Fifty-year change in air pollution in Kaohsiung, Taiwan

Chiu-Hsuan Lee1 · Peter Brimblecombe1 · Chon-Lin Lee1,2,3,4

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Abstract
The change in air quality in cities can be the product of regulation and emissions. Regulations require enforcement of emission reduction, but it is often shifting economic and societal structures that influence pollutant emissions. This study examines the long-term record of air pollutants in Kaohsiung, where post-war industrialisation increased pollution substantially, although improvements are observed in recent decades as the city moved to a more mixed economy. The study tracks both gases and particles across a period of significant change in pollution sources in the city. Concentrations of SO2 and aerosol SO4 2− were especially high ~1970, but these gradually declined, although SO4 2− to a lesser extent than its precursor, SO2. While twenty-first century emissions of SO2 and NOx have declined, this has been less so for NH3, because it arises from predominantly agricultural sources. The atmosphere in Kaohsiung continues to have high concentrations of O3, and these have risen in the city, likely a product of less titration by NO. The changes have meant that ozone has become an increasing threat to health and agriculture. Despite a potential for producing (NH4)2SO4 and NH4NO3 aerosols, a product of a relatively constant supply of NH3, visibility has improved in recent years. Emissions of SO2 and NOx should continue to be reduced, as these strongly affect the amount of fine secondary aerosol. However, the key problem may be ozone, which is difficult to control as it requires careful consideration of the balance of NOx and hydrocarbons so important to its production.

Keywords Aerosols · Economic change · Health effects · Agricultural effects · Visibility

Introduction

Air quality in many cities has improved in line with changes in their economy and the regulation of emissions. Declining concentrations of air pollutants, most notably SO2, but later NOx, can be ascribed to changes in industries and their control, and a more modern vehicle fleet (Brimblecombe 2005; Power and Worsley 2018). However, the link between emission control and reduced air pollutant concentrations is weakened because of a mediating atmospheric chemistry, best characterised by O3 formation. Its production is affected by hydrocarbons and nitrogen oxides. Ozone concentrations can increase when nitrogen oxide emissions decrease, so air pollution regulation needs to go beyond simple emission control and requires the application of air quality management (Elsom 1992). Particulate matter is an important contributor to air pollution, yet a significant fraction of the urban aerosol is again produced through reactions of primary pollutants in the atmosphere that lead to both inorganic components (Ravishankara 1997), such as sulphates and nitrates. Secondary sulphate aerosol was probably more abundant in the past when SO2 levels were high. Secondary
organic components were best represented by the carboxylic acids, perhaps most notably low volatility dicarboxylic acids (Kawamura et al. 2001).

Taiwan (Fig. 1) experienced a rapid transformation from the large agrarian colony left by the Japanese after World War 2, when it saw innovative expansion, such as that in the semiconductor industry. Taiwan’s industrial growth saw per-capita gross domestic product in US dollar increase from $397 to $8200 by 1990 and just over $28,000 by 2020. Kaohsiung could share in this as during the colonial period, its harbour became a focus for shipping and rail transport that allowed the city’s development as a major hub for Taiwan’s south, with an industrial base in steel, cement, petrochemicals, paper making, etc. The Taiwan Economic Miracle (Tsai 1999) saw rapid growth of industrial infrastructure (~1960–1990), which contributed to pollutant emissions that, in the early pre-regulated stages, paralleled the strengthening economy. Today, Kaohsiung remains an industrial city, though with an increasing shift in the local economy (Fig. 1c) towards financial services, tourism, and the arts, with plans for the waterfront to become a landscape resource (KEC 2021).

Post-war industrialisation led to regulations needed to improve air quality, initially under the Air Pollution Control Act of 1975. The relaxation of Martial Law (late 1980s) saw newly democratised systems, and though electoral politics can stifle environmental debate, Taiwan established a cabinet-level Environmental Protection Administration (TEPA) in 1987. However, the 1975 Act was only effective after 1992, when stricter rules were implemented (Tang 1993). Under democratisation, this “credit for the improvement has been given to the air emission fee program that was first implemented” in 1995 (Tang and Tang 2000). Before then, “the traditional command-and-control program and tax-allowance subsidy were the two major instruments used for air pollution control ...” (Shaw and Hung 2001). Emission standards were established for power facilities 1994-05-04, while 1995-07-01 saw the introduction of an air pollution control fee for SO$_x$ emissions, and subsequent regulations to reduce volatile organic emissions (see supplementary table in Chen et al. 2014).

Post-war Kaohsiung expanded with an urban population of 168,008 in 1947 to 1,512,798 in 2017, where there were 2,776,912 in the municipal area. In parallel, there was a growth of heavy industry (1976–1986) and a mature stage for the heavy chemical industry (1986–1996). Industrial zones were established in Fengshan (1974), Yongan, and Linyuan (1974–1975). Although the service industry has surpassed manufacturing in terms of value, steel, petrochemical, cement, shipbreaking, and processing, exports remain substantial, though characterised by pollution. Challenge 2008: National Development Key Project encouraged investments in infrastructure such as High Speed Rail, Kaohsiung MRT (Mass Rapid Transit), but with newer less polluting developments emerging from 2009: (i) biotechnology, (ii) tourism, (iii) green energy, (iv) medical care, (v) low intensity agriculture, and (vi) cultural creativity (KCG 2010).

The long history of industrial emissions has promoted many studies of air pollution in Kaohsiung, with deposit gauge measurements from the 1960s (Hsu and Wei 1971; Selya 1975; Wei 1966), and more modern measurements from the 1970s (Chow et al. 1983). In Europe and North

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**Fig. 1** a Regional and b local maps showing main places mentioned in the text within the Kaohsiung City special municipality and neighbouring Pingtung County, where ammonia was sampled. The large squares mark the Environmental Protection Administration sites mentioned in the text. Shading reveals the 2009 population density. c Financial capital for the various economic sectors in Kaohsiung
America, there are estimates of long-term air pollutant loads in urban air using modelling (Brimblecombe 1977), observations of smoke days (Davidson 1979) or pollutant deposition (Brimblecombe 1982), but fewer from Asia (Ishikawa and Hara 1997). Taiwan’s National Health Administration planned an optimal monitoring network for SO2 in Kaohsiung from 1975 to 1977 (Liang and Lee 1980). There is a long series of studies of health (Yang et al. 1998), visibility (Lee et al. 2005; Lee 2006), and PAHs (Lai et al. 2017). Early problems seem dominated by primary pollutants, but the situation with ozone has caused concern across recent years (Hung-Lung et al. 2007; Shiu et al. 2007; Tsai et al. 2012). Although there are also contemporary claims that for two decades, atmospheric visibility in Kaohsiung has worsened (Lee and Lai 2018); they are not supported by Maurer et al. (2019), who suggest 2000–2015 saw improvement in Kaohsiung, paralleling change elsewhere in Taiwan, but not fully explained by lower RH or PM10.

Ammonia has not often been measured in Taiwan, but it may be important, given the density of pigs and poultry (Cheng et al. 2011). Additionally, Hsieh and Chen (2010) measured NH3 at industrial parks in southern Taiwan at Neipu, Pingtung, and Pingnan over two consecutive days each (2003-09/2004-12), finding means of 90.4 ppb, 72.8 ppb, and 84.9 ppb, while at the National Pingtung University of Science and Technology campus dormitory and a bamboo grove near the village of Laopi in Pingtung County mixing ratios were 52.2 ppb and 4.6 ppb showing the significance in the region. Vehicles represent a potential source of NH3 in urban areas; e.g. in urban Guangzhou, vehicles produce 19% of the ammonia (Liu et al. 2014), although 2006 estimates across the Pearl River Delta suggest vehicles accounted for just 2.5% (Zheng et al. 2012).

This paper explores a 50-year history of air pollution in Kaohsiung to understand the changes and assess the relevance of shifts in the economy and regulatory activity. The city represents an interesting and somewhat isolated location compared to the Greater Taipei Area in the north. Additionally, some pollutants are transported across the Taiwan Strait from Mainland China, but that has limited impact on urban concentrations (Lai and Brimblecombe 2021). In Kaohsiung, there have been great changes as it moved from an unregulated industrial centre for manufacturing, steelmaking, oil refining, and shipbuilding to a place aspiring to be noted for international exhibitions, tourism, and the arts. This transition has required more stringent urban planning and concerns over air quality and visibility. Although coastal cities have been well studied, the difficulty of maintaining sites may limit measurement duration (e.g. Alastuey et al. 2004; Galindo et al. 2020), but the longer record used here means we can explore how urban transformation is reflected through a half century of development. We give special attention to changes in the threat to health, agricultural production, and visibility.

Method

Economic, air pollution, and meteorological data

The project used economic data from the Kaohsiung City Government, Department of Budget, Accounting and Statistics as plotted in Fig. 1c (KCG 2021; KCGDG 2021) and population data (DHR 2021). Energy use has grown since 1970 with about 4 TWh and 7.5 TWh from oil and coal to level values of around 700 TWh and 400 TWh for these fuels from 2000 (BP 2021). Gas has become more important and now amounts to 200 TWh (BP 2021). A network of sites (Taiwan Air Quality Monitoring Network, TAQMN) is maintained by the TEPA to measure air pollutants (https://airtwp.epa.gov.tw). In Kaohsiung, it began with sites at Sanmin, Fengshan, Fuxing, and Qianjin providing data, though incomplete, from 1984. Nanzih came 2 years later, and a widening network added observations from Qianzhen, Daliao, Renwu, Xiaogang, Meinong, Linyuan, Qiaotou, and Zuoying from 1993 onwards (Fig. 1). Early monitoring in Kaohsiung was weighted towards crowded industrial areas of the city, but became more widespread over time, with a site placed at Meinong, a Hakka farming community on the Laonong River, 40 km from the centre of Kaohsiung. Corrections to PM2.5 from 2014 adopted the USEPA Non-Federal Reference Method, which led to a reduction in average values (~25%), but has little effect on our work as we largely avoid using the fine particulate data. Emission estimates (https://teds.epa.gov.tw/) are tuned to meet the boundaries of current Kaohsiung City, now a county-sized special municipality (area ~2950 km2). These along with concentration and mixing ratios (c) are plotted in Fig. 2. Aerosol composition is less frequently measured in the region, and much has been done as a part of research projects, rather than regular monitoring, though the most relevant data is given in the supplement. Daily visibility data for Kaohsiung (WMO_ID:467440) were extracted as daily observations from the historical record (https://e-service.cwb.gov.tw/HistoryDataQuery/index.jsp) on the CODiS of Taiwan’s Central Weather Bureau. The records displayed in Fig. 2 come from a single source, so the methodology remains consistent, except for a change to the TEPA methodology for PM10 in 2010 as seen in emissions in Fig. 2(b). Additionally, there was a decade-long break to the ozone record for Fuxing.

The number of data points was often large, so we used parametric methods (e.g. Welch’s t-test), but where small and the distribution undefined, non-parametric techniques were preferred along with the median and quartile ranges.
The Wilcoxon signed-rank test (rather than a t-test) was used where the data set was small and occasionally Kendall \( \tau \) and Theil-Sen slopes were determined as these are more robust against outliers than a classical linear regression (Vannest et al. 2016).

**Results and discussion**

The record of pollutants PM\(_{10}\), PM\(_{2.5}\), SO\(_2\), NO\(_x\), NO\(_2\), and O\(_3\) as measured by TAQMN are shown in Fig. 2.

**Decadal change in primary pollutants**

Monthly average pollutant concentrations and mixing ratios from the TAQMN site in Kaohsiung and estimated emissions for the region 2002–2020 are shown in Fig. 2. There are distinct annual cycles to the primary pollutants (Fig. 2(a, c, e, g)), higher values occurring each winter (Lee et al. 2018; Tsai et al. 2013); seasonal cycles 2000–2020 appear as insets. Trends across this period suggest continuous improvement to the primary pollutants such as NO\(_x\) and PM\(_{10}\) (\( \tau = -0.40, p < .0001 \) and \( \tau = -0.23, p < .0001 \)), and notably for SO\(_2\) (\( \tau = -0.62, p < .0000 \)). Especially low SO\(_2\) values are evident at the rural Meinong site (Fig. 2(c)). There is evidence of a weaker mid-cycle in annual cycle for NO\(_x\) in rural areas (Fig. 2(e)). The SO\(_2\) mixing ratios were especially high before 1994, when measurements were made at crowded urban locations: Sanmin, Fengshan, Fuixing, Qianjin, and Nanzih. The mixing ratios typically continued to be higher than at other sites that entered the record after 1994. However,
even at these crowded sites, levels declined over time, continuing improvement perhaps a result of sulphur emission fees beginning in 1995.

The mixing ratios of CO, NOx, and less clearly PM10 rose at first, but these decreased from the early 1990s, in a way typical of the changing pollutant levels during the historic development of cities (Brimblecombe 1977). By contrast, oxidants have increased (Chen et al. 2014), with O3 mixing ratios on the rise (Fig. 2(i)). In the eastern parts of Kaohsiung, toluene from paint and solvent industries plays an important role in O3 production as in inland areas production is often limited by the NMHCs, i.e. volatile organic compounds (Hung-Lung et al. 2007). Ozone production in the air aloft, often reflecting long-range transport, can be NOx limited (Hung-Lung et al. 2007). The seasonal cycle of ozone, with a bimodal structure, is more complex than the primary pollutants (inset of Fig. 2(i)).

Estimated emissions from the Kaohsiung area for a range of pollutants from 2002 onward (Fig. 2(b, d, f, h, j)) reflect emission reduction policies (Chen et al. 2014). Emission inventories are error prone, with a factor of two errors possible for NOx and hydrocarbons and an even larger three-fold error found for CO and particulate matter (Smit et al. 2010), but when a consistent methodology is applied year by year trends can nevertheless be clear. However, the sudden change in PM10 in 2010 relates to an altered assessment methodology for industrial emissions, adopted by the TEPA. Some 24 kt a⁻¹ ammonia was emitted from poultry farms in Taiwan (Cheng et al. 2011), which makes Kaohsiung’s estimated emissions substantial at 18.74 kt a⁻¹ in 2002.

The decline in emissions appears to be smaller than that for concentration. This anomaly may arise because most pollutant concentration measurements are made in the built up and increasing residential area of Kaohsiung, while the emissions are for the county-sized special municipality.

Particulate matter has been measured for many years. Selya (1975) listed the 2-year average for total suspended matter as 371 μg m⁻³ and SO₄²⁻ at 21.4 μg m⁻³. Although this early SO₄²⁻ concentration is high, it seems compatible with later measurements for 1994/1995, 11.5 μg m⁻³ (Yang et al. 1998), and 1998/1999, 14.34±5.10 μg m⁻³ (Lin 2002), as tabulated in the supplement. It is supported by the trends in SOx mixing ratios in the early part of the record (Fig. 2(c)) and the suggestions of high levels from Chow et al. (1983).

Figure 3(a) shows the mole ratio of nitrogen to sulphur oxides in the gas phase (i.e. $n_{NOx}/n_{SO2}$ as points and a shaded interquartile range). Measurements from the late 1960s (Selya 1975) would suggest that in the particulate phase, $n_{NO3}/n_{SO4}$ (~0.24 in the 1960s) was lower than that at present in a sulphur-dominated atmosphere with uncontrolled industrial use of soft coal, a cheap fuel widely used in factories, hotels, dwellings, schools, etc. Such low values could have reflected large amounts of sulphate present in coarse fly ash. From the late 1960s, soft coal was banned in Taipei, so some entrepreneurs in Kaohsiung may have for a short time increased its use (Selya 1975). Such changes are attributed to the shift from coal to petroleum, and in Kaohsiung are reflected in the change in $n_{NOx}/n_{SO2}$ from 1.5 in the 1980s to 4.5 over the last decade.

Overall, these observations of long-term change in air pollutants in Kaohsiung show a pattern among primary pollutants similar to other cities along with social change that has shifted fuel use (e.g. London in Brimblecombe 2006). Pressure for regulation in Taiwan led to reductions in mixing ratios of SO₂ first, with NOx and PM seeming to reach a maximum in the last decade of the twentieth century.
However, as substantial as many of the improvements have been, the changing economy of the city has made a significant contribution to reductions. The NO$_x$/$SO_2$ ratio was probably low in the 1970s and grew after that. The NO$_x$/$SO_2$ ratio in the atmosphere of Kaohsiung has increased, which follows early reduction of sulphur emissions, and enhanced by NO$_x$ from a growing vehicle fleet that has been difficult to keep in check. In Taiwan, vehicle registrations are increasing at 0.24 million a year (https://tradingeconomics.com/taiwan/car-registrations), but there are additionally 0.9 million polluting motor-scooters (Everington 2018). Despite this, the overall mixing ratios on NO$_x$ have declined in line with emissions (Fig. 2(e, f)) suggesting some regulatory success in responding to an enlarged automobile fleet, i.e. private vehicles from 1994, 432,228, to 2020, 763,975 (KCGDG 2021).

**Change in secondary pollutants**

The changing volatile organic components as non-methane hydrocarbons (Fig. 2(j)) and the increasing dominance of NO$_x$ (Fig. 3(a)) encourage the formation of secondary pollutants. Toluene has been shown to be particularly relevant to the formation of ozone (Hung-Lung et al. 2007), although Kuo et al. (2015) indicated that VOC was not significantly correlated with ozone variability in the few episodes studied in Kaohsiung. From 1994 to 2003, Shui et al. 227 (2007) found that the mixing ratios of NO$_2$ in southern Taiwan decreased while those of ozone increased, which could be accounted for by (i) the reduction in NO$_2$, due to lower NO titration, or (ii) the more reactive precursor NMHCs (Chang et al. 2005). Figure 3(b) shows the increasing fraction ($f_{NO_2}$= NO$_2$/NO$_x$) of NO$_x$ present as NO$_2$ at urban Fuxing and in rural Meinong. Despite being in an urban area, Fuxing has become increasingly less industrial over the decades, now focussed on commercial and residential activities. Over time, decreasing amounts of NO$_x$ have allowed the available O$_3$ to oxidise larger fractions of NO to NO$_2$. A quarter century back, the 5-year average O$_3$ at the urban site of Xiaogang was 19.9±6.9 ppb (1993-08/1998-07), but much higher at rural Meinong 29.4±6.6 ppb. More recently (2016-01/2020-12), the differences had narrowed to 26.4±7.7 ppb and 27.4±6.2 ppb. The increases in urban areas are typical of Southern China where titration of O$_3$ by NO has decreased with declining emissions of NO$_x$ (Li et al. 2022a).

These changes are likely accompanied by the formation of secondary inorganic aerosols that have been easy to trace in Hong Kong as the record of aerosol is detailed since 1995 (Brimblecombe 2022). The record of aerosol composition is less complete in Kaohsiung and fails to reveal a satisfying and coherent picture of change (see supplement Fig. S2), although it is likely that in the 1970s the sulphate was high. The Kaohsiung special municipality is agricultural, so the hinterland provides NH$_3$ to neutralise acidity and these emissions have changed only a little over time (Fig. 2(h)). Aerosol NH$_4^+$ is probably insensitive to the total NH$_3$, but highly sensitive to total H$_2$SO$_4$ and HNO$_3$ (Cheng and Wang-Li 2019). Nevertheless, the special municipality has not been able to greatly reduce its agricultural NH$_3$, but it is probably more critical to ensure that emissions of SO$_2$ and NO$_x$ continue to be reduced as these strongly affect the amount of fine secondary aerosol.

Chemical transformations mean that particulate SO$_2^{2−}$ and NO$_x^{+}$ concentrations might not necessarily follow regulatory improvements to their precursors. However, in Kaohsiung, it is likely that over longer timescales, particulate sulphate has declined in parallel with SO$_2$. The Theil-Sen slope for the medians of the SO$_2^{2−}$ suggests a decline of ~0.3 μg m$^{-3}$ a$^{-1}$ from 1970, which would accumulate to ~80% over time. Since the mid-1990s, it decreased from 11.5–14.3 μg m$^{-3}$ (Lin 2002; Yang et al. 1998) to 3.9–4.4 μg m$^{-3}$ at present (Shen et al. 2020), i.e. ~65% decline. This is proportionally less than the decrease in SO$_2$ from ~25 μg m$^{-3}$ in the 1990s to ~3 μg m$^{-3}$ at present (~90% decrease).

**Changing health risk**

Air pollution poses both long- and short-term health risks. The risk of daily hospital admissions due to air pollution can be calculated based on exposure to pollutants, and here we adopted the method used to calculate the Air Quality Health Index of Hong Kong (GovHK, 2014). The risk is determined as the sum of percentage added health risk ($R_{AHR}$) for daily hospital admissions attributable to the 3-h moving average mixing ratios of NO$_2$, SO$_2$, O$_3$, and particulate matter (here taken as PM$_{10}$). These risk factors were derived from health statistics and air pollution data from Hong Kong and are therefore not exact for Kaohsiung, but given similar population activity and climate, there should be a reasonable proportionality. The $R_{AHR,i}$ for each pollutant $i$, as

$$R_{AHR,i} = 100 \left[ \exp \left( \beta_i c_i \right) - 1 \right]$$  
and $c_i$ is the 3-h moving average concentration of pollutants (μg m$^{-3}$), with the factors $\beta_{SO_2} = 0.0004462559$, $\beta_{SO_2} = 0.0001393235$, $\beta_{O_3} = 0.0005116328$, and $\beta_{PM_{10}} = 0.0002821751$ (Wong et al. 2012). Although it would make more sense to use PM$_{2.5}$ in these calculations, PM$_{10}$ was used as the record was more complete, but PM$_{10}$ can provide a reasonable estimate of the health risk, and it includes PM$_{2.5}$ (Brimblecombe 2021).

Figure 4 shows the added daily health risk averaged for each month at (a) Fuxing, (b) Xiaogang, and (c) Meinong. The risk is much higher at the urban site in Xiaogang, but declines over time, in much the same way as the risk in rural Meinong. The risk from PM$_{10}$ is relatively constant across the sites reflecting the broad distribution that arises from...
a multiplicity of sources. The balance of risk arises differently at the sites, so at Meinong a larger proportion comes from O₃, while the effect of SO₂ on health risk is very much lower compared with Fuxing, especially in the earlier years at this site. The proportions are clearer in the ternary plot of Fig. 4(d), which shows monthly risk across the 5-year period 2016–2020. It reveals the contemporary situation where the urban sites of Fuxing and Xiaogang are distinct from that in Meinong. The time trends for the relative risk over years 1993–2020 are shown in the ternary diagram of Fig. 4(e). This illustrates the transition to lower risk from particulate matter, but a greater proportion of risk that arises from O₃, especially at the rural site, but the change has also been evident in the urban areas. Secondary pollutants are more difficult to control, separated as they are from their sources through a mediating chemistry. This difficulty was recognised with the discovery of photochemical smog 70 years ago (Brimblecombe 2014), and stresses a continued need for management of air quality that can address secondary pollutants.

**Agriculture**

Agricultural crops are sensitive to O₃ (Fuhrer et al. 1997), so this places pressure on food security (Wang et al. 2017). The hinterland to Kaohsiung is important in the production of a range of crops (ABKMG 2021): fruit (banana 57 900 t a⁻¹, guava 71 468 t a⁻¹, and pineapple 56 971 t a⁻¹) and vegetables (bamboo shoots 20 497 t a⁻¹, green soybeans 19 665 t a⁻¹, tomatoes 12 902 t a⁻¹, and radishes 11 496 t a⁻¹).

As crops accumulate damage over time, it is common to express risk to vegetation as AOT₄₀ (Accumulated Ozone exposure over a threshold of 40 ppb during the day as ppb h). Summation is usually made over daylight hours during the crop’s growing season, although it is sometimes reported for each month. In Europe, the target value is 9000 ppb h considered over 5 years. The long-term objective is 3000 ppb h. In Europe, the growing season is typically May to July. Since Taiwan has a tropical climate, the growing season is more difficult to define because crops are grown all year round. Summer days are also not particularly long, so daylight hours are considered shorter in our calculations: 07:00 to 17:00. There are two periods (Fig. 2(i)) of high O₃ levels, March to May and September to November (Chen et al. 2004). The 3-month long AOT₄₀ for the two urban sites and the rural site at Meinong is shown in Fig. 5(a, b) for each O₃ season. The mixing ratios of O₃ increased over the late parts of the twentieth century, but it is somewhat uncertain because the records are difficult to overlap for cross-checking. We can see that Meinong is typically the highest, and the Wilcoxon signed-rank test shows it to be higher (p<.0001) than both Xiaogang (1994 to 2020) and Fuxing (2004 to 2020). Despite increases in O₃ in general in the Kaohsiung area (Fig. 2(i)), there are hints the AOT₄₀ is in decline particularly late in the year. The number of hours each year where late season O₃ exceeds 40 ppb is plotted in Fig. 5(c), which suggests that although AOT₄₀ might be decreasing, the number of hours above 40 ppb is relatively stable over recent years. The decline in AOT₄₀ is mostly caused by a decrease in hours with high O₃ (i.e. >80 ppb). These have declined recently especially in the late part of the year, but since 2001, in both ozone seasons, high O₃ periods have become less common.

There are only a few studies of O₃ and crop damage in Taiwan (e.g. Sheu and Liu 2003). In addition, there are few studies on major crops found in the Kaohsiung region,
except for some studies on soybeans and tomatoes. Soybeans show visual damage after taking up several thousand ppb h over the entire growing season (Gosselin et al. 2020), conditions that were exceeded at the Meinong site (Fig. 5(a, b)). Over a period of weeks, tomatoes (Lycopersicon esculentum Mill. H-11) exposed to O₃ at 200 and 350 ppb for 2.5 h at 3 days a week showed extensive foliar injury, defoliation, and reduction in biomass, although fruit yield was only lower at the higher mixing ratio (Oshima et al. 1975). However, such high values are not experienced at Meinong, and even an hourly mixing ratio >150 ppb is found less than 40 times since 1993. Nevertheless, most years exceed the European guideline value of 9000 ppb h, especially during the September to November period. The long-term objective 3000 ppb h is always exceeded. However, the experiments of Reinert et al. (1997) show that the Tiny Tim cherry tomato (L. esculentum L. cv. Tiny Tim) shows a 20% reduction in vegetative dry weight after 13 weeks exposure to just 80 ppb.

In Kaohsiung and the surrounding region, concentrations of O₃ have remained high for the last quarter century. This represents both a risk to health and a threat to agriculture, so should be a matter of continued regulatory concern.

Visibility

Visibility is an important issue in the region. It is a publicly perceptible marker of changes in air pollution over long periods (Brimblecombe 2021), but studies from southern Taiwan do not always use the most recent data (Lee and Lai 2018). Maurer et al. (2019) were able to use the record up to 2016, which hints at the influence of PM_{10} on visibility and supports the notion that it has generally improved in Taiwan over recent decades. Yuan et al. (2006) suggest an empirical equation for the light scattering coefficient, $b_{sp}/\text{km}^{-1}$ as:
where $c$ is the concentration of aerosol components and $c_0$ a remainder term. Yang et al. (2005) determined the amount of $(NH_4)_2SO_4$ and $NH_4NO_3$ assuming that all $SO_4^{2-}$ and $NO_3^-$ was present as the ammonium salt, which requires that $NH_4^+$ be in excess, a reasonable assumption in Kaohsiung, and increasingly so given the decline in $SO_2$ and $NO_x$. However, as Li et al. (2022b) show for Beijing, the secondary aerosol can make a contribution to visibility that can outweigh the effects of primary emissions. The calculated visibility from aerosol measurements listed in the supplementary materials can be compared with observed visibility in Kaohsiung (Fig. 6), although such improvement might not be large enough to be obvious to the general population.

### Conclusion

This study has revealed long-term reductions in air pollution as a city transformed from an industrial base to a broader economy, with industry increasingly located around sites such as Xiaogang. On a day-to-day basis, mixing ratios of precursors and secondary pollutants might not correlate well, but it is likely that the effects of pollution chemistry and meteorology are smoothed out over the years, so primary and secondary pollutants seem to follow similar patterns. However, the response is non-linear, so the reduction in secondary $SO_4^{2-}$ in Kaohsiung has not been as large as the reduction in $SO_2$ over the last decade. In parallel, the N/S ratio has increased with the decline in sulphur emissions. Air pollution concentrations in Kaohsiung have declined to a greater extent than the reduction in emissions. Both regulations and economic changes have enabled improvements in air quality in recent decades, yet $O_3$ remains a problem. This secondary pollutant is difficult to control as it requires careful consideration of the balance of $NO_x$ and hydrocarbons, especially as the NMHC emissions are no longer in sharp decline.

Future work could compare primary and secondary pollutant concentrations with emissions via modelling, although it may be difficult to collect data for spatially resolved emissions over long time periods. However, economic records could reveal fuel imports and farming statistics which would suggest the magnitude and distribution of emissions. Changes in secondary organic compounds were neglected in this study, although this would be an interesting topic for further research. Evaluating the impact of an emission reduction on change in visibility or health effects is important for formulating regulatory policy, and while modelling is available to link emissions to concentrations, nonlinear effects on exposure or health outcomes can be more difficult to represent.

### Supplementary Information

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### Author contribution

Conceptualisation of the investigation (CLL, PB), methodology (PB), supervision (CLL), undertaking investigation (CHL), preparing data (CHL), statistical analysis (CHL, PB), original draft (CHL, PB), figures (PB), and final editing (PB). All authors have read and agreed to the published version of the manuscript.

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### Data availability

The data is publicly available as denoted by URLs in the text.

### Declarations

**Ethical approval** No human or animal subjects were used in this research.

**Consent to participate** All authors agreed with being involved in the research project.

**Consent to publish** All authors agreed with the content and that all gave explicit consent to submit and that they obtained consent from the responsible authorities at the institute/organisation where the work has been carried out, before the work was submitted.

**Competing interests** The authors declare no competing interests.

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### References

ABKMG (2021) Agricultural Production (title in Chinese). Agricultural Bureau of Kaohsiung Municipal Government. https://agri.kcg.gov.tw/AgriculturalService/Agriculture/Produce.htm. Accessed 2 July 2022

Alastuey A, Querol X, Rodriguez S, Plana F, Lopez-Soler A, Ruiz C, Mantilla E (2004) Monitoring of atmospheric particulate matter around sources of secondary inorganic aerosol. Atmos Environ 38(30):4979–4992

BP (2021) BP’s Statistical Review of World Energy 2021. Pureprint Group Limited, London

Brimblecombe P (1977) London air pollution, 1500–1900. Atmos Environ 11(12):1157–1162

Brimblecombe P (1982) Trends in the deposition of sulphate and total solids in London. Sci Total Environ 22(2):97–103
Brimblecombe P (2005) The globalization of local air pollution. Globalizations 2(3):429–441
Brimblecombe P (2006) The Clean Air Act after 50 years. Weather 61(11):311–314
Brimblecombe P (2014) Deciphering the chemistry of Los Angeles smog, 1945–1995. In: In Toxic Airs: Body, Place, Planet in Historical Perspective. University of Pittsburgh, Pittsburgh, pp 95–108
Brimblecombe P (2021) Visibility driven perception and regulation of air pollution in Hong Kong, 1968–2020. Environments 8(6):51
Brimblecombe P (2022) Trends in secondary inorganic particles in Hong Kong, 1995–2020. Atmos Environ 268:118801
Chang CC, Sree U, Lin YS, Lo JG (2005) An examination of 7:00–9:00 PM ambient air volatile organics in different seasons of Kaohsiung city, southern Taiwan. Atmos Environ 39(5):867–884
Chen KS, Lin CF, Chou YM (2001) Determination of source contributions to ambient PM2.5 in Kaohsiung, Taiwan, using a receptor model. J Air Waste Manage Assoc 51(4):489–498
Chen KS, Ho YT, Lai CH, Tsai YA, Chen SJ (2004) Trends in concentration of ground-level ozone and meteorological conditions during high ozone episodes in the Kaoping Airshed, Taiwan. J Air Waste Manag Assoc 54(1):36–48
Chen SP, Chang CC, Liu JJ, Chou CCK, Chang JS, Wang JI (2014) Recent improvement in air quality as evidenced by the island-wide monitoring network in Taiwan. Atmos Environ 96:70–77
Cheng B, Wang-Li L (2019) Responses of secondary inorganic PM2.5 to precursor gases in an ammonia abundant area in North Carolina. Aerosol Air Qual Res 19(5):1126–1138
Cheng WH, Chou MS, Tung SC (2011) Gaseous ammonia emission from poultry facilities in Taiwan. Environ Eng Sci 28(4):283–289
Chow JC, Watson JG, Chaung CY (1983) Air pollution in the Republic of China (Taiwan). J Air Pollut Control Assoc 33(8):768–770
Davidson CI (1979) Air pollution in Pittsburgh: a historical perspective. J Air Pollut Control Assoc 29(10):1035–1041
DHR (2021) Dept. of Household Registration. Ministry of the Interior, Taiwan. https://www.ris.gov.tw/appen. Accessed 2 July 2022
Elsom DM (1992) Atmospheric pollution: a global problem, 2nd edn. DHR (2021) Dept. of Household Registration. Ministry of the Interior, Taiwan. https://www.ris.gov.tw/appen. Accessed 2 July 2022
Elsom DM (1992) Atmospheric pollution: a global problem, 2nd edn. DHR (2021) Dept. of Household Registration. Ministry of the Interior, Taiwan. https://www.ris.gov.tw/appen. Accessed 2 July 2022
Everington K (2018) Taiwan to ban gasoline-powered scooters in 2035, Taiwan News (2018/11/05). https://www.taiwannews.com.tw/en/news/3568172. Accessed 2 July 2022
Fuhrer J, Skirby L, Ashmore MR (1997) Critical levels for ozone effects on vegetation in Europe. Environ Pollut 97(1-2):91–106
Galindo N, Yubero E, Clemente Á, Nicolás JF, Varea M, Crespo J (2014) Deciphering the chemistry of Los Angeles aldehydes and inorganic species in Los Angeles. Atmos Environ 35(23):3917–3926
KCG (2010) The current situation and challenges of the development of Kaohsiung’s cities (in Chinese). https://orgws.kcg.gov.tw/001/KcgOrgUploadFiles/334/relfile/id/69818/33333bf1-b27e-4a50-b5dd-c937bf20905.pdf. Accessed 24 Dec 2021
KCG (2021) Survey of Budget Department of Budget, Kaohsiung City Government, Accounting and Statistics. https://kas.kcg.gov.tw/. Accessed 2 July 2022
KCGDG (2021) Statistical Information Network of Kaohsiung City, Kaohsiung City Government. https://kcgdg.kcg.gov.tw/kcgstat/page/default.aspx. Accessed 22 July 2021
KEC (2021) Economy, industries & development, Kaohsiung Exhibition Center. http://www.kecc.com.tw/cityEconomy.asp. Accessed 22 Dec 2021
Kuo YM, Chiu CH, Yu HL (2015) Influences of ambient air pollutants and meteorological conditions on ozone variations in Kaohsiung, Taiwan. Stoch Env Res Risk A 29(3):1037–1050
Lai YC, Tsai CH, Chen YL, Chang-Chien GP (2017) Distribution and sources of atmospheric polycyclic aromatic hydrocarbons at an industrial region in Kaohsiung, Taiwan. Aerosol Air Qual Res 17(3):776–787
Lee CG (2006). Study of the effect of aerosol characteristics and meteorological parameters on visibility in Urban kaohsiung. Thesis, Environmental Engineering, National Sun-Yat Sen University.
Lee CG, Lai WL (2018) Effects of different factors on the visibility in Kaohsiung Area using hierarchical regression. In: In International Conference on Genetic and Evolutionary Computing. Springer, Singapore, pp 361–371
Lee CG, Yuan CS, Chang JC, Yuan C (2005) Effects of aerosol species on atmospheric visibility in Kaohsiung city, Taiwan. J Air Waste Manage Assoc 55(7):1031–1041
Lee CS, Chang KH, Kim H (2018) Long-term (2005–2015) trend analysis of PM 2.5 precursor gas NO2 and SO2 concentrations in Taiwan. Environ Sci Pollut Res 25(22):22136–22152
Li XB, Yuan B, Parrish DD, Chen D, Song Y, Yang S, Liu Z, Shao M (2022a) Long-term trend of ozone in southern China reveals future mitigation strategy for air pollution. Atmos Environ 269:118869
Li Z, Sun Y, Wang Q, Xin J, Sun J, Lei L, Li J, Fu P, Wang Z (2022b) Nitrate and secondary organic aerosol dominated particle light extinction in Beijing due to clean air action. Atmos Environ 269:118833
Liang WL, Lee CT (1980) The optimization evaluation method of SO2 air pollution and monitor network in Kaohsiung area. J Chin Inst Eng 3(2):105–115
Lin JJ (2002) Characterization of the major chemical species in PM2.5 in the Kaohsiung City, Taiwan. Atmos Environ 36(12):1911–1920
Liu T, Wang X, Wang B, Ding X, Deng W, Liu S, Zhang Y (2014) Emission factor of ammonia (NH3) from on-road vehicles in China: tunnel tests in urban Guangzhou. Environ Res Lett 9(6):064027
Maurer M, Klemm O, Lokys HL, Lin NH (2019) Trends of fog and meteorological parameters on visibility in Urban Kaohsiung. Thesis, Environmental Engineering, National Sun-Yat Sen University.
Maurer M, Klemm O, Lokys HL, Lin NH (2019) Trends of fog and visibility in Taiwan: climate change or air quality improvement? Aerosol Air Qual Res 19(4):896–910
Oshima RJ, Taylor OC, Braegelmann PK, Baldwin DW (1975) Effect of ozone on the yield and plant biomass of a commercial variety of tomato. Am Soc Agron Crop Sci Soc Am Soil Sci Soc Am 4(4):463–464
Power A, Worsley A (2018) Historical urban pollution. In: Charlesworth SM, Colon A (eds) Booth Urban Pollution: Science and Management. Wiley, Oxford, pp 7–27
Ravishankara AR (1997) Heterogeneous and multiphase chemistry in the troposphere. Science 276(5315):1058–1065
Reinert RA, Eason G, Barton J (1997) Growth and fruiting of tomato as influenced by elevated carbon dioxide and ozone. New Phytol 137(3):411–420
Selya RM (1975) Water and air pollution in Taiwan. J Dev Areas 9(2):177–202
Shaw D, Hung MF (2001) Evolution and evaluation of air pollution control policy in Taiwan. Environ Econ Policy Stud 4(3):141–166
Shen H, Cheng PH, Yuan CS, Yang ZM, Ie IR (2020) Chemical characteristics, spatiotemporal distribution, and source apportionment of PM2.5 surrounding industrial complexes in Southern Kaohsiung. Aerosol Air Qual Res 20(3):557–575
Shen BH, Liu CP (2003) Air pollution impacts on vegetation in Taiwan. Air Pollut Impacts Crops For-a Global Assess:145–163
Shia CJ, Liu SC, Chang CC, Chen JP, Chou CC, Lin CY, Young CY (2007) Photochemical production of ozone and control strategy for Southern Taiwan. Atmos Environ 41(40):9324–9340
Smit R, Ntziachristos L, Boulter P (2010) Validation of road vehicle and traffic emission models–a review and meta-analysis. Atmos Environ 44(25):2943–2953
Tang DTC (1993) The environmental laws and policies of Taiwan: A comparative law perspective. Pac Rim Law Policy J 3:89
Tang CP, Tang SY (2000) Democratizing bureaucracy: the political economy of environmental impact assessment and air pollution prevention fees in Taiwan. Comp Polit:81–99
Tsai PL (1999) Explaining Taiwan’s economic miracle: are the revisionists right? Agenda: J Policy Anal Reform 6:69–82
Tsai HH, Liu YF, Yuan CS, Chen WH, Lin YC, Hung CH, Jen YH, Ie IR, Yang HY (2012) Vertical profile and spatial distribution of ozone and its precursors at the inland and offshore of an industrial city. Aerosol Air Qual Res 12(5):911–922
Tsai JH, Chang LP, Chiang HL (2013) Size mass distribution of water-soluble ionic species and gas conversion to sulfate and nitrate in particulate matter in southern Taiwan. Environ Sci Pollut Res 20(7):4587–4602
Vannest KJ, Parker RI, Gonen O, Adiguzel T (2016) Single case research: web based calculators for SCR analysis. (Version 2.0) [Web-based application]. College station: Texas A&M University. http://www.singlecaseresearch.org/calculators/theil-sen. Accessed 29 June 2021
Wang T, Xue L, Brimblecombe P, Lam YF, Li L, Zhang L (2017) Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects. Sci Total Environ 575:1582–1596
Wei W-H (1966) Air pollution control on Taiwan. Environmental Sanitation Bureau, Taipei
Wong TW, Tam WWS, Lau AKH, Ng SKW, Yu ITS, Wong AHS, Yeung DA (2012) Study of the air pollution index reporting system, report to the air services group; Tender Ref. AP 07-085. The Environmental Protection Department of HKSAR, Kowloon
Yang CY, Wang JD, Chan CC, Hwang JS, Chen PC (1998) Respiratory symptoms of primary school children living in a petrochemical polluted area in Taiwan. Pediatr Pulmonol 25(5):299–303
Yang H, Yu JZ, Ho SSH, Xu J, Wu WS, Wan CH, Wang X, Wang X, Wang L (2005) The chemical composition of inorganic and carbonaceous materials in PM2.5 in Nanjing, China. Atmos Environ 39(20):3735–3749
Yuan CS, Lee CG, Liu SH, Chang JC, Yuan C, Yang HY (2006) Correlation of atmospheric visibility with chemical composition of Kaohsiung aerosols. Atmos Res 82(3-4):663–679
Zheng JY, Yin SS, Kang DW, Che WW, Zhong LJ (2012) Development and uncertainty analysis of a high-resolution NH3 emissions inventory and its implications with precipitation over the Pearl River Delta region, China. Atmos Chem Physics 12(15):7041–7058

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