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Export of nutrients and suspended solids from major Arctic rivers and their response to permafrost degradation

ZHANG Shu-Min^a, MU Cui-Cui^{a,b,c,d,*}, LI Zhi-Long^a, DONG Wen-Wen^a, WANG Xing-Yu^a, Irina STRELETSKAYA^e, Valery GREBENETS^e, Sergey SOKRATOV^e, Alexander KIZYAKOV^e, WU Xiao-Dong^c

^a Key Laboratory of Western China's Environmental Systems (Ministry of Education), College of Earth and Environmental Sciences, Lanzhou University, Lanzhou, 730000, China

^b Southern Marine Science and Engineering Guangdong Laboratory (Zhuhai), Zhuhai, 519000, China

^c Cryosphere Research Station on the Qinghai–Tibetan Plateau, State Key Laboratory of Cryospheric Science, Northwest Institute of Eco-Environment and

Resource, Chinese Academy of Sciences, Lanzhou, 730000, China

^d University Cooperation of Polar Research, Beijing, 100875, China

^e Geographical Faculty, Lomonosov Moscow State University, Moscow, 119991, Russia

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Abstract

The rapid warming of the Arctic has led to permafrost degradation, accelerating the transport of terrestrial materials by rivers. The quantitative assessment of riverine nutrients and total suspended solids (TSS) flux is important to clarify the land—ocean connections in the Arctic. However, much is unknown about the estimates of these components from direct measurements in the Arctic rivers and the response of the components to permafrost degradation. Here, we report the results from the Arctic Great Rivers Observatory (Arctic-GRO) for the six major Arctic rivers (Yenisey, Lena, Ob', Mackenzie, Yukon, and Kolyma) to investigate the riverine exports of TSS, total dissolved nitrogen (TDN), nitrate (NO_3^-), bicarbonate (HCO_3^-), total dissolved phosphorus (TDP), and phosphate (PO_4^{3-}). The results showed that from 2004 to 2017, the annual TSS, TDN, and NO_3^- exports to the Arctic Ocean were approximately 106,026 Gg, 692 Gg, and 130 Gg, respectively, and the HCO_3^- , TDP, and PO_4^{3-} exports were approximately 79,092 Gg, 32 Gg, and 18 Gg, respectively. There were remarkable variations in component concentrations and fluxes between seasons. More than 80% of the TDN, TDP, PO_4^{3-} , and TSS exports mainly occurred in spring and summer, and a high HCO_3^- flux was recorded in summer, while a high NO_3^- flux in some rivers occurred in winter. The active layer thickness was significantly positively correlated with the annual TDN, NO_3^- , and HCO_3^- exports. In addition, the HCO_3^- flux of the six Arctic rivers increased by 247 Gg per year during 2004–2017. The positive relationship between the active layer thickness and river discharge indicates that permafrost degradation accelerated riverine carbonate, nitrogen, and phosphorus exports. This study demonstrates that riverine exports play an important role both in the Arctic terrestrial and marine ecosystems, and permafrost degradation will likely increase the riverine material exports to the ocean.

Keywords: Arctic rivers; Carbonate; Nutrients; Total suspended solids; Permafrost

* Corresponding author. Key Laboratory of Western China's Environmental Systems (Ministry of Education), College of Earth and Environmental Sciences, Lanzhou University, Lanzhou, 730000, China.

E-mail address: mucc@lzu.edu.cn (MU C.-C.).

1. Introduction

The Arctic region is an important component of the global climate system, and it has been warming at a rate of more than twice the global average for the period of 1961–2014 (AMAP, 2017). The rapid warming has resulted in the thawing of

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permafrost in the Arctic region (Biskaborn et al., 2019). The Arctic permafrost regions store a large amount of soil organic carbon, up to 1300 Pg C (Hugelius et al., 2014; Schuur et al., 2015). Permafrost thaw stimulates plant-available N in surface soils (Salmon et al., 2016), and large amounts of N are stored in deeper soils (Harden et al., 2012). Permafrost-affected peatlands store 185 Pg C of organic carbon and 7.1 Pg of total nitrogen (Hugelius et al., 2020). Meanwhile, phosphorus, an essential nutrient for all life forms (Ruttenberg et al., 2003), is also related to organic carbon in soil organic matter accumulation.

The Arctic Ocean accounts for 1% of the global ocean volume and receives more than 10% of the global river discharge (McClelland et al., 2012). The six largest Arctic rivers, the Ob', Yenisey, Lena, Mackenzie, Yukon, and Kolyma, cover approximately 67% of the pan-Arctic watershed (Goldman et al., 2012). The massive input of terrestrial materials strongly affects the Arctic Ocean. The organic matter composition in rivers is also related to seasonal changes in thawing depth (Neff et al., 2006), active layer thickness (O'Donnell et al., 2016; O'Donnell et al., 2010), groundwater discharge (O'Donnell et al., 2016), and permafrost coverage in watersheds (O'Donnell et al., 2016; Olefeldt et al., 2016). River exports of nutrients and organic matter affect the biogeochemistry of Arctic estuaries and the overall Arctic Ocean (Holmes et al., 2011). For example, river runoff significantly influences the dissolved inorganic carbon (DIC) and total alkalinity patterns in coastal regions, affecting aragonite saturation state, partial pressure of CO₂, and sea-air CO₂ flux seasonality (Gomez et al., 2020). In addition, an increase in Arctic river discharge (Mu et al., 2019; Savelieva et al., 2000; Lammers et al., 2001) can influence the ecosystems in Arctic coastal water and ocean (Carmack et al., 2016). Nitrate is the limiting nutrient in the Arctic Ocean, and the phosphorus supply can also affect the ecosystem productivity, salinity, and biogeochemical cycle of marine ecosystems (Emmerton et al., 2008; Hawkings et al., 2016); moreover, the riverine exports of phosphorus may affect the ecosystem in the estuaries and oceans. The total suspended solids (TSS) concentration is also related to hydrology and land use/cover (Murphy, 2020). Presently, a prevailing view is that the inorganic nutrient fluxes in permafrost-affected rivers are increasing (Shogren et al., 2019; Toohey et al., 2016; Drake et al., 2018). With the deepening of the active layer thickness, a seasonally thawed active layer contributes to more groundwater movement into rivers (Oelke et al., 2004; Rushlow et al., 2020), and the enhanced groundwater input will lead to increased matter transport in dissolved forms (Pokrovsky et al., 2020). Therefore, permafrost degradation is expected to increase the riverine exports of carbon, nitrogen, phosphorus, and suspended solids in Arctic rivers.

The Pan-Arctic River Transport of Nutrients, Organic Matter, and Suspended Solids project, which began in 2002, provides a dataset of dissolved organic matter and inorganic nutrients obtained via identical sampling and analytical methods for the six largest Arctic rivers (the Arctic Great Rivers Observatory, Arctic-GRO; www.arcticgreatrivers.org/).

Based on this dataset, several reports on materials exports have been provided, considering dissolved organic carbon and inorganic carbon (Griffin et al., 2018; Tank et al., 2012) and particulate organic carbon and nitrogen (Holmes et al., 2011; McClelland et al., 2016; Raymond et al., 2007); however, much is unknown about the exports of TSS, total dissolved nitrogen (TDN), NO_3^- , HCO_3^- , total dissolved phosphorus (TDP), and PO_4^{3-} . In addition, little is known about the relationship between material exports and permafrost degradation.

The current study, using data on the Yenisey, Lena, Ob', Yukon, Mackenzie, and Kolyma rivers, aimed to quantify the following: 1) the seasonal concentrations of TSS, TDN (including NO_3^- , NO_2^- , NH_4^+ , and dissolved organic nitrogen), NO_3^- , HCO_3^- , TDP (PO_4^{3-} and dissolved organic phosphorus), and PO_4^{3-} ; 2) annual and seasonal exports of TSS, TDN, NO_3^- , HCO_3^- , TDP, and PO_4^{3-} ; 3) the possible relationship between the active layer thickness and the exports of TSS, TDN, NO_3^- , HCO_3^- , TDP, and PO_4^{3-} . Through the analysis of observation data, this study provides insights into the effects of permafrost degradation in relation to nutrient export from Arctic lands to the Arctic Ocean and ultimately to the global carbon and nitrogen cycle.

2. Data and methods

The six Arctic rivers of Yenisey, Lena, Ob', Mackenzie, Yukon, and Kolyma were considered (Fig. 1). The Ob' River has the largest area and the smallest permafrost coverage, while the Kolyma River has the smallest area and 100% permafrost coverage (Table 1). The dataset of water quality and daily discharge of the Arctic rivers for 2004–2017 (excluding 2007 and 2008) was obtained from the Arctic-GRO (https://arcticgreatrivers.org/), including the HCO₃⁻, TDN, NO₃⁻, and TDP concentrations. The period for phosphate (PO₄³⁻) calculation was 2009–2017, and that for TSS was 2004–2018. The active layer thickness data for 2004 to 2017 in the Arctic regions were obtained from the Arctic Circumpolar Active Layer Monitoring network and the Global Terrestrial Network for Permafrost.



Fig. 1. Study area, mainly including the basins of the Yenisey, Lena, Ob', Mackenzie, Yukon, Kolyma Rivers.

Table 1					
Environmental	conditions	of the	six	Arctic	watersheds

Watershed	Drainage area (10 ⁶ km ²)	Permafrost coverage (%)	Continuous permafrost coverage (%)	Discontinuous permafrost coverage (%)	Average active layer thickness (cm)
Yenisey	2.56	88	33	11	86
Lena	2.40	99	79	11	37
Ob'	2.95	26	2	4	120
Mackenzie	1.75	82	16	29	107
Kolyma	0.65	100	100	0	82
Yukon	0.83	99	23	66	51

The details of sampling protocols are available on https:// arcticgreatrivers.org/. The field monitoring was based on the U.S. Geological Survey (USGS) equal discharge incremental sampling protocol, whereby samples were collected across the river channel. During open-water periods, five deep integrated samples were collected using the USGS D-96 deep integrated sampler equipped with an insulated nozzle and an insulated collection bag. The samples were fed into a 14 L insulated mixer to form composite samples, which were used to clarify the vertical and horizontal heterogeneities. During the ice-covered periods, the samples were collected at the central point of the maximum flow through a separate channel and an intermediate channel hole about 10 cm below the bottom of the surface ice layer. Total suspended solids were transferred into high-density polyethylene bottles from the sampling churn. The dissolved components, including HCO_3^- , inorganic nutrients (NO₃⁻, NH₄⁺, PO₄³⁻), and TDN, TDP), were filtered through AquaPrep 600 capsule filters with 0.45 µm pore size and frozen in a precleaned high-density polyethylene bottle. Alkalinity was determined via titration using a Hach digital titrator and the Gran method, with the online USGS calculator. The TDN concentration was determined using a Shimadzu total organic carbon/total nitrogen analyzer. The TDP concentration was measured via the ascorbic acid method following persulfate oxidation. The NO₃⁻ and PO₄³⁻ concentrations were analyzed using a Lachat Quickchem FIA+ 8000 instrument.

We categorized the river discharge, TSS, and nutrient concentrations by three seasons: spring (May to June, 61 d), summer (July to October, 123 d), and winter (November to April, 181 d) (Striegl et al., 2005). An uncertainty analysis was conducted through the percentile method, and we used a quartile confidence interval to find the range of carbon and nutrient exports. We employed min—max normalization to quantify the annual increases in active layer thickness and discharge and the relationship between them. The min—max normalization can transform the minimum and maximum data to 0-1 while retaining the original distribution of scores. We used simple linear regressions to examine the relationships between nutrient concentration, discharge, and active layer thickness (Mu et al., 2019).

3. Results

3.1. HCO_3^- , TSS, TDN, NO_3^- , TDP, and PO_4^{3-} concentrations

Strong relationships were observed between the discharge and HCO_3^- , NO_3^- , TDP, and TSS concentrations (p < 0.05),

and there were seasonal variations in the concentrations for Lena and Mackenzie rivers (Fig. 2). For the HCO_3^- and NO_3^- , the highest concentrations occurred in winter, while the lowest occurred in spring and summer. There were no significant patterns for the TDN, TDP, and PO_4^{3-} .

3.2. Export of TSS, TDN, NO_3^- , HCO_3^- , TDP, and PO_4^{3-}

The seasonal and annual exports of nutrients and TSS in the Arctic rivers are summarized in Table A1. The six rivers exported nearly half of HCO_3^- in summer. The TDN, TDP, and PO_4^{3-} exports in spring and summer were equivalent, accounting for more than 80% of the total flux (Fig. 3). However, there were great variations in seasonal NO_3^- exports among the six rivers. Compared with the river exports of TDN and TDP at the continental scale, nutrient fluxes exported in Arctic rivers were remarkably lower (Table 2).

The average flux ratio of TDN to TDP was 25.6 in the Arctic rivers (Fig. 4), with the Ob' River having the lowest value (8.7) and the Yukon River having the highest value (53.0). There were great seasonal variations in the TDN to TDP ratio, with higher values in the winter and lower values in summer and spring.

3.3. Relationship between active layer thickness and nutrients and TSS exports

The HCO₃⁻ flux had a significant positive correlation with the active layer thickness (Table 3). The TDN and NO₃⁻ fluxes in most rivers, except Yenisey, had positive relationships with the active layer thickness. However, a negative correlation existed between NO₃⁻ and the active layer thickness in Mackenzie. The TDP, PO₄³⁻, and TSS had no significant linear relationship with the active layer thickness (Table 3).

The active layer thickness and discharge of the six Arctic rivers both increased significantly as the year progressed between 2004 and 2017, and the active layer thickness was also significantly correlated with the discharge (Fig. 5), indicating that the active layer thickness and discharge of the Arctic rivers increased gradually during the past two decades.

From 2004 to 2017, the HCO_3^- flux of the six Arctic rivers increased at a rate of 247 Gg per year. In the study period, the exports of HCO_3^- in Yenisey, Lena, Ob', Mackenzie, Yukon, and Kolyma rivers increased by 3792 Gg, 4371 Gg, 3516 Gg, 2056 Gg, 3290 Gg, and 1109 Gg, respectively, with an average value of 3022 Gg. However, there was no significant



Fig. 2. Discharge with HCO_3^- , total dissolved nitrogen (TDN), NO_3^- , total dissolved phosphorus (TDP), PO_4^{3-} , and total suspended solids (TSS) concentrations in Lena and Mackenzie rivers (p < 0.05).



Fig. 3. Percentage of river flow and material fluxes in different seasons during 2004-2017 (PO₄³⁻ during 2009-2017).

Table 2										
Comparison of	of TDN	fluxes	between	the	Arctic	and	other	continents	(unit:	Pg
per year).										

Region	TDN	TDP
Arctic	0.69	0.03
Oceania	3.80 ^a	0.18 ^a
North America	4.30 ^a	0.20 ^a
South America	10.30 ^a	0.41 ^a
Europe	3.00 ^a	0.24 ^a
Australia	0.30 ^a	0.01 ^a
Northern Asia	3.70 ^a	0.25 ^a
Southern Asia	6.90 ^a	0.26 ^a
Africa	4.00^{a}	0.21 ^a

Note:

^a Riverine exports of nutrients by continent (Seitzinger et al., 2005).

increasing or decreasing trend for other nutrients. The TSS exports during 2004–2018 showed significant decreasing trends (Fig. 6).

4. Discussion

The estimated annual NO_3^- , TDN, and TDP fluxes of the six Arctic rivers in 2004–2017 averaged approximately 130.3 Gg, 692.0 Gg, and 31.9 Gg, respectively, which are similar to the results obtained via calculation using the Load Estimator



Fig. 4. Annual and seasonal flux ratios of total dissolved nitrogen to total dissolved phosphorus in the six rivers.

program model for 1999–2008 (Holmes et al., 2011). The Load Estimator is a program for estimating constituent loads in streams and rivers. The mean load estimates, standard errors, and 95% confidence intervals are developed on a monthly and (or) seasonal basis. The estimated annual PO_4^{3-} flux of the six Arctic rivers in 2004–2017 in this study was approximately 18.2 Gg, which was lower than the 37.6 Gg for

Table 3 Linear regression between carbon, nitrogen, and phosphorus fluxes and active layer thickness in the Arctic rivers in 2004–2017.

Element		Yenisey	Ob'	Lena	Mackenzie	Yukon	Kolyma
HCO ₃	slope	0.65*	0.84*	0.61*	0.82*	0.63*	0.54*
TDN	slope	_	0.83*	0.55*	0.63*	0.60*	0.50*
NO_3^-	slope	_	0.60*	0.48*	-0.20*	0.65*	0.53*
TDP	slope	_	0.81*	0.67*	_	_	_
PO_4^{3-}	slope	_	0.59*	_	_	_	_
TSS	slope	-	-0.19*	_	-	-3.46*	_

Note: -p > 0.05; *p < 0.05.



Fig. 5. Changes in active layer thickness and discharges and their linear relationship for the Arctic rivers during 2004–2017 (The data were normalized through the min–max method, and there were no units).

1985–1995 derived from the Unified Federal Service for Observation and Control of Environmental Pollution Nutrient dataset and the R-ArcticNet discharge data (www.R-arcticnet. sr.unh.edu/) (Holmes et al., 2000). The difference between the results of the studies can be attributed to the differences in the



Fig. 6. Variation in HCO_3^- flux between 2004 and 2017 (a) and total suspended solids flux between 2004 and 2018 (b).

source and timespan of the datasets and the calculation methods (Holmes et al., 2011). We estimated the annual DIC flux from six Arctic rivers during 2004–2017 to be 79 Tg, which accounted for 19.7% of the global DIC export from land to ocean (Ren et al., 2015). Our estimate is higher than the 57 ± 9.9 Tg per year for the full Arctic catchment during 2000-2009 (Tank et al., 2012), which was calculated based on the relationship between the DIC yield and runoff, the presence of carbonates, glacial coverage, and continuous permafrost. The difference between the two results is not only due to the different calculation methods but also due to the rapid increase in the DIC export in recent decades. For example, from 1978 to 2015, the exports of DIC from the Yenisei and Ob' Rivers increased by 185% and 134%, respectively (Drake et al., 2018). Our study estimated that the annual TSS export in the six Arctic rivers was approximately 108 Tg, indicating that the TSS export in the Arctic rivers is an important component of the terrestrial material exports to the Arctic Ocean (Holmes et al., 2002). The TSS output of Arctic rivers is lower than those of temperate and tropical rivers (Syvitski, 2002), which can be explained by the low erosion processes of the land areas

because of the long frozen period, low precipitation, and high vegetation coverage in the circumarctic regions (Favaro and Lamoureux, 2015; Syvitski, 2002).

The seasonal variation of nutrients and TSS export can be attributed to the changes in discharge and the nutrient and TSS concentrations (Tank et al., 2012; Holmes et al., 2000). The HCO_3^- flux in winter accounted for 26% of that of the whole year, while those in spring and summer accounted for 26% and 48% of that of the whole year, respectively. The HCO_3^- concentrations in winter and summer were higher and slightly lower than that in spring, respectively (the durations of summer and winter were four months and six months, respectively, while the spring duration was only two months). Although the HCO_3^- concentration was lower in summer, the higher discharge and longer timespan led to the higher HCO₃⁻ export in summer. In winter, the base flow of water partly originated from glacier outlet water and subglacial sediments and thus had considerably high DIC concentration (Tank et al., 2012). The patterns for other nutrients, including TDN, NO₃⁻, TDP, and PO_4^{3-} , were more variable than that for HCO_3^- across the river basin and seasons, revealing the strong control of the river network on biogeochemical signals (Shogren et al., 2019).

We also examined the seasonal patterns of TSS, TDN, NO_3^- , TDP, and PO_4^{3-} fluxes. On average, TDN accounted for about 41% and 38% of the annual flux in spring and summer, respectively, but only 21% of the flux in winter. The TDN fluxes of Yenisey, Lena, and Kolyma in spring exceeded 50%, while the flux in winter in the Ob' River exceeded 30% of the annual flux. The difference in contributions between seasons can be explained by the different seasonal dynamics among the different rivers (Dittmar and Kattner, 2003). For example, 43% and 21% of the NO_3^- and TDN exports, respectively, occurred in winter. The TDN concentrations in spring and summer were much higher than the NO_3^- concentrations, and TDN includes not only inorganic nutrients such as NO₃⁻ and NH_4^+ but also dissolved organic nitrogen. It has been suggested that dissolved organic nitrogen is the main component of TDN in Arctic rivers, while dissolved inorganic nitrogen only constitutes a small part of the TDN (Guo et al., 2004). The organic matter composition in rivers is also related to the seasonal changes in thawing depth (Neff et al., 2006), active layer thickness (O'Donnell et al., 2016; O'Donnell et al., 2010), groundwater discharge (O'Donnell et al., 2016; O'Donnell et al., 2012), and permafrost coverage in the watersheds (O'Donnell et al., 2016; Olefeldt et al., 2016). Moreover, NO₃⁻ mainly originates from terrestrial ecosystems (Dornblaser and Striegl, 2007), and the cold temperature in winter limits the NO_3^- assimilation by plants. Furthermore, the basic flow in winter contains a large amount of groundwater (Frey and McClelland, 2009), which may also partly explain the high nitrate concentration and flux ratio in winter. In this study, the seasonal patterns of TDP and PO_4^{3-} were similar, with approximately 40% in spring and summer but only 20% in winter. Most phosphorus in rivers is derived from chemical weathering (Dornblaser and Striegl, 2007). The fluxes of TSS in spring, summer, and winter were 49.2%, 48.8%, and 2.0% respectively. During the early period, suspended solids delivery was not similar to the discharge until the maximum TSS concentration appeared in June. Since then, owing to the complete river channel connection and the limited influence of snow in the river channel, the sediment transport and discharge exhibited similar patterns and showed extremely low values in winter (Favaro and Lamoureux, 2015). Moreover, the TDN/ TDP value in the Arctic rivers was higher than the Redfield ratio (16:1), which suggests the relative lack of phosphorus in river water transported to coastal zone. The TDN/TDP value of the Ob' River was lower than the Redfield ratio, indicating that the phosphorus content transported by the Ob' River was higher than those transported by other rivers (Holmes et al., 2011).

The continuous increase in HCO_3^- flux in recent years may be explained by the permafrost degradation, which can also cause an increase in the runoff and carbonate flux by increasing the contribution of groundwater (Frey and McClelland, 2009). In addition, owing to the deepening of active layer thickness, water permeates into deeper mineral soil, which aggravates weathering (Frey and McClelland, 2009). Changes in the streamflow regime may ultimately result in changes in the TSS concentration of a river (Murphy, 2020). This process can include geomorphological changes such as increased bank eroding and channel bottom scouring and resuspension (Murphy, 2020). Therefore, the increases in the active layer thickness and the permafrost degradation will affect the TSS transport through rivers and soil erosion, but these increases were not the causes of the decrease in TSS. There were no significant changes in TDN, NO₃⁻, TDP, and PO_4^{3-} with the year. Our results show that the TDN export in Arctic rivers, except for the Yenisey River, can be affected by the active layer thickness. Samples of Russian rivers entering the Arctic Ocean were taken from near the estuary at salinities below 0.01, and the sample from the Yenisey River had a salinity of 1.68, showing a slight mixing with seawater (Lobbes et al., 2000). The Yenisey River featured the most alkaline pH (9.4), whereas among the Russian rivers, the Lena River had the minimum pH value (7.2) (Lobbes et al., 2000). It has been suggested that nutrient concentration decreases with salinity (Lobbes et al., 2000; Hessen et al., 2010). Furthermore, although the active layer thickness had a positive relationship with the TDN and NO_3^- outputs, several other factors affect the TDN and NO₃⁻, such as groundwater (Walvoord and Striegl, 2007), terrestrial organics, and salinity (Hessen et al., 2010). Different from TDN and NO_3^- , TDP and PO_4^{3-} did not show linear relationships with the active layer thickness. This may be because the TDP and PO_4^{3-} concentrations in permafrost regions were low (Frey and McClelland, 2009; Frey et al., 2007), and thus, it is difficult to observe the weak trends between the active layer thickness and the phosphorus concentration and flux.

5. Conclusion

Using data from Arctic-GRO, we calculated the concentrations of riverine materials and their relationships with active layer thickness. We estimated the exports of HCO_3^- , TDN, NO_3^- , TDP, PO_4^{3-} , and TSS discharged from six Arctic rivers to the Arctic Ocean. The seasonal variation in nutrients and TSS export can be attributed to the changes in discharge and the nutrient and TSS concentrations. Our results indicate that in the past 20 years, the active layer thickness and river discharge exhibited increasing trends, and these changing trends will likely lead to increasing nutrient exports, while there will be a declining trend in the TSS.

Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.accre.2021.06.002.

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