Fifty-Year Change in Air Pollution in Kaohsiung, Taiwan

Chiu-Hsuan Lee

National Sun Yat-sen University College of Marine Sciences

Peter Brimblecombe (✉ p.brimblecombe@uea.ac.uk)

National Sun Yat-sen University College of Marine Sciences <https://orcid.org/0000-0002-2233-8761>

Chon-Lin Lee

National Sun Yat-sen University College of Marine Sciences

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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 a 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 dramatically, 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 SO_2 and aerosol SO_4^{2-} were especially high ~1970, but these gradually declined, although the SO_4^{2-} to a lesser extent than its precursor, SO_2 . While 21st century emissions of SO_2 and NO_x have declined, this has been less so for NH_3 , because it arises from predominantly agricultural sources. The atmosphere in Kaohsiung continues to have high concentrations of O_3 , 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 the production of $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 aerosols, a product of a relatively constant supply of NH_3 , visibility has improved in recent years. The emissions of SO_2 and NO_x 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 needs careful consideration of the balance of NO_x and hydrocarbons so important to its production.

Highlights

- Kaohsiung emissions and primary pollutants decline, but NO_x/SO_2 and O_3 increases
- Air quality change reflects regulation and a move from an industrial to mixed economy
- The increasing O_3 threatens on health and agriculture
- Visibility has improved despite agricultural contributions to $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3

1. 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 SO_2 , but later NO_x can be ascribed to changes in industries and their control, and a more modern vehicle fleet (Brimblecombe, 2005; Power & Worsley, 2018). However, the link between emission control and reduced air pollutant concentrations is weakened because of a mediating atmospheric chemistry, best characterised by O_3 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 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 sulfates and nitrates. Secondary sulfate aerosol was probably more

abundant in the past when SO₂ levels were high. Secondary organic components 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 largely agrarian colony left by the Japanese after World War 2, when it saw innovative expansion, such as that in the semiconductor industry. During the colonial period, Kaohsiung's 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 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).

Postwar 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 (1976-86) and a mature stage for the heavy chemical industry (1986-96). Industrial zones were established in Fengshan (1974), Yongan and Linyuan (1974-75). 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, 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 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 deposit (Brimblecombe, 1982), but fewer from Asia (Ishikawa & Hara, 1997). Taiwan's National Health Administration planned an optimal monitoring

network for SO₂ in Kaohsiung from 1975-1977 (Liang & 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 & 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 concentrations 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 fifty-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, where there have been great changes as it moved from an unregulated industrial centre for manufacturing, steel-making, 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.

2. Method

2.1 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 (DHR, 2021). A network of sites (Taiwan Air Quality Monitoring Network, TAQMN) is maintained by the Taiwan Environmental Protection Administration (TEPA) to measure air pollutants (<https://airtw.epa.gov.tw>). In Kaohsiung, it began with sites at Sanmin, Fengshan, Fuxing and Qianjin providing data, though incomplete, from 1984. Nanzih came two years later, and a widening network added observations from Qianzhen, Daliao, Renwu, Xiaogang, Meinong, Linyuan, Qiaotou and Zuoying, 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 $PM_{2.5}$ from 2014 adopted the USEPA Non-Federal Reference Method, which led to a reduction in average values ($\sim 25\%$), 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 $\sim 2950 \text{ km}^2$). These along with concentration (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 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 (CWB).

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 Theil Sen slopes were determined as these are more robust against outliers than a classical linear regression (Vannest et al., 2016).

3. Results

The record of pollutants PM_{10} , $PM_{2.5}$, SO_2 , NO_x , NO_2 and O_3 as measured by the TAQMN are shown in Fig. 2.

3.1 Decadal change in primary pollutants

Monthly average pollutant concentrations 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. 2a,c,e,g), higher values occurring each winter (Lee et al, 2018; Tsai et al., 2013); seasonal cycles 2000-2020 appear as insets. The trends suggest continuous improvement to the primary pollutants, and often, though most notably for SO_2 , low values are evident at the Meinong rural site (Fig. 2c). There are hints of a weaker annual cycle for rural NO_x , which can be noticed mid-cycle (Fig. 2e). The SO_2 concentrations were especially high before 1994, when measurements were made at crowded urban locations: Sanmin, Fengshan, Fuixing, Qianjin and Nanzih. The concentrations typically continued to be higher than at other sites added in the record beyond 1994. However, even at these crowded sites, levels declined over time; continuing improvement perhaps a result of charges for sulfur emissions from 1995.

Although concentrations of CO , NO_x and non-methane hydrocarbons (NMHCs) decreased 1995-2012, oxidants have increased (Chen et al., 2014). Ozone concentrations have risen (Fig. 2i), although the precursor NMHCs declined from ~ 2002 (Fig. 2j), but this would be easily understood if Kaohsiung's hydrocarbons (Chang et al., 2005) were more reactive. In the eastern parts of Kaohsiung toluene from paint and solvent industries plays an important role in O_3 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 NO_x limited (Hung-Lung et al., 2007). The seasonal cycle of ozone, with a bimodal structure, is more complex than the primary pollutants (inset to Fig. 2i).

Estimated emissions from the Kaohsiung area for a range of pollutants from 2002 onward (Fig. 2b,d,f,h,j) reflect emission reduction policies (Chen et al., 2014). However, the sudden change in PM₁₀ in 2010 relates to altered assessments of industrial emissions. 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.

Selya (1975) listed the two-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 SO₂ concentrations in the early part of the record (Fig. 2c) and the suggestions of high levels from Chow et al (1983).

Figure 3a shows the mole ratio of nitrogen to sulfur in the gas phase (i.e., $n_{\text{NO}_x}/n_{\text{SO}_2}$ as points and a shaded interquartile range). Measurements from the late 1960s (Selya, 1975) would suggest that $n_{\text{NO}_3}/n_{\text{SO}_4}$ was lower than at present (~0.24) in a sulfur dominated atmosphere with uncontrolled industrial use of soft coal; a cheap fuel widely used in factories, hotels, dwellings, schools etc. 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 $n_{\text{NO}_x}/n_{\text{SO}_2}$ from 1.5 in the 1980s to 4.5 over the last decade.

3.2 Change in secondary pollutants

The changing volatile organic components as non-methane hydrocarbons (Fig. 2j) and the increasing dominance of NO_x (Fig. 3a) encourages the formation of secondary pollutants. Toluene has proved particularly relevant to the production of ozone (Hung-Lung et al., 2007), although Kuo et al (2015) suggested that VOC did not significantly correlate with ozone fluctuations in the few episodes examined in Kaohsiung. Over the years 1994 to 2003, Shui et al (2007) noticed that the mixing ratios of NO₂ in Southern Taiwan decreased, while those for ozone increased, which could be accounted for by the reduction in NO₂, because of decreased NO titration. This is illustrated in Fig. 3b, which shows the increasing fraction of NO_x present as NO₂ 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 O₃ to oxidise larger fractions of NO to NO₂. A quarter century back, the five-year average O₃ 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 O₃ 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 sulfate was high.

3.3 Changing health risk

Air pollution poses both long- and short-term risks to health. The risk of daily hospital admissions from air pollution can be calculated on the basis of exposure to the pollutants and here we have adopted the method used in calculating Hong Kong's Air Quality Health Index (GovHK, 2014). The risk is determined as the sum of percentage added health risk (R_{AHR}) for daily hospital admissions attributable to the 3-hour moving average concentrations of NO₂, SO₂, O₃ and particulate matter (here taken as PM₁₀). These risk factors were derived from Hong Kong health statistics and air pollution data, so will not be exact for Kaohsiung, but a reasonable proportionality is likely to exist given a similar population activity and climate. The $R_{\text{AHR},i}$ for each pollutant i , as:

$$R_{\text{AHR},i} = 100[\exp(\beta_i c_i) - 1] \dots (\text{eq. 1})$$

and c_i the 3-hour moving average concentration of pollutants ($\mu\text{g m}^{-3}$), with the factors: $\beta_{\text{NO}_2} = 0.0004462559$; $\beta_{\text{SO}_2} = 0.0001393235$; $\beta_{\text{O}_3} = 0.0005116328$ and $\beta_{\text{PM}_{10}} = 0.0002821751$ (Wong et al., 2012). Although it would make more sense to use PM_{2.5} in these calculations PM₁₀ was used as the record was more complete, but the PM₁₀ can make 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 PM₁₀ 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. 4d, which shows monthly risk across the five-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 years 1993-2020 in the ternary diagram of Fig. 4e. 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.

3.4 Agriculture

Agricultural crops are sensitive to O_3 (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, tomato 12 902 t a⁻¹ and radish 11 496 t a⁻¹).

Crops accumulate damage over time, so it is usual to express the risk to vegetation in terms of an AOT40 (Accumulated Ozone exposure over a Threshold of daytime 40 ppb as ppb h). The summation is typically made over daylight hours over the growing season for the crop, although sometimes it is expressed each month. In Europe the target value is 9000 ppb h considered over five years. The long-term objective is 3000 ppb h. In Europe the growing season is typically May to July. As Taiwan is a tropical country the growing season is more difficult to define as crops tend to be grown throughout the year. The summer days are not especially long either, so daylight hours are taken as being shorter in our calculations: 07:00 to 17:00. There are two periods (Fig. 2i) of high O_3 levels, March-May and September to November (Chen et al., 2004). The three-month long AOT40 for the two urban sites and the rural site at Meinong are shown in Fig. 5ab for each O_3 season. The O_3 has increased dramatically over the late parts of the 20th century, but it is a little uncertain as records don't easily overlap so the cross checking is poor. We can see that Meinong is typically the highest and 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 O_3 in general in the Kaohsiung area (Fig. 2i), there are hints the AOR40 is in decline particularly late in the year. The number of hours each year where late season O_3 exceeds 40 ppb is plotted in Fig. 5c, which suggests that although AOT40 might be decreasing, the number of hours above 40 ppb are relatively stable over recent years. The decline in AOT40 is mostly caused by a decrease in hours with high O_3 (i.e. >80 ppb). These have declined recently especially in the late part of the year, but since 2001, in both ozone seasons, high O_3 periods have become been less common.

There are only a few studies of O_3 and crop damage in Taiwan (e.g. Sheu & Liu, 2003). Additionally, there has been little research on major crops found in the Kaohsiung area except for some studies of soybean and tomato. Soybean shows visual damage after accruing several thousand ppb h across the growing season (Gosselin et al., 2020), conditions certainly exceeded at the Meinong site (Fig. 5a,b). Over a period of weeks, tomatoes (*Lycopersicon esculentum* Mill. H-11) exposed to O_3 at 200 and 350 ppb for 2.5 h at 3 days a week showed extensive foliar injury, defoliation, reduction in biomass, although fruit yield was only lower at the higher concentration (Oshima et al., 1975). However, such high values are not experienced at Meinong, and even an hourly concentration >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.

3.5 Visibility

Visibility is important as a publicly perceptible marker of air pollution over long periods (Brimblecombe, 2021), although studies from southern Taiwan do not always use the most recent data (Lee & Lai, 2018). Maurer et al (2019) were able to use the record up to 2016, which hints at the influence of PM₁₀ 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}/km^{-1} as:

$$b_{sp} = 0.0046c_{(\text{NH})_4\text{SO}_4} + 0.0067c_{\text{NH}_4\text{NO}_3} + 0.0033c_{\text{TC}} + 0.0032(c_{\text{PM}_{2.5}} - c_0) \dots (\text{eq. 2})$$

where c is the concentration of aerosol components and c_0 a remainder term. Yang et al (2005) determined the amount of $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 assuming that all SO_4 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 . The calculated visibility from aerosol measurements listed in the supplementary materials can be compared with observed visibility in Kaohsiung (Fig. 6). Improvement is not always dramatic, and as Li et al (2022b) show for Beijing the secondary aerosol can make contributions to visibility that can outweigh the effects of primary emissions.

4. Discussion

The long-term change in air pollutants in Kaohsiung, shows 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 concentrations of SO_2 first, with NO_x and PM seeming to reach a maximum in the last decade of the 20th century. However as dramatic as many of the improvements have been, the changing economy of the city has made a significant contribution to reductions. The $\text{NO}_3^-/\text{SO}_4^{2-}$ 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 sulfur 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 concentrations on NO_x have declined in line with emissions (Fig. 2ef) suggesting some regulatory success in responding to an enlarged automobile fleet, i.e. private vehicles 1994: 432 228 to 2020: 763 975 (KCGDG, 2021). 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. 2h). Aerosol NH_4^+ is probably insensitive to the total NH_3 , but highly sensitive to total H_2SO_4 and HNO_3 (Cheng & 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.

Concentrations of O_3 , as a secondary pollutant, have remained high for the last quarter century and represent a risk to health and a threat to agriculture. 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). Chemical transformations mean that particulate SO_4^{2-} and NO_3^- concentrations might not necessarily follow regulatory improvements to their precursors. However, in Kaohsiung it is likely that over longer timescales, particulate sulfate has declined in parallel with SO_2 . The Theil-Sen slope for the medians of the SO_4^{2-} suggests a decline of $\sim 0.3 \mu\text{g m}^{-3} \text{ a}^{-1}$ from 1970, which would accumulate to $\sim 80\%$ over time. Since the mid-1990s it decreased from $11.5\text{--}14.3 \mu\text{g m}^{-3}$ (Lin, 2002; Yang et al., 1998) to $3.9\text{--}4.4 \mu\text{g m}^{-3}$ at present (Shen et al., 2020), i.e., $\sim 65\%$ decline. This is proportionally less than the decrease in SO_2 , from $\sim 25 \mu\text{g m}^{-3}$ in the 1990s to $\sim 3 \mu\text{g m}^{-3}$ at present ($\sim 90\%$ decrease).

5. 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 around sites such as Xiaogang. On a day-to-day basis, concentrations 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 sulfur emissions. Both regulation and economic changes have made improvements in air quality possible over recent decades, yet O_3 remains a problem. This secondary pollutant difficult to control as it needs 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 data for spatially resolved emission over long periods can be difficult to assemble. However, economic records could reveal fuel imports and farming statistics which would suggest the magnitude and distribution of emissions. Changes in secondary organic compounds have been neglected in this study, though it would be interesting topic for further research. Assessing the effects of emission reduction on visibility change or health outcomes is important when 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.

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/organization where the work has been carried out, before the work was submitted.

Author Contributions: 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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Availability of data and materials: The data is publicly available as denoted by URLs in the text

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Figures

Figure 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.

Figure 2

Monthly average TEPA measurements from sites in Kaohsiung, with seasonal patterns (averaged 2000-2020) as insets and annual emissions from the Kaohsiung special municipality. (a) PM₁₀ (black) and PM_{2.5} (grey) concentrations, (b) PM₁₀ emissions, (c) SO₂ concentrations (rural Meinong noted as low

concentration series), (d) SO₂ emissions, (e) NO_x concentrations, (f) NO_x emissions, (g) CO concentrations, (h) NH₃ emissions, (i) O₃ concentrations, (j) NMHC emissions.

Figure 3

(a) Monthly average ratios of nitrogen (NO_x) to sulphur (SO₂) from TEPA measurements at sites in Kaohsiung. (b) The fraction of NO_x as NO₂ at Fuxing (red squares) and Meinong (green circles).

Figure 4

Monthly added contributions to health risk from PM₁₀, NO₂, SO₂ and O₃ at (a) Fuxing, (b) Xiaogang and (c) Meinong. (d) Ternary diagram showing monthly added risk PM₁₀, NO₂, SO₂ and O₃ at Fuxing (red-brown squares), Xiaogang (red diamonds) and Meinong (green circles) from 2016-2020. (e) Added average annual contributions to health risk from PM₁₀, NO₂, SO₂ and O₃ at Fuxing (red-brown line), Xiaogang (few points as diamonds) and Meinong (green line).

Figure 5

(a) AOT40 for the season March-May at Xiaogang, Fuxing and Meinong. (b) AOT40 for the season September-November at the three sites (c) Number of hours each year when O₃ exceeded 40 ppb for the September-November season at the three sites Month. Inset shows the number of hours when O₃ exceeded 80 ppb at Meinong for the March-May (small dots and fine line) and September-November (large squares and thick line) seasons.

Figure 6

The light scattering coefficient (inverse visibility) in Kaohsiung with a line showing annual values from Maurer et al (2019), extended to the present, and small shaded squares monthly values from CODiS. The large open squares are medians and the interquartile range calculated from the aerosol measurements using eq. (2) Note: see supplement for details on the data sources. In calculations data from Chen et al (2001) SO₄²⁻ was set to 5 µg m⁻³ and Yang et al (1998) PM_{2.5} was set to 30 µg m⁻³.

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