ooe, M. Copper Recycling Technique Using Electrochemical Processes. *SEI Tech. Rev.* **2017**, *190*, 84–87.
79. Frias, C.; Mejias, A.; Martin, D.; Diaz, G. Solvent Extraction Applied to Mixed Copper and Zinc Bearing Materials. 2010. Available online: <https://ddtp.tecnicasreunidas.es/wp-content/uploads/2016/11/P-Solutions-for-primary-zinc-materials-MIXED-ORES.pdf> (accessed on 21 December 2017).



© 2018 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).