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Analysis of supercritical water oxidation for detoxification of waste organic solvent in university based on life cycle assessment

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ABSTRACT

Spray incineration and supercritical water oxidation (SCWO) processes have been used for detoxifying waste organic fluids in the University of Tokyo. In this study, we aim to elucidate the environmental aspects of these waste treatment processes by life cycle assessment (LCA). Through the investigation of actual plants, the inventory data and other characteristics of actual plants were collected and analyzed. To confirm the potential of SCWO, three modification types of the process and operation were considered and assessed on the basis of estimated inventory data. The results demonstrate that spray incineration has less environmental impact than SCWO in all scenarios. However, SCWO has various advantages for installation as a treatment process in universities such as negligible risk of creating dioxins and particulate matter. Proper choice of the treatment method for organic waste fluid requires a comprehensive analysis of risks. Spray incineration poses the risk of providing dioxins and particulate matter, while SCWO has such risk at negligible level. This means that waste including concerned materials related to such emission should be treated by SCWO. Using the right technologies for the right tasks in the detoxification of hazardous materials should be implemented for sustainable universities.

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1. Introduction

The treatment of hazardous waste has become an important issue related to pollution prevention and environmental management. In addition to local chemical risk on safety, health, and environment, regional/global environmental impacts such as acidification, soil/water/air pollution, and global warming should be considered in hazardous waste treatment. Various methods of treating hazardous waste had existed such as incineration, immobilization, landfill and off-shore disposal, and underground storage [1]. Technologies have been developed [2] and applied in various countries [3,4], although legislations on hazardous waste disposal have been enacted and, for example, landfill of wastes must meet requirements [5]. Not only the installation of waste treatment, but also the avoidance of waste generation should also be taken into account, such as the minimization of generated waste quantity and the replacement of chemicals to avoid waste generation [6]. In this regard, however, the condensation of hazardous materials for volume reduction can cause higher risk rather. Among developed waste-treatment methods, incineration has been used mainly for treating waste fluid containing organic chemical compounds, partly because of its applicability to a wide range of waste types. Incineration also has economic advantage except for diluted waste. In this regard, however, incineration treatment should be managed with measures against the generation of toxic gases [7]. At the same time, the risk of generating dioxins has also been a source of concern. To manage and sustain hazardous waste treatment sites, risk communication with neighbors living within the sites must be carefully addressed with reliable information on risk [8,9].

Supercritical water has become an interesting medium for chemistry over the last two decades [10]. One of the most studied reactions is an oxidative reaction, i.e., supercritical water oxidation (SCWO), which has been regarded as a technology applicable to waste treatment [11]. Above its critical temperature of 647 K and pressure of 22.1×10^6 Pa, water becomes a nonpolar solvent and dissolves various organic compounds and gases. Supercritical water has adequate properties for decomposing organic waste. Organic compounds are rapidly oxidized by reacting with O_2 in supercritical water. One of the most important characteristics of using SCWO is its nonemission of dioxins and particulate matter. As well as economic advantage of SCWO due to its use of water, the emission of smaller amounts of toxic acid gases, e.g., thermal NO_x, makes the method suitable for adoption in waste treatment. These attributes show the high potential of SCWO for detoxifying hazardous organic substances. Several applications were discussed for applying SCWO to wastewater treatment [12]. A bench-scale

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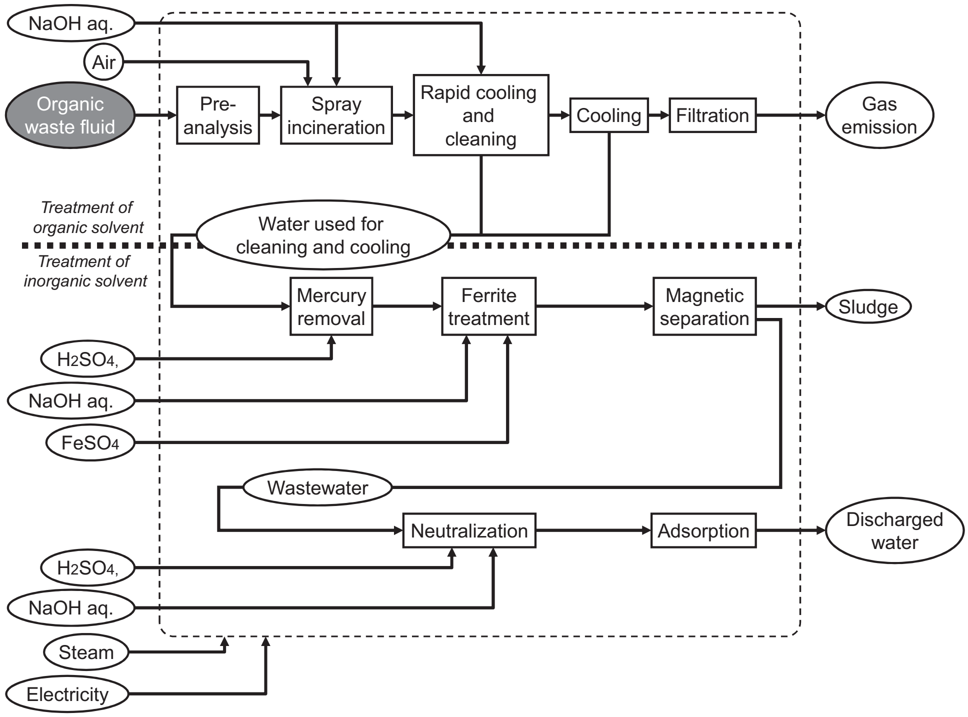

Fig. 1. Description of spray incineration process operated in Hongo campus of the University of Tokyo.

reactor for decomposing chemical agents was demonstrated [13]. Wastewater from textile production [14] and acrylonitrile manufacture [15] has been treated by SCWO. On the other hand, SCWO has some technical problems such as corrosion and salt precipitation, and operational problems related to high temperature and high pressure [16]. To solve these problems, some concepts of the SCWO reactor were studied and suggested [17–19]. The use of the transpiring wall reactor, one of the SCWO reactor concepts, was suggested as a decomposition process for treating industrial wastewater [20], and the experimental result of treating salt-contained wastewater using the reactor was reported [21], which demonstrated that the reactor can treat salt-contained waste without causing any reactor blockage.

The University of Tokyo is one of the largest sources of greenhouse gas (GHG) emission in Tokyo and therefore has launched a project for a sustainable campus [22]. Regarding general waste from workers and students, strategic approaches to its reduction and treatment using management systems have been recommended in the UK, Canada, and New Zealand [23–25]. Research activities generate various waste fluids containing toxic chemical substances. In universities, the generated waste fluid contains many sorts of solvent and reagent, which often change according to the research activity. Industrial waste fluid may be constant in terms of quality and quantity, so that a treatment process can be designed for its products and research plans. Because research plans in the university is redesigned more frequently than those in the industry, waste fluid generated in a university is unpredictable and treatment processes should be able to address their characteristics. This is why a waste treatment process applying SCWO for used reagents in the University of Tokyo was developed and started in 2003. In the University of Tokyo, the total amount of waste fluid is about 204 ton per year, and that of organic waste fluid is about 176 ton and inorganic waste fluid is about 28 ton in 2009 FY [22]. To treat and detoxify these hazardous wastes, the Environmental Science Center was established in the University of Tokyo, and two processes have been constructed and started at the Hongo and Kashiwa campuses. At the Hongo campus, organic waste fluid is treated by spray incineration. At the Kashiwa campus, organic waste fluid is processed by SCWO. Because some universities are located in urban areas, various concerns related with such detoxification processes, e.g., dioxin creation and toxic gas emission from smokestacks must be addressed and identified quantitatively or qualitatively to communicate with residents around campuses. At that time, environmental impact has become one of the concerns in constructing new processes. Although the SCWO processes of treating sewage waste treatment and transformer oil were analyzed in previous research studies [26–29], there are only a few comparative analyses of the environmental aspects of various conventional methods.

In this paper, the applicability of SCWO to organic waste fluid detoxification in universities is analyzed with a case study of the University of Tokyo. The analysis is focused on the environmental impact of SCWO compared with that of conventional spray incineration. To quantify environmental impact, life cycle assessment (LCA) [30,31] is applied, which has been increasingly utilized to assess technologies toward sustainable development [32,33]. By using LCA, existing environmental impact due to SCWO technology can be analyzed comprehensively through the life cycle of not only the solvents and reagents to be treated, but also the utilities used in treatment processes. As well as LCA results, the characteristics of SCWO are also interpreted, especially the negligible risk of creating particulate matters and dioxins. This discussion leads to the clarification of its true detoxification potential.

2. Materials and methods

2.1. Waste treatment processes operated in the University of Tokyo

In the University of Tokyo, waste fluid is segregated and collected on the basis of a campus rule, where wastes are divided into 13 categories [34]. This rule is applied in both Hongo and Kashiwa campuses. Fig. 1 shows the studied process operated in the Hongo

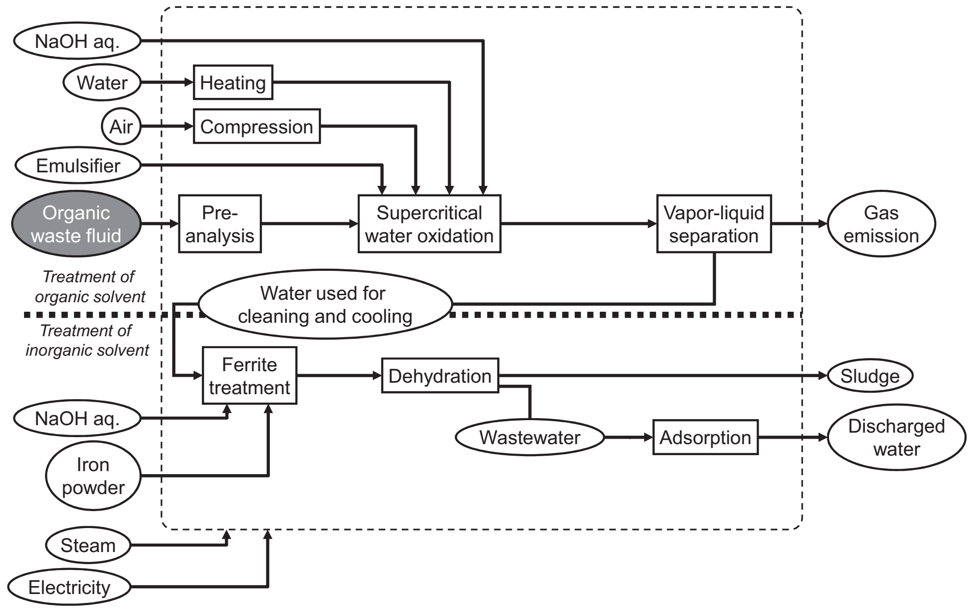

Fig. 2. Description of SCWO process operated in Kashiwa campus of the University of Tokyo.

campus of the University of Tokyo, which is mainly divided into organic and inorganic waste fluid treatments. The throughput of the incinerator is 70 L/h of waste fluid. In the preanalysis, the content and heat quantity of waste fluid is qualified, because spray incineration cannot treat halogenated chemicals. Before feeding fluid into the reactor, the heat quantity of waste fluid is adjusted by blending collected waste fluid to keep the calorie of fed fluid constant. Organic waste fluid without halogenated chemicals is sprayed into the burner reactor at 1223 K, where organic compounds are combusted or thermally decomposed. The gas emission from the spray incinerator has a high temperature and contains hazardous substances such as inorganic metals, NO_x and particulate matter. Therefore, the gas must be cooled and cleaned by an alkaline water solution, filtered to remove remaining hazardous materials, and then emitted to the air through smokestacks. The alkaline water solution used contains inorganic metals and pumped to the secondary treatment processes for inorganic substances. After removing mercury, other heavy metals are decontaminated by ferrite treatment. Ferrous sulfate is added to the wastewater containing heavy metals, which initiates a reaction forming ferrite crystals under alkali conditions at 330–345 K. After ferrite compounds are separated from water by a magnetic separator, the wastewater is purified by adsorption onto activated alumina to achieve the requirement of the regulation on sewage systems. At present, this spray incineration process at the Hongo campus is started up and shutdown in a day. The burner reactor is started up using city gas to heat up the process every morning, treats organic waste fluids, and then it is shutdown using kerosene, which can gradually turn it off and clean its inside.

The studied SCWO process operated in the Kashiwa campus of the University of Tokyo is schematically shown as a block flow diagram in Fig. 2 and its configuration is organized in Table 1. The throughput of this SCWO reactor is 15 L/h. In this process, there is a preanalysis step for adjusting the heat quantity fed to the SCWO reactor. In this system, excess oxidizer for treating wastes is fed to the reactor. Although the water contained in the fed fluid can be acceptable for SCWO reactor rather than spray incineration, the amount of incombustible waste fluids treated in SCWO reactor has not been larger than that of combustible waste fluids. This is because the ion concentration such as chloride, sulfate, or phosphate is not preferable for SCWO reactor regarding material

Table 1
Configuration of SCWO reactor at the Kashiwa campus of the University of Tokyo.

Construction characteristics	Type	MODAR type [35,36] with dual-shell pressure balanced vessel (DSPBV) [37]
	Diameter [mm]	460 (outer) 348 (inside)
	Length [mm]	1745 (outer) 1180 (inside)
	Volume [m^3]	0.107
	Material	Chrome molybdenum steel (external cylinder) Titanium-alloy (inner cylinder)
	Oxidant	Air
Operational conditions	Residence time [h]	7.1
	Flow rate of waste [L/h]	15
	Reactor temperature [K]	843–903
	Reactor pressure [MPa]	23.3–23.7

corrosion of reactor. The amount of incombustible waste fluids is less than that of combustible ones in this process. Organic waste fluid is pumped into the SCWO reactor and decomposed. At that time, a small amount of NaOH is added as neutralizer for effluent gas and liquid. As for the effluent from the SCWO process, the gas, composed mainly of CO_2, small amounts of other oxides such as SO_x and NO_x, and water are emitted. The wastewater may contain heavy metals; therefore, it is pumped to the secondary process, i.e., inorganic treatment process, where iron powder is applied to coagulate and separate heavy metals from the wastewater. After dehydration of sludge waste and removal of remaining metals by adsorption, the wastewater is discharged to the sewerage. The SCWO in the Kashiwa campus is also started up and shutdown in a day. To control the inside temperature of the SCWO reactor during the start-up and shut-down, ethanol or 2-propanol is loaded as waste to be decomposed in the reactor. These are fed to the reactor to start-up and shut-down it mildly and to clean up the pipes around reactor after treating waste fluid.

2.2. Assessment

The life cycle system defined in this study is shown in Fig. 3. The functional unit of the scenarios compared is set as the execu-

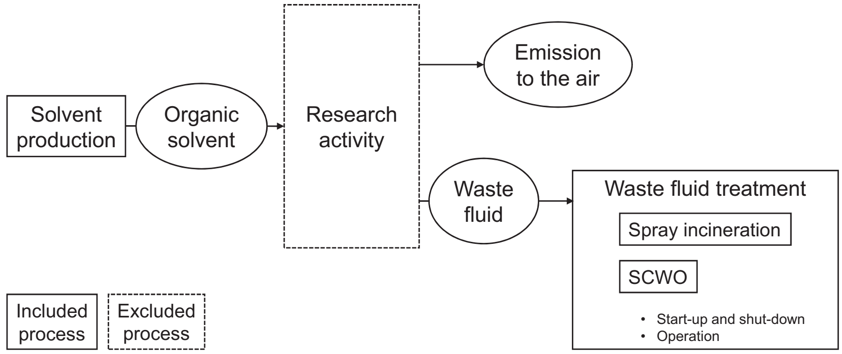

Fig. 3. Life cycle system of organic solvent.

tion of research activities using unit amount of an organic solvent. In this study, the solvent was acetone, which is one of the most utilized solvents in the University of Tokyo. The production processes of the agent and utilities such as fuels and electricity, and the processes of waste fluid treatment, i.e., spray incineration and SCWO, are included. As for the inventory of this life cycle, the direct emission to the air from research activities is taken into account. In this regard, it is assumed that the research activities are same in all the scenarios assessed in this study. In the impact assessment of LCA, the indicator was defined as disability-adjusted life years (DALY), the definition of which is that DALYs for a disease or health condition are calculated as the sum of the years of life lost (YLL) due to premature mortality in the population and the years lost due to disability (YLD) for incident cases of the health condition [38]. This indicator has been applied in various life cycle impact assessment methodologies for quantifying human health damage due to environmental impact [39]. Global warming, photochemical oxidant creation, air pollution in urban areas, and human toxicity were included in the impact assessment. The life-cycle impact assessment method based on endpoint modeling (LIME) version 2 [40,41] was applied. LIME2 is the method for quantifying environmental impacts originating from the emission from Japan [39–41]. The meteorological and geographical conditions in Japan are taken into account to define impact factors for acidification, photochemical oxidant creation, urban air pollution, and so on.

2.3. Scenario definition

On the basis of the system boundary, scenarios are defined and their settings are organized in Table 2. To confirm the environmental aspects of the detoxification process, scenario 0 is set as the research activity without waste treatment process, which means that all the solvent used is directly emitted to the environment, i.e., the air. In scenarios 1 and 2, the solvent used is collected within the two campuses and treated by spray incineration and SCWO under actual conditions. The inventory data on waste treatment processes was obtained through the investigation of actual operation results. Some of the obtained data were measured and others were estimated by the specifications of devices. All the processes were started up and shutdown in a day as actual operations, for example, the temperature or pressure of reactor must be increased from ordinary temperatures and pressures. During such unsteady-state operations, additional process chemicals were required and accumulated, for example, city gas for starting up spray incineration or ethanol for SCWO. Such utilities for unsteady-state operations were allocated to the waste fluid treated in the day's operation. The emission from waste treatment processes is monitored in each process, and the average measured data were applied, except that for CO_2, which was estimated under the assumption of the complete oxidization of waste fluid. The production inventories of process chemicals listed in Figs. 1 and 2, fuels, and other utilities were obtained and estimated from existing LCA databases [41,42] and the literature [43] except that for the emulsifier.

Although spray incineration is now an established conventional technology for detoxifying hazardous materials, SCWO is still regarded as a developing technology. This means that SCWO has not been optimized to increase its energy efficiency. Three types of possible alternative processes on operation and devices have been generated on the basis of interviews and communications with engineers regarding the SCWO process operated in the University of Tokyo. On the basis of these scenarios, the potential of the SCWO process is analyzed and discussed.

Because the pressure and temperature conditions of the SCWO process are quite severe, the energy use for the start-up and shutdown operation (of the SCWO process) is considerably large. To reduce the energy use in such an unsteady-state operation, a continuous operation of the process might be effective. As organized in

Table 2
Scenario settings.

	Scenario						
	0	1	2	3	4	5	6
Agent and amount	Acetone 1 L	Acetone 1 L	Acetone 1 L	Acetone 1 L	Acetone 1 L	Acetone 1 L	Acetone 1 L
Treatment	N/A	Spray incineration	SCWO	SCWO	SCWO	SCWO	SCWO
SCWO setting							
Start-up and shut-down operation	N/A	In a day	In a day	In a week	In a year	In a year	In a year
Oxidizer	N/A	Air	Air	Air	Liquid oxygen	Air	Liquid oxygen
Compressor for oxidizer	N/A	Normally installed	Normally installed	Normally installed	Normally installed	Two units installed	Two units installed
Power recovery	N/A	N/A	N/A	N/A	N/A	Power recovery turbine	Power recovery turbine

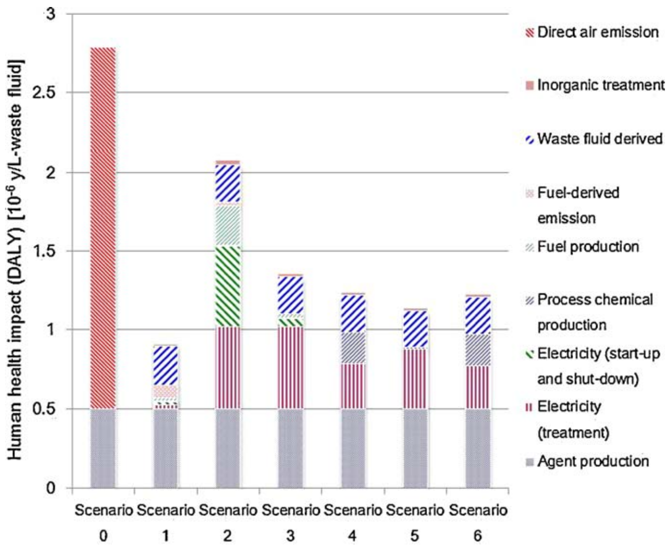

Fig. 4. The result of life cycle assessment on the basis of acetone used (1 L).

Table 2, scenario 3 takes into account the weekly continuous operation, the running time of which is from Monday at 9 am to Friday at 5 pm. The startup and shutdown operations are performed once a week, respectively, whereas ten times under the current daily operation. In scenarios 4–6, the annual continuous operation of SCWO is assumed. Note that continuous operation test of the SCWO process for 5 days has already been performed at the University of Tokyo and no problem was encountered.

Regarding electricity use during operation, the compression of the oxidizer, i.e., air, has been regarded as a factor of the significant increase in the required energy. Nearly half of the electricity usage of the Kashiwa SCWO process is used for compressing air as the oxidizer. To reduce the energy used for compression, liquid oxygen can be an alternative oxidizer, because liquids need less energy to increase their pressure. Liquid oxygen is utilized as the oxidizer for the SCWO process in scenarios 4 and 6. To estimate the inventory data utilizing liquid oxygen, some assumptions were set as follows. The amount of liquid oxygen was defined as the same amount of gaseous oxygen contained in the air. Instead of the high-power compressor for air, an additional pump was installed to feed liquid oxygen.

In addition to the adoption of liquid oxygen, the installation of a multistage compressor and a power recovery turbine can be measures to reduce the electricity usage [44] of the SCWO process. In the actual SCWO process, a compressor unit but not a turbine was installed; the effluent gas pressure is decreased through pressure control valves. In scenario 5, the doubling of the compressor and the installation of a power recovery turbine are taken into account. A device for air compression is added in Fig. 2. Regarding the installation of a power recovery system, a turbine is added before emitting gas to the air. The outlet gas from the vapor–liquid separator, 23.5 MPa at 493 K, is decompressed to 10 MPa at 417 K. To estimate the inventory change caused by installing the devices mentioned, the process simulator Aspen Plus^{TM} was applied. In the simulation, an isentropic compressor and a turbine were adopted, and the single- and multicompressor processes, were simulated. The estimated data on power use in scenarios 5 and 6 were extrapolated from simulation results and actual plant data. As for the estimation of physical property, the Soave–Redlich–Kwong (SRK) equation was employed.

3. Results

Fig. 4 shows the results of LCA for defined scenarios. In this figure, the graph legend "inorganic treatment" means environmental load generated by iron powder treatment or ferrite treatment, and processes after either treatment including additive production. "Waste fluid derived" means the emission from waste fluid by spray incineration or SCWO. "Fuel-derived emission" means the emission from incineration or oxidation of city gas, kerosene, and ethanol. "Fuel production" means the environmental load of city gas, kerosene, and ethanol production. "Process chemical production" means the environmental load of additives production used for inorganic treatment. "Electricity (start-up and shut-down) means the environmental load of electricity used to start up and shut down the organic treatment equipment. "Electricity (treatment)" means the environmental load of electricity used for the organic treatment. From the results of scenarios 0–2, the damage due to the direct emission of solvent to the air was considerably higher than that due to treating it in two processes. This is because acetone has a photochemical oxidant creation potential resulting in high human health impact, for example, respiratory diseases [45,46]. Even though other environmental loads such as CO_2, SO_x, and NO_x are increased by treating the solvent, the damage originating from the emission cannot upend the dominance of the waste treatment. Fig. 4 shows that the treatment reduces the environmental impact of organic solvents on human health. Comparing scenarios 1 and 2, it is revealed that SCWO has a larger human health impact than spray incineration.

The contribution of the start-up and shut-down operation in the SCWO process to the total damage of scenario 2 is about 38%, while that in spray incineration is about 13% of the total damage of scenario 1. This means that such an unsteady-state operation for the SCWO reactor needs much energy to increase the temperature and pressure of the equipment and water. The result of scenario 3 demonstrates that the modification of the operation schedule from the daily start-up and shut-down to a weekly continuous operation can reduce the total impact by about 35% of scenario 2. In scenario 3, the contribution of the unsteady-state operation to the total impact is about 5%. Through the evaluation of SCWO for different operation periods, the continuous start-up and shut-down operation for more than 30 days has less damage contribution to the total impact than 1%. Therefore, under the annual continuous operation in scenarios 4–6, the contribution of the start-up and shut-down operation can be considered as negligible. Under the assumption of neglecting the start-up and shut-down operation, the total impact can be reduced.

The result of scenario 4 demonstrates the effect of changing the oxidizer from air to liquid oxygen with continuous operation. By comparing scenario 4 with scenario 2, the total impact can be reduced by about 40%. Because the compression of liquids needs less energy than that of gases, the electricity use for treating waste fluid can be reduced by 45% from that in scenario 2. Despite the reduction, the reduction ratio was not so high in scenario 4, because of the large impact of the production of liquid oxygen. The result of scenario 5 demonstrates that the installation of an additional compressor and an additional turbine can more effectively reduce the total impact than the adoption of liquid oxygen. The total impact can be reduced by 45% from scenario 2. Owing its high impact in terms of the production of liquid oxygen, scenario 6 has a larger impact than scenario 5, but a smaller impact than scenario 4.

4. Discussion

Although the results of scenarios 1–6 demonstrate that SCWO has a larger environmental impact than spray incineration, it has a different function as a waste treatment process and should be

Table 3
Characteristics of treatment processes operated in the University of Tokyo.

	Spray incineration	SCWO
Emission risk		
Dioxin creation	Not negligibleOperational measures required	Negligible
Particulate matter creation	Not negligible Device measures required	Negligible
Operational risk	Fire	Explosion
Solvent		
Halogenated	Untreated and outsourced	Treated
Hydro carbon and alcohol	Treated	Treated
Incombustible	Treated with rate limitation	Treated
Other aspects		
Cost	Low	High
Chimney	Required	Not required
Odor	Concerned	Not concerned
Plant scale	Relatively large	Relatively small

carefully interpreted together with their characteristics. Table 3 organizes the characteristics of spray incineration and SCWO. Because spray incineration has a risk of dioxin creation, it cannot be used to treat halogenated solvents. Likewise, a filtration system must be installed adequately before the system for gas emission from the spray incineration process because of its risk of creating particulate matter, which has considerable health risks [47]. These emission risks are not a source of concern in the SCWO process. Additionally, SCWO has advantages over spray incineration. SCWO requires no smokestack or permanent odor control system. These advantages make the site of SCWO smaller than that of spray incineration. In this regard, however, there are some disadvantages of SCWO, such as the occupational risk due to the high pressure and temperature. Such severe conditions might lead to the high cost of operation to keep the process safe.

On the basis of the results of scenarios 0–2, the necessity of waste treatment was confirmed for acetone use. This is because acetone has potential impact on human health through photochemical oxidant creation. For various solvents, the impact originating from its treatment as scenarios 1 and 2 can be examined with those by direct emission. In Fig. 5, the human health impact due to the production and emission of the solvent generally used in the University of Tokyo are shown. On the basis of this result, the treatment by SCWO was addressed under the conditions of scenario 2. It was revealed that the treatments of 2-propanol and benzene by SCWO have larger impacts than the direct emission of these chemicals to the air without treatment. This is because the environmental impact of the emission of the chemicals is less than that of the energy use of the SCWO process. In this regard, however, the SCWO process

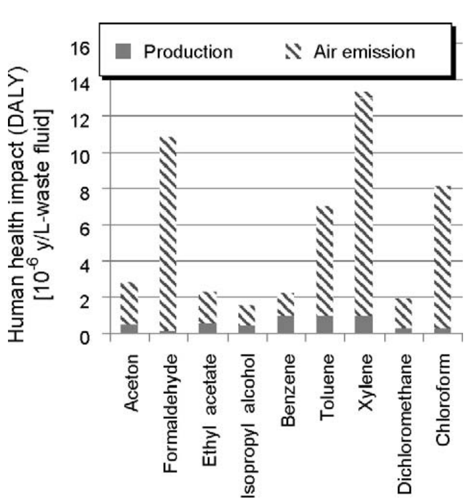

Fig. 5. Human health impact due to production and emission of solvent used in the University of Tokyo.

under the conditions of scenarios 3–6 can reduce the impact better than direct emission. Based on the results of this study, an analysis for other hazardous materials such as polychlorinated biphenyls (PCBs) and dioxins can be performed as the same way. Because these chemicals are relatively high persistent or bioaccumulative properties, the impact factors in LCA are also large. Note that the impact factors utilized in this study are based on the LIME2 method, which is customized for use in Japan. By using other impact assessment methods, the results may be different in terms of the original aspects [39].

In universities, waste fluid is a high-mix low-volume waste originating from laboratory research activities. Waste fluid can include various solvents with various minor constituents. At the same time, universities are usually located in residential areas. To address such conditions, treatment processes should be carefully selected on the basis of evaluation results and other characteristics. Spray incineration has less environmental impact than SCWO; therefore, it is acceptable to set up a treatment process with low environmental impact and cost. In this regard, however, spray incineration has disadvantages such as the risks of creating dioxins and particulate matter. SCWO can be regarded as an acceptable process because it requires no smokestack and odor control system, and poses negligible risk of creating dioxin and particulate matter. Halogenated solvents can also be treated by SCWO in the same way as other combustible wastes. To prevent blockage and corrosion of the SCWO reactor, substances generating precipitates in the reactor must be considered and the mixing rate of halogenated chemicals must be carefully adjusted.

For both processes, the means of outputting waste resulting from research activities is strongly related to the total performance of the treatment processes and the environmental impact. As shown in the evaluation results, an appropriate treatment can reduce environmental impact significantly. This means that researchers should try to collect all the solvents used, even if they are highly volatile. Moreover, the waste content should be adjusted and segregated properly on the basis of process characteristics. By involving researchers, a treatment system can be effectively established and the sustainability of research activities in universities can be discussed.

5. Conclusion

The results demonstrate that spray incineration has less environmental impact than SCWO. In this regard, however, SCWO has various advantages for installation as a treatment process in universities such as negligible risk of creating dioxins and particulate matter. The shift of operation to continuous, the adoption of liquid oxygen, and the installation of additional compressors and power recovery turbines can significantly reduce the environmental impact of SCWO, which is mainly related to electricity use. The selection of a treatment method for organic waste fluid requires a comprehensive analysis of risks. Moreover, not only the performance of the process such as cost, but also the completeness of the detoxification of hazardous materials should be taken into account. Spray incineration has the risk of creating dioxins and particulate matter. This means that waste including the studied materials related with such emission should be treated by SCWO. Utilizing the right technology for the right task in the detoxification of hazardous materials should be addressed for sustainable universities. It also requires cooperation by researchers to segregate waste to appropriate treatment processes. A treatment system involving technologies and users is needed for hazardous waste management to support sustainable development and research activities.

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