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Changes in the impact of anthropogenic effects on river water quality during the last 50 years in Japan

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Abstract Japan's rapid urbanisation over the last 50 years has resulted in land use and lifestyle changes, all of which are likely to have changed the quality of river water, and consequently the wetland and coastal environment. We examined changes in river water quality over this period by means of a review of previous studies. Around the 1950s, the weighted average of chloride using discharge of Japan's 30 major rivers was 6.1 mg/l while in the 2000s it was 11.3 mg/l. Because there were no significant changes in the natural conditions, we have attributed the increase to the urbanisation of the last 50 years. Nitrate levels in the mountain streams of southern Japan have increased, particularly in the western part of the Kanto region. As this area is located on the leeward side of the Tokyo Metropolitan Area, depositions from aerosols are thought to be the main cause of the increased nitrate concentration. These two findings suggest that certain uses of land may affect river water quality differently over time, and that changes in land use may also affect river water quality in remote areas.

Keywords Urbanisation Watershed . Chloride · Nitrate · Lifestyle

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Introduction

Many countries experienced rapid urbanisation in the 20th century, a trend that will continue in the 21st century. The United Nations ([2006\)](#page-6-0) stated that the urban proportion of the global population changed from a mere 13% in 1900 to 29% in 1950 and reached 49% in 2005, with 60% of the global population expected to live in cities by 2030. Because the coastal domain (sea level ± 200 m) contains two thirds of the world's cities (LOICZ [1996\)](#page-6-0), and approximately 37% (2.07 billion) of the world's population live within 100 km of a coastline (Cohen et al. [1997\)](#page-5-0), urbanisation will directly alter the health of coastal ecosystems, including wetlands.

Urbanisation is characterised by land use changes from unaltered natural land to artificial land use (e.g. roads, residential areas, commercial areas). This change affects the water quality in rivers running through urban watersheds. Baker [\(2003](#page-5-0)) reported that urban areas are the largest source of chloride. Wayland et al. [\(2003\)](#page-6-0) found that urbanized areas are associated with higher concentrations of sodium, potassium and chloride in river water, which they attributed to the practice of road salting. Other studies have also reported the positive correlation between the area of urbanisation and the concentration of chloride in river water (Hirose and Kuramoto [1981](#page-5-0); Jarvie et al. [2002;](#page-5-0) Schoonover et al. [2005\)](#page-6-0).

Cultivation is another anthropogenic effect which alters river water quality. In the United States,

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agriculture is the primary source of stream impairment (US-EPA [2000\)](#page-6-0). Woli et al. [\(2004](#page-6-0)) investigated the nitrogen budget in livestock farming areas and found that intensive livestock farming areas had the highest impact on the budget, and that mixed agriculture and livestock farming areas had a medium influence, while grassland-based dairy cattle and horse farming areas had the lowest impact. Smart et al. ([1985\)](#page-6-0) and Neill [\(1989](#page-6-0)) reported a positive correlation between the proportion of farmed land in drainage basins and the nitrate concentration in river water.

Watershed management and catchment scale studies have become increasingly important in determining the impact of human activities, such as urbanisation and cultivation, upon water quality within watersheds as well as receiving waters. It is impossible to separate the water quality management of rivers from that of riparian wetlands or enclosed bays including salt marshes and tidal flats. Many studies reported rivers as major paths bringing eutrophication inducing contaminants to downstream wetlands (e.g. Collavini et al. [2005](#page-5-0); Panigrahi et al. [2007](#page-6-0)). This is also true for the remediation of downstream ecosystems, because these ecosystems suffer direct damage from the eutrophication induced by the increased nutrient flux from the watersheds.

However, effects of urbanisation, i.e. in the efflux of certain elements from an area, may not be constant over long time spans. As an extreme example, Edo City, which until 1867 was the name for what is now part of the Tokyo Metropolitan Area, was an urban area with a population of more than 1 million after 1721. However, it is safe to assume that the efflux of elements of Edo City and that of the present-day Tokyo Metropolitan Area would be quite different due to the changes in lifestyle over time, in particular the drastic changes in both land use and lifestyle in Japan during the past 50 years. Therefore, the effect of land use on river water quality may not have remained the same over the entire period. Moreover, some human activities, such as industrialization and motorized transportation, increase the volatile efflux of inorganic elements, which may result in the increase of certain elements in remote areas.

To elucidate the effects of lifestyle change and urban expansion upon water quality, we selected chloride as a target ion and compared the concentrations found in river waters 50 years ago with those found today. We also compared the nitrate concentration of mountain rivers during this period to examine the effects of lifestyle change and/or urbanisation on remote areas.

Materials and methods

To compare the chloride concentration in river waters between the present and 50 years ago, we used the data described by Kobayashi [\(1961a\)](#page-6-0) and by Tagami and Uchida [\(2006](#page-6-0)).

Kobayashi [\(1961a\)](#page-6-0) analysed major ions in water samples taken from more than 225 rivers across Japan. The samples were mostly taken in the 1950s, with the remainder having been taken in the 1930s or 1940s. They were collected at least six times a year, and only the average of all the analytical results was recorded.

Tagami and Uchida [\(2006](#page-6-0)) analysed some of the major inorganic ions, including chloride, in water samples taken from 30 rivers from 2002 to 2005. The samples were each taken only once at each location, and mostly during summer. The rivers analysed by Kobayashi [\(1961a\)](#page-6-0) include all the 30 rivers studied by Tagami and Uchida ([2006\)](#page-6-0), although Kobayashi [\(1961a\)](#page-6-0) took his sample from only one location along each river, while Tagami and Uchida [\(2006](#page-6-0)) analysed water sampled from ten different points along each river. To minimize the effect of location within the river, we chose the data from Tagami and Uchida [\(2006](#page-6-0)) that was sampled at the same or the closest point to that described by Kobayashi [\(1961a](#page-6-0)). As the sampling time for the 2000s is only once a year, we added mean value, standard deviation (SD) and coefficient of variance (CV) of electric conductivity (EC) for the rivers in the results and discussion section to show the annual fluctuations, using data from more than 800 water monitoring stations maintained by the Ministry of Land, Infrastructure, Transport and Tourism, Japan (MLIT). We selected the data from MLIT [\(2002](#page-6-0)) as their sampled year coincides with the data of Tagami and Uchida [\(2006](#page-6-0)). The annual average of the EC value was calculated from observed data 6 times in a year. As we could not get the data of the Shou River, taken in the 2000s, we chose data taken in the nearest year, 1999. Because there were no data for the Yubetsu River, we did not state the EC data for it.

Nitrate concentrations were compared according to two different regional focuses: the whole of Japan and the Kanto region alone. Here, we first compared the data of Kobayashi [\(1961a\)](#page-6-0) and Konohira et al. [\(2006](#page-6-0)). While Kobayashi ([1961a](#page-6-0)) took river water samples from the mid- to downstream parts of the rivers, located on the alluvial plain, Konohira et al. [\(2006](#page-6-0)) took samples from 1296 points in mountain streams, and averaged the data by each prefecture. Because we could not compare the nitrate concentration at different times with the data sampled from the same and/or a close position, we averaged the data within the eight regions, i.e. Hokkaido, Tohoku, Kanto, Chubu, Kinki, Chugoku, Shikoku, and Kyushu. Thus, the average nitrate concentration of the river waters in each district using the data in Kobayashi [\(1961a\)](#page-6-0) would represent the downstream part at around 50 years ago, while the same average using the data in Konohira et al. [\(2006\)](#page-6-0) would represent the mountain streams at present.

Secondly, to determine more details of the change in nitrate concentration in the Kanto region, data from both the Kobayashi [\(1955](#page-6-0)) and Konohira et al. ([2006\)](#page-6-0) studies regarding the Kanto region were compared. Kobayashi [\(1955](#page-6-0)) tested 65 water samples throughout the Kanto region (this region comprises the 6 prefectures of Chiba, Gunma, Ibaraki, Kanagawa, Saitama, and Tochigi, along with Tokyo), 22 of which were sampled higher than 100 m above sea level and were selected for our study. Although the sampling points in Ibaraki and in the southern part of Kanagawa are located slightly lower than 100 m, these two sampling locations were incorporated in the analysis because they were the only points sampled in the Kuji and the Sakaga Rivers respectively. These data were chosen as they were also from the upper reaches of the river waters (mountain river water), and averaged for each prefecture for comparison with the data of Konohira et al. [\(2006](#page-6-0)).

Results and discussion

Figure 1 shows the increase in chloride concentration during the past 50 years. The river water samples analysed by Kobayashi ([1961a](#page-6-0)) had been sampled before the beginning of rapid urbanisation in Japan. Because natural effects such as geological transformations would not change so drastically within 50 years, we can therefore attribute most of the increase to anthropogenic effects.

Fig. 1 Increase in the chloride concentration in Japanese rivers in the last 50 years determined by subtracting Kobayashi's data [\(1961a](#page-6-0)) from that of Tagami and Uchida ([2006\)](#page-6-0)

A remarkable increase in chloride ions is observed in the river water received from drainage basins which contain an intensively urbanised area. Chloride concentration in the River Ishikari (see Fig. 1, #3), whose drainage basin contains Sapporo City, increased by 17.1 mg/l (from 11.3 to 28.4 mg/l (EC: mean 15.4 mS/m, SD 5.0, CV 32.6)). The Ara River (#12), whose drainage basin contains Saitama City and the northern and eastern part of the Tokyo Metropolitan Area, saw an increase in chloride concentration of 11.2 mg/l (from 7.0 to 18.2 mg/l (EC: mean 39.9 mS/m, SD 12.2, CV 30.6)). The Yodo River (#20), whose drainage basin contains Osaka City, saw an increase in chloride concentration by 13.8 mg/l (from 4.9 to 18.7 mg/l (EC: mean 16.2 mS/m, SD 1.6, CV 9.8)). In contrast, the Kiso River (#19), whose drainage basin contains only the edge of Nagoya City (making the sampling point of Kobayashi ([1961a](#page-6-0)) and Tagami and Uchida ([2006\)](#page-6-0) slightly upstream of the urban area), showed an increased chloride concentration of a mere 0.8 mg/l (from 1.4 to 2.2 mg/l (EC: mean 6.8 mS/m, SD 0.95, CV 13.9)).

Chloride concentration in the Ishikari (#3), Ara (#12) and Yodo (#20) rivers increased by 2.5, 2.6 and 3.8 times, respectively. Meanwhile, the population in the corresponding river drainage basins had increased by 1.6, 3.1 and 1.7 times, respectively (statistics from the Ministry of Land, Infrastructure, Transport and Tourism of Japan). The data shows a strong correlation between areas of increased urbanisation and higher chloride concentrations, which suggests that these higher concentrations are caused by the process of urbanisation itself.

Rivers whose chloride concentrations had decreased have not experienced heavy urbanisation within their drainage basin. The Teshio River (#1) with the lowest value $(-2.5 \text{ mg/l}$ and changed from 1.4 to 2.2 mg/l (EC: mean 6.8 ms/m, SD 0.95, CV 13.9)), does not flow through a highly developed area. Likewise, the Yubetsu River (#2), the Seki River (#15) and the Shou River (\#16) , which showed slight decreases of -0.2 mg/l (from 2.2 to 2.0 mg/l), -0.4 mg/l (from 17.8 to 17.4 mg/l (EC: mean 17.4 mS/m, SD 6.5, CV 37.3)) and -0.9 mg/l (from 3.2 to 2.3 mg/l (EC: mean 6.7 mS/m, SD 0.5, CV 7.4)) respectively, do not flow through urbanised areas.

Kobayashi ([1961a](#page-6-0)) noted the high concentration of chloride along the coast of the Sea of Japan, which he attributed to the excessive supply of sea salt transported by westerly winds. However, the data shown in Fig. [1](#page-2-0) is the subtracted values of the 1950s from the 2000s, and as there is no significant increase on the coast of the Sea of Japan, we can say the sea salt supply has not changed in the 50 years. The effects of the area's geology would not change drastically during the period. Therefore, we can conclude that the effects of urbanisation are the main reasons for the increased chloride concentration.

Some previous studies (e.g. Jarvie et al. [2002](#page-5-0); Baker [2003](#page-5-0); Schoonover et al. [2005](#page-6-0)) found that increased urbanisation induced an increase in chloride concentration, and other reports focused on the practice of road salting to melt ice in winter as a likely cause (e.g. Wayland et al. [2003](#page-6-0)). Since Tagami and Uchida ([2006\)](#page-6-0) took their samples in the summer and autumn, road salting could not be the cause of the increase in chloride concentration found in this study.

Sewage is a major cause of increases in chloride concentration (Hirose and Kuramoto [1981;](#page-5-0) Wayland et al. [2003](#page-6-0)), and this factor can be applied to the situation in this study. In 1953, the Japanese government brought in a law to maintain the safety of tap water. The new law required that tap water be chlorinated with a level of 0.1 mg/l free residual chlorine or 0.4 mg/l combined residual chlorine. This may also be the part of the reason for the chloride increase in urbanised areas. The disinfection process in wastewater treatment facilities using chlorination may also contribute to the increase in the chloride concentration in the rivers and lakes around highly populated areas.

Using the same dataset reported in his earlier paper [\(1961a\)](#page-6-0), Kobayashi [\(1961b](#page-6-0)) also compared the chloride concentration of Japanese rivers with those of the rest of the world in the 1950s, and both sets of data showed almost the same concentrations (5.8 and 5.7 mg/l, respectively).

To compare how the average chloride concentration has changed over 50 years in the 30 rivers for Japan as a whole, weighted averages using discharge were calculated. The results were 6.1 mg/l in the 1950s and 11.3 mg/l in the 2000s. The urbanisation that has taken place during the past 50 years has significantly increased the chloride concentration in Japanese rivers.

Figure [2](#page-4-0) shows the difference in nitrate concentration averaged within the eight general regions of Japan. We compared the average nitrate concentration of the river waters in each region using the data in Kobayashi's 1961 study [\(1961a\)](#page-6-0), which were samples taken from the mid- to downstream area around 50 years ago, with the same average using the data from Konohira et al. [\(2006](#page-6-0)) recently sampled from mountain streams.

There is a large contrast between northern and southern regions. Nitrate concentrations in the Hokkaido and Tohoku regions decreased over time, and most areas of these regions are mountainous compared with those of southern parts in the country.

Southern regions though, showed an increase in nitrate over the last 50 years: the nitrate concentrations in the Chubu, Kinki, Chugoku, Shikoku and Kyushu regions ranged from 0.08 to 0.26 mg/l. The Kanto showed the highest increase, with a value of 0.62 mg/l. In the last 50 years, these areas have developed dramatically, and the South also has a large number of industrial regions and heavily populated cities along the Pacific coast.

145 $130°$ 140° 135 45 45 Hokkaido 0.6 0.2
 0.1 $40^{^{\circ}}$ 40 -0.4 -0.1 Tohoku $NO₃-N$ [mg/L] Chubu Chugoku 35 35 Kanto Kinki Shikoku Kyushu 145° 140 130 135

Fig. 2 Increase in the nitrate concentration in eight Japanese regions in the last 50 years determined by subtracting Kobayashi's data ([1961a\)](#page-6-0) from that of Konohira et al. ([2006\)](#page-6-0)

Because we did not compare results from the same locations in each river, we checked the trend in the nitrate concentration within the river for the present and 50 years ago. Figure 3 shows nitrate concentration changes according to the flow from the head waters in some rivers in the Kanto region in the 1950s. Almost all rivers show lower nitrate concentration at the headwaters than in the mid- to downstream parts.

The NIRS ([2007\)](#page-6-0) sampled river water throughout Japan from 2002 to 2006. The nitrate concentration data showed the same trends as those in Fig. 3: lower nitrate concentration at the headwaters than in the mid- to downstream. Thus, the higher concentration of nitrate in the mountain rivers in the 2000s than in the mid- to downstream areas in the 1950s (Fig. 3) denotes a higher overall nitrate concentration in mountain rivers now than 50 years ago. Because there have been no significant changes of the land use/cover in the mountain river watersheds of Japan over this 50 year period, the emission of gaseous wastes from industrial plants or vehicles from urbanised areas is thought to be the main cause of the nitrate increase. Okubo et al. ([2007\)](#page-6-0) analysed the change in nutrient concentration in inflow rivers to Lake Biwa, located in the Kinki region, over the past

Fig. 3 Nitrate concentration changes according to the flowdown from the headwaters in the Kanto Region. The horizontal line shows the station distance from the headwaters. Data are from Kobayashi [\(1955](#page-6-0)). The Rivers are labelled alphabetically (A: Kuji river, B: Naka river, C: Kinu river, D: Tone riv., E: Ara river, F: Tama river, F: Sagami river and G: Sakaga river). These characters correspond with the labels in Fig. [4](#page-5-0)

25 years, and found that nitrate had increased, while ammonium and phosphate had decreased. Okubo et al. ([2007\)](#page-6-0) attributed the nitrate increase to the emissions in the air influenced by the motorized transportation in the neighbouring regions.

Figure [4](#page-5-0) shows the increase in nitrate concentration in the mountain rivers in the Kanto region. There is a large contrast between the western region (Gunma, Saitama, Tokyo and Kanagawa) and the eastern region (Tochigi and Ibaraki). The western region showed a large increase, whereas that in the eastern region was relatively low. Sakio ([1999\)](#page-6-0) analysed and compared the pH and nitrate concentrations of rain water sampled in a region neighbouring an area of air pollution with samples from the mountainous region in Saitama Prefecture; most of the values were identical. Aoi et al. ([2004\)](#page-5-0) reported a high concentration of nitrate in the river and rain water on a forested mountain in Gunma Prefecture. Based on the positive correlation between nitrate concentration and wind direction during sampling, they attributed the source of nitrate to the Tokyo Metropolitan Area.

Shibata et al. [\(2001](#page-6-0)) reported significantly higher nitrate concentrations in stream water in central Japan, including the Kanto region, compared with the other regions. They concluded that the large amounts of precipitation during the warm period from June to September, along with atmospheric nitrogen deposition,

Fig. 4 Increase in nitrate concentration in the Kanto region in the last 50 years determined by subtracting Kobayashi's data ([1955\)](#page-6-0) from that of Konohira et al.([2006\)](#page-6-0). Because the samples were not taken by Konohira et al. ([2006\)](#page-6-0), data from Chiba Prefecture has not been plotted. Major rivers are labelled alphabetically (A: Kuji river, B: Naka river, C: Kinu river, D: Tone river, E: Ara river, F: Tama river, G: Sagami river and H: Sakaga river), and their watersheds coincide with the prefecture boundaries in most of the cases in the upper stream site. Kobayashi's [\(1955](#page-6-0)) sampling stations are displayed on the same distribution map

caused the increase in nitrate concentrations in stream water. Kaneyasu et al. [\(1999\)](#page-6-0), based on results from several widely separated sampling stations in the Kanto region, reported that nitrate is added to the soil in the form of photochemical smog. These reports suggest that atmospheric deposition plays an important role in the nitrate concentration of the stream water in the Kanto region. High nitrate concentration is not simply a problem in mountain streams alone, it also causes eutrophication in the downstream wetlands. There is a possibility that not only fertilizers but also atmospheric deposition are the main factors inducing eutrophication.

Conclusion

When river water quality data taken in the 1950s was compared with that from the 2000s, both chloride and

nitrate concentrations were found to have increased throughout Japan. Chloride concentration has increased greatly in urbanised areas. Nitrate data showed significant input of nitrate into river water in mountainous areas, presumably due to same process of atmospheric deposition from distant urbanised areas that has seen increasing quantities of atmospheric anthropogenic fixed nitrogen reported even in open ocean (Duce et al. 2008).

From the viewpoint of river management and the protection of river/ wetland water quality, we need to take account of both on-site effects (watershed effect) and off-site effects (including atmospheric deposition). Furthermore, lifestyle changes may modify the contaminant output of urbanised areas. To determine a more accurate relationship between watershed management and river water quality, analyses based on the GIS will be useful. Before applying this method we should consider the effects of changed lifestyles, especially when comparing past and present.

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