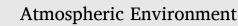
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Historical dry deposition of air pollution in the urban background in Oslo, Norway, compared to Western European data

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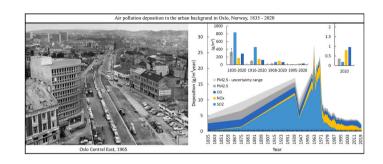
HIGHLIGHTS

G R A P H I C A L A B S T R A C T

- Historical 1835–2020 dry deposition of SO₂, NO_x, O₃ and PM_{2.5} in Oslo, Norway.
- From fuel consumption, to emissions, to air concentrations, to dry deposition.
- Oslo air pollution loads are later and lower than London and Paris.
- Trends of Oslo PM_{2.5} pre-date, SO₂ shortcut, NO₂ follow Kuznetsov Curve.

ARTICLE INFO

Keywords: Air pollution Historical emissions Dry deposition Oslo Buildings Kuznets curve



ABSTRACT

The historical (1835-2020) dry deposition of major air pollutants (SO₂, NOx, O₃ and PM_{2.5}) in the urban background in Oslo, Norway, in a situation that could represent the building facades, was approximated from reported fuel combustion, emission factors, air concentrations since 1960, and dry deposition velocities. The annual accumulated dry deposition (and thus not considering the removal processes) of the pollutants, together, was found to have varied from about 2.3 to 27 g m $^{-2}$, with the maximum in the 1960s caused by high SO₂ emissions from the combustion of fuel oils, and with 1.6 kg m^{-2} having deposited over all the years. The deposition of PM_{2.5} was found to have dominated from 1835, have increased to a maximum in 1875 and then slowly decreased. The SO₂ deposition decreased to a low value around 1990. The NO_x deposition was also at its highest in the 1960s to about 1970, it became the largest from the 1980s, and then showed a clear decrease from about 2010. The O_3 deposition was lower in the years of the maximum total and NO_x deposition. The dry deposition of O_3 and NO_x were found to be about similar in 2020, more than two times that of $PM_{2.5}$ and more than four times that of SO_2 . The trends of the NO_x emissions were found to reflect the relative (1975) and absolute (\sim 2000) turning points of the environmental Kuznets curves (EKC) that has been suggested for Norway, whereas the trend of the SO_2 emissions seems to have "shortcut" this development by the strong regulations in the emissions from 1970 that lead to near simultaneous relative and absolute reductions. The gradual decrease of the PM_{2.5} emissions from about 1945 seems to correspond with the decrease in combustion energy intensity in the economy as wood was substituted with more energy efficient fuels and then with the continued reduction in the wood burning.

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1. Introduction

Anthropogenic air pollution damages health (WHO, 2013; WHO, 2005), natural ecosystems (Maas and Grennfelt, 2016), and built structures (Watt et al., 2009; Brimblecombe, 2003) which are the topic of this work. The effects are local (Grøntoft, 2019), regional (EMEP, 2021), and global (WHO, 2005). To increase the awareness about this problem, it is important to report its different aspects in easily understandable terms. Information about the risks and damages, should engage the economic and societal interest, including private and public owners, involved in maintaining the built environment. The mass deposition of air pollution to buildings varies depending on the local air concentrations, meteorological conditions, and surface characteristics. The air pollutants wet deposit with precipitation and dry deposit. As the precipitation will affect the accumulation of the pollutants in different ways and often also lead to rain washing, the deterioration effects of the wet deposition are less predictable (Grossi and Brimblecombe., 2004). In interaction with the climatic conditions, the deposition increases weathering, corrosion, and soiling (Watt et al., 2009; Graedel and Leygraf, 2000). The particle soiling and combined effect with SO₂ and sulfate (SO_4^{2-}) in the formation of black crusts on limestone (Brimblecombe, 2003; Inkpen, 2004) and corrosion of steel and other metals (Graedel and Leygraf, 2000) are among the most observed damages. Corrosive effects of ozone have been reported both on inorganic and organic materials, outdoors and indoors (Lee et al., 1995). Synergistic effects of O3 have been observed with SO2 in the laboratory in the corrosion of metals and calcareous stone (Screpanti and De Marco, 2009; Kucera and Fitz, 1995) and outdoors in for example the corrosion of copper (Kucera et al., 2007), and with NO2 in the formation of nitric acid and corrosion of limestone (Kucera, 2005a). O3 can corrode rubbers (Brimblecombe and Grossi, 2010; Ryhl-Svendsen, 2008) and polymer coatings (Brimblecombe and Grossi, 2010; Reichert and Pohsner, 2006). After ventilation to the indoors O₃ can react on surfaces (Rim et al., 2016; Lamble et al., 2011; Cass et al., 1991) and damage sensitive materials such as paper ((Menart et al., 2014)), colourants (Whitmore and Cass, 1989), and pigments (Shaver et al., 1983). O₃ can react indoors with NO₂ to form nitrate radicals, and heterogeneously on surfaces to nitrous (HNO₂) and nitric acid (HNO₃), that can damage sensitive materials (Graedel and Levgraf, 2000; Nazaroff and Weschler, 2021; Weschler and Shields, 1997; Hackney, 2016).

The tolerable¹ (CLRTAP, 2014) impact of the air pollution before maintenance of facades and cultural heritage has been reported as the amounts of wear (Spezzano, 2021; Kucera, 2005b) or the air concentrations which will result in the wear (Kucera, 2007). The costs of the air pollution, and savings from its reduction, have been reported (Grøntoft, 2019; Rabl et al., 2014a; Grøntoft, 2020; Grøntoft et al., 2019). The reports of the concentrations, mass deposition, impacts and costs are needed for mitigation and regulation purposes.

Measurement programs that were initiated in the mid-1950s (Egnér and Eriksson, 1955) showed that long-range transport of sulfureous emissions from western Europe contributed to acid rain and increasing SO₂ concentrations in Scandinavian rural areas from about 1960 (Oden, 1968). An engaged debate about the recorded and potential negative effects started, including about corrosion of buildings and monuments (Oden, 1968). The long period impacts of air pollution on buildings have been studied in several works. Damage functions were used to estimate the deterioration of carbonate stone, iron and copper and the change in

the blackening of stone surfaces in London (Brimblecombe and Grossi, 2009) and weathering of materials at the Notre Dame cathedral (Brimblecombe and Lefèvre, 2021) in Paris over the last millennium, and the haze on glass in Paris since 1850 (Ionescu et al., 2012). These studies applied historical values for the air pollution derived from non-instrumental weather records and pollution models (Brimblecombe and Grossi, 2009), besides fuel use data (Brimblecombe and Lefèvre, 2021; Ionescu et al., 2012) and available air pollution measurement data from the 20th century as also in this work, and modelling predictions until 2100. They found that the deterioration was especially intense in London from the 18th until the end of the 20th century, and that "the centuries where pollution controlled the damage to durable building materials seems to be over" (Brimblecombe and Grossi, 2009). A clear correlation has been found between the chemical composition of crusts on limestone sculptures and their exposure to historical periods with different air pollution over the last 350 years in Oxford, UK (Wilhelm et al., 2021). A decreasing rate of erosion was measured on horizontal surfaces of the St. Paul's cathedral in London from 1980 to 1990 to 1990-2000, which was explained with decreasing SO₂ levels (Trudgill et al., 2001). In Paris, the period of damage from aggressive pollutants was found to be later and briefer than in London (Ionescu et al., 2012). It was reported that the increasing use of coal fuel through the 19th century "enhanced the sulfur dioxide concentration and caused the rapid increase in haze formation that peaked about 1950", and that "damages to medieval glass likely followed a similar pattern". The changes were interpreted in terms of a U-shaped Environmental Kuznets curve (EKC) (Brimblecombe and Grossi, 2009; Grytten et al., 2018), which describes environmental degradation that first increases in the initial phases of economic growth, and then declines as the growth of developed economies enters a certain level. In Norway it was found that the energy consumption per gross domestic product (GDP) per capita increased at a lower rate from 1975 and fell from 2002 (Grytten et al., 2018), and it was concluded that 1975 and 2002 were relative and absolute turning points of the EKC that were likely to be reflected in the environmental loads.

This work approximates the accumulated mass of dry deposition (and is not thus considering removal processes) since 1835 of the major air pollutants, sulfur dioxide (SO₂), nitrogen oxides (NO_x = nitrogen dioxide (NO₂) + nitric oxide (NO)), ozone (O₃) and particles with mean aerodynamic diameter less than 2.5 µm (PM_{2.5}), in a general urban background situation in Oslo, Norway, that could represent the building surfaces. Bearing in mind that the additional effects of wet deposition may increase or change the pollution impacts. Lacking reports on dry deposition velocities to buildings, their values were chosen in the lower ranges of those reported on land. It should be noted here, already, that this choice involves considerable uncertainty as the variations in the dry deposition velocities in the "general urban outdoor" including to building surfaces are expected to be large, as also reported to general land surfaces (including water, vegetation, and the different seasons). The estimated deposition fluxes can be adjusted to different values of the dry deposition velocities by their direct proportionality (see sections below).

2. Method and data

2.1. General

The mass dry deposition of SO₂, NO₂ and PM_{2.5} was estimated from the area averaged, urban background concentrations of the air pollutants and their dry deposition velocities. For the years before 1960 the concentration of the air pollution in Oslo was approximated from the reported energy consumption from fuel combustion (in Petajoule, PJ) from the burning of wood, coal and coke, oil, and gas, excepting the merchant navy (Lindmark and Minde, 2018; Grytten et al., 2018). For the time after 1960, concentrations reported from detailed emission-dispersion modelling or from measurements were used (Gram, 2005; Lindberg, 1969; Larssen and Hoem, 1990; Bartonova et al., 2002;

¹ Although negative environmental impacts could always be considered unacceptable, it is ordinary to set ("acceptable") pollution limit, or target, values, for example for health exposure, based on precent risk information and/or what could be practically achieved. The term "acceptable" corrosion of materials is according to ((CLRTAP, 2014)) reserved for technical constructions while "tolerable" is used in connection with degradation of cultural heritage. The term "tolerable" is used in this paper, covering both meanings.

Tidblad et al., 2014; Grøntoft and Ferm, 2017; Grøntoft and Roux, 2020; Air quality monitoring data, 2020).

A fixed correlation was assumed before 1960 between the concentrations of the single air pollutants in Oslo and their national emissions. The correlation factor was determined as the proportion between the national emissions and reported concentrations in 1960. It was thus assumed that in the years before 1960 the fuel combustion represented the same fraction of the emissions to the air concentrations as in 1960. It was further assumed that the fraction of the concentrations of the air pollution in the centre of Oslo to the total Norwegian emissions were the same from 1835 to 1960. The variations in the fuel consumption, and emissions, in Oslo as compared to the Norwegian total before 1960 is not well known. Although the use of different fuels will have been differently distributed over time throughout the country, it seems a reasonable assumption that during the nineteenth century the emissions of air pollutants in the quickly growing and early industrialized city of Oslo correlated strongly with the national fuel consumption. The population growth will have been a major factor in this correlation. After about 1900, this correlation may have become weaker. Then after 1945 and especially after about 1970 with the new concerns about the urban environment and air quality, it is likely that the temporal correlation between the national and Oslo emissions and concentrations of air pollution weakened and changed. A more detailed description of the urban development, energy use and emissions in Oslo since 1835 are given in Supplementary materials 1 (S1). The uncertainties in these approximations will be discussed along with the more detailed explanation of the estimation procedures, where an uncertainty assessment was applied to the reported results. In other cases, to make the reading easier, main uncertainties will be noted and discussed later in the Discussion and uncertainty section.

2.2. Emissions and emission factors

The calculations of the pollution emissions were performed for breaking points in the historical record of the energy content of fuel consumption (1835-start, 1875, 1940, 1945, 1960) and, in addition, every 10th year from 1970 until 2020, with interpolation between these years. The historical record showed near linear trends in the fuel consumption between these years. The reported energy consumption in the breaking point years was multiplied with the energy contents of the fuels, of 10, 28.1, 40 and 40.4 GJ T^{-1} for wood, coal and coke, oil, and gas (Applied R Code and Data Science for Immediate Application, 2014; Norskenergi, 2021), to obtain the mass consumption of the fuels. The Norwegian mass emissions of the air pollutants were then calculated by multiplying the fuel mass consumption with the emission factors (kg T^{-1}) for the release of the air pollutants from the fuel burning found to best approximate the combustion situation in 1960. The emission factors were averaged over the fuel types and combustion technologies. These values for the emission factors were also used to calculate the emissions in the period from 1835 to 1960.

Some information was available about emission factors for SO₂ and NOx in Oslo back to 1960 (Gram, 2005). Tabulated values of emission factors for specific fuel and combustion technologies were partly available from 1990, and mostly available for recent years (2015) (Informative Inventory Report (IIR), 2017). Values for the emission factors before 1990, and more so for the emission factors before 1960, are clearly uncertain. Values were for example not found for the changes in the sulfur content in the mix of fuel oils combusted in Oslo before 1960. The historical (before the 1960s) emissions of particles from wood burning seemed most uncertain. The uncertainty in the NO_x emissions is of less importance for the evaluation as they were much smaller than the emissions of SO2 and PM2.5 before the increasing use of petroleum products after 1945. The particle emissions from typical iron ovens used in the 19th to early 20th century have been reported to be 1.5 to 2 times higher than the present environmental limits (Seljeskog and Kausch, 2018). Based on this information and the assessment that the

combustion of coke and coal with earlier technologies probably also emitted more particles, an uncertainty range up to a 100% increase in the value of the emission factor for wood combustion in 1960, is suggested for the years before 1960, to represent a probability range of the overall particle emissions. With interpolation from 1960 to the 100% uncertainty in 1945, to obtain a smooth range. For SO₂ and NO₂, a value for the uncertainty is not suggested. It is only noted here that the uncertainty is higher in earlier years, mainly before 1960. The values for the emission factors in 1960, the rational for their derivation and literature sources are given in Table 1.

The, tentative, curves for the emissions from the fuel use in the categories also for the time after 1960 was, as a comparison to the concentrations, obtained, by curve fitting of the emissions calculated by the factors to the reported emissions of the pollutants from combustion (Norwegian Environment Agency, 2021), that were available from 1975 (1995 for PM_{2.5}) to 2017. These approximated emission factors from curve fitting to reported values and a discussion of their comparison with the concentration trends in Oslo are given in Supplementary material 2 (S2).

2.3. Air pollution concentrations

In addition to the air pollution concentrations (of SO₂, NO_x and PM_{2.5}) that were estimated from the fuel consumption and emissions in the years from 1835 to 1960, reports of annual concentrations of SO₂ and NO_x with high spatial resolution (1 \times 1 km) were available for Oslo for the period 1960-2000, based on emission inventories and dispersion modelling (Gram, 2005). Concentrations reported from measurements at urban background stations were obtained for the years after 1959, to compare with the values from the dispersion modelling until year 2000, and to use in the dry deposition estimations, with linear interpolation over periods of years with missing data, from 2000 to 2020. The ozone concentrations were obtained from those of NO_x for the whole period from 1835 to 2020 by: $O_3 = 60.5 \text{ x} \exp(-0.014) \text{ x} \text{ NO}_x$ (Kucera et al., 1995). The technicalities of the derivations of the concentrations and evaluation of these data to be representative for the urban background situation in the centre of Oslo are provided in Supplementary material 3 (S3). The concentrations are given in the Appendix.

2.4. Deposition

The dry deposition fluxes of the gaseous (SO₂ and NO₂) and particulate ($PM_{2.5}$) pollutants were calculated to be proportional with the concentration in air by Equation (1) (Seinfeld and Pandis, 2016).

$$F = v_d \times C \tag{1}$$

where *F* (g m⁻² s⁻¹, reported in the following as g m⁻² a⁻¹) is the pollution flux to the surface, v_d (m s⁻¹, reported in the following as cm s⁻¹) is the dry deposition velocity and *C* (g m⁻³, reported in the following as µg m⁻³) is the pollutant concentration.

The dry deposition velocity is influenced by the air transport of the pollution to the surface and the surface reactivity, that can be described in more detail by such factors as the atmospheric stability, the boundary layer conditions (turbulence intensity), the particle size (of aerosol), the air-surface friction velocity, and collecting properties of the surface (Donateo and Contini, 2014). The surface uptake finally happens by diffusion, or sedimentation of larger particles, through a so called quasi-laminar sublayer of more or less stationary air of thickness on the order of millimetres (Seinfeld and Pandis, 2016). Besides the meteorological conditions, due to the variations in the total areas and other properties of surfaces, depending for example on the amount and type of vegetation and buildings, the ranges of reported dry deposition velocities is considerable (Rabl et al., 2014b). Area averaged values will include local variations for example between forests and urban areas. A precise determination of the air pollution dry deposition velocities to

Table 1

Air pollution emission factors for fuel combustion in Norway in 1960 derived from the literature and applied back to 1835. The suggested uncertainty range for the emission factor of PM_{2.5} from wood combustion before 1960 is given in brackets.

Pollutant	SO ₂				NO _x		PM _{2.5}		
Energy source ^a Year	$Wood^1$	Coal and coke ²	Oil ^{3,4}	Wood ⁵	Coal and coke ⁶ Emission factor (kg T ⁻¹)	Oil ^{3,7}	Wood ⁸	Coal and coke ⁹	Oil ¹⁰
1835–1960 ^b	0.2	25	31	1	1.6	22	22 (44 [°])	1	1

Emission factors in 1960.

^a Gas was only used as a primary fuel source after 1980.

^b The emission factors were derived for the year 1960 and applied also in the estimations of the emissions back to 1835.

^c Upper limit of suggested uncertainty range for the years 1835–1960, interpolated from 44 to 22 kg T⁻¹ from the 1945 to 1960.

¹ (Gram, 2005; Informative Inventory Report (IIR), 2017).

² Average of factors for the stationary combustion of coke and coal in 1960, with the assessed same amount of coke and coal consumption (Gram, 2005).

³ (Gram, 2005) Weighted average of five types of petroleum oils used for heating, petrol, and diesel for cars, consumed in the industry and by the ships in the harbour of Oslo in 1960.

⁴ (Informative Inventory Report (IIR), 2017) reports a value of 17 for heavy fuel oils in 2015.

⁵ (Gram, 2005; Informative Inventory Report (IIR), 2017).

⁶ (Gram, 2005) Weighted average for coke and coal consumption for heating and in the industry in Oslo in 1960. (Informative Inventory Report (IIR), 2017) report a value of 3 for combustion in small stoves.

⁷ The value was calculated to be the same as that of factors from (Informative Inventory Report (IIR), 2017) weighted with reported emission amounts in combustion categories for Norway in 2019 (Statistics Norway, 2019).

⁸ (Informative Inventory Report (IIR), 2017) Fuel wood combustion in small stoves in 1990. Very little change was reported between 1990 and 2000. The same value was used for 1960.

⁹ (Informative Inventory Report (IIR), 2017) Approximate average of reported combustion technologies in 2015 = 0.8. A value of 1 was used for 1960, as for PM_{2.5} emissions from oil.

¹⁰ (Informative Inventory Report (IIR), 2017) Reported values for heavy sulfur containing fuel oils in 1990 = 0.76 and in 2000 = 0.68. Extrapolated back to 1960 with the rate of change reported between 1990 and 2000.

buildings in an urban area, like Oslo, is difficult. Different reduction effects on the concentration of airborne particles have been found for green areas in urban spaces with different green coverage rates (for example Qiu et al., 2019), which might imply an overestimation of the deposition of air pollution to buildings by general reported deposition velocities to the ground. The area fraction of the tree canopy (leaf) to green spaces in the inner city and the total built zone of Oslo were reported to be 3 and 1.2 (Hanssen et al., 2019). The fraction of the total building to sealed surface areas, in the inner city and in the total built zone, were reported to be 0.7-1, and 0.4-0.6, if the total building façade (including roof) area is, on average, 2 to 3 times the land area coverage of the buildings. Thus, the total surfaces of canopy (leaf) in the city seem to be about 2-3 times those of the building surfaces (when assuming that green spaces and sealed surfaces have the same geometric areas), which would influence the amount of dry deposition. It is reported that the reduction in concentrations of primary PM10 by realistic ambitious planting of trees is in the range 2-10% and of PM from all sources and for practical planting schemes only a few percent (The Air Quality Expert Group of the Department for Environment et al., 2018). The effect of the tree plantings on the air pollution concentrations is related to the deposition and dispersion in complex ways depending on their characteristics and location (Janhäll, 2015; Setälä et al., 2013; Tong et al., 2013). It is not a direct relationship between concentration and deposition, as both elevated concentrations due to for example reduced dispersion among trees, and reduced concentrations, might imply higher deposition. The relatively low effect of tree planting on concentrations may indicate a similar low effect on the overall deposition, but this would depend on the pollution air transport and verification by deposition measurements seems needed. In unbuilt areas outside the city more variation is expected, between for example dense forests, cropland, and heaths. In all, it seems usually the deposition velocities are higher to tree covered and most vegetated surfaces as compared to buildings. Therefore, values in the lower general ranges of reported dry deposition velocities were used to estimate the deposition in the urban background, that could represent buildings, in this work, except for O₃, for which a value reported to represent a situation for "Norway" was used. Table 2 gives the literature sources, values, and explanations of the derivations of dry deposition velocities used in the estimations. The uncertainty in these values is, clearly, considerable and should be kept in mind.

Table 2	
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Suggested dry deposition velocities to the urban background repre-
senting buildings, in Oslo, Norway.

Pollutant	Dry deposition velocity (cm s^{-1})
SO ₂	0.35 ^a
NO ₂	0.14 ^b
O ₃	0.13 ^c
PM _{2.5}	0.16 ^d

^a Low estimate of (Rabl et al., 2014b). A mean dry deposition velocity of 0.45 cm s^{-1} , within the range $0.1-0.7 \text{ cm s}^{-1}$ was used for deposition estimations at Norwegian background stations by (Aas et al., 2018).

^b (Nowlan et al., 2104) They report the value of 0.14 cm s⁻¹ to be more realistic and in better agreement with the literature (as summarized in (Holland et al., 2005)), than a previous value of 0.34 cm s⁻¹ reported by (Holland et al., 2005), who also noted, "that their NO₂ dry deposition velocity (of 0.34 cm s⁻¹) was larger than most previously published values". (Aas et al., 2018) report values used for dry deposition estimations at Norwegian background stations, in a range from 0.1 to 0.5 cm s⁻¹ (Aas et al., 2012) report a general low dry deposition velocity regulated by stomatal control in the range 0.2–0.4 cm s⁻¹, but in winter of 0.02 cm s⁻¹.

^c Value reported for the latitude of Oslo (60°) by (Falk and Haslerud, 2019).

^d "Best guess" for PM₁₀, of (Rabl et al., 2014b), multiplied with the fraction 0.45 of the available mean annual concentrations of PM_{2.5} to PM₁₀ measured on air quality measurement stations in Oslo from 2006 to 2019. The PM dry deposition have been found to significantly correlate with the PM concentration (Wu et al., 2018). This is in the lower range of values reported in other works (Wu et al., 2018; Saylor et al., 2019; Szep et al., 2016).

The concentration and dry deposition velocity of PM_{10} are typically the double of those of $PM_{2.5}$ (Rabl et al., 2014a). Thus, a four times higher dry deposition (flux) of PM_{10} than of $PM_{2.5}$ could be expected. Relatively more of the larger particles in the PM_{10} range and above will tend to settle by gravitation and thus on horizontal surfaces. They will not follow the air flow as closely as the gases and smaller particles and the larger extent of impaction can lead to increased localized dry deposition (Seinfeld and Pandis, 2016).

3. Results

Fig. 1 shows the approximated Norwegian mainland emissions of the air pollutants from fuel combustion since 1835, compared with the air concentrations in Oslo.

It can be seen in Fig. 1 that until about 1950 coal and coke was the main source of the SO₂ emissions. Then, from about 1950 to 1980, sulfur containing heavy fuel oils was the main source of the SO₂ emissions. The SO₂ concentrations reported from the emission-dispersion modelling are somewhat lower than those reported from measurement before 1990. The concentration of SO₂ in Oslo was from 1970 sharply reduced, and it seems more quickly than the national emissions. Apart from that, the trend of the concentrations in Oslo follows the trend of the national SO₂ emissions.

The small and slowly increasing emissions of NO_x from 1835 to 1940 seem to have come mainly from wood combustion until about 1880, then increasingly from petroleum products to become larger than from the wood combustion before 1900, with a contribution from coal and coke that had also become larger than from wood before 1940. From 1945 the emissions from the combustion of petroleum products increased sharply to become, until the present, the absolutely dominating source of the NO_x emissions. The reported NO_x concentrations from the dispersion modelling are much lower than the measurement values from the central traffic stations in Oslo, and the trend of the concentrations and emissions (Gram, 2005) in Oslo is negative from 1970, whereas the national emissions are increasing in the 1990s before a reduction was reported from about 2000. The Norwegian emissions of $PM_{2.5}$ have probably decreased since 1875, except for the years 1940–1945. Wood burning has in all the years since 1835 been the major emission source of $PM_{2.5}$. The emissions of $PM_{2.5}$ from petroleum products increased from about 1950, to become a notable fraction also due to a slight reduction in the contribution of wood burning over the same period. From about 1985 to 1995, the concentration of $PM_{2.5}$ in Oslo seems to have decreased relatively more than the national emissions (Fig. 1).

Fig. 2 shows the approximated annual dry deposition since 1835, in the central urban background in Oslo. It also shows the total accumulated dry deposition of the pollutants since 1835 (185 years), in three shorter periods before the present, from 1916 (104 years), 1968 (52 years) and 1995 (25 years), and in the year 2020. The natural and anthropogenic removal processes, such as rain washing, and façade maintenance and cleaning, are not considered.

The variation in the total (of the assessed pollutants) annual dry deposition in the Oslo central urban background is seen in Fig. 2 to be due to a maximum in the particle deposition in 1875 and then a subsequent gradual decrease, an increase in the SO₂ deposition until 1940 and then a peak from 1960 to 1970, an increase in the NO_x deposition from about 1950 to 1970 and a subsequent slow decrease, and a reduced deposition of O₃ in the peak years for the NOx emissions around 1970. The O₃ dry deposition was found to be the highest of the pollutants in the first and last 10 years of the evaluation period (from 1835 to 1845 and after 2010). A peak in the total dry deposition is seen during the years of high SO₂ emissions between 1960 and 1970. The displayed accumulated values reflect these trends. In 2020 the NO_x, PM_{2.5} and SO₂ deposition

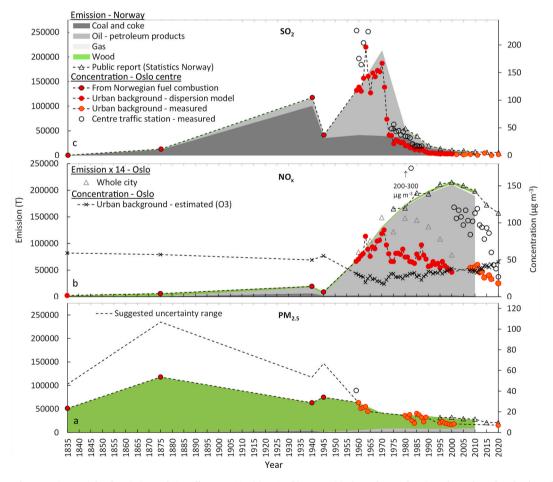


Fig. 1. Approximated Norwegian mainland emissions of air pollutants, SO_2 (a), NO_x (b), $PM_{2.5}$ (c), from the combustion of wood, coal and coke, oil, and gas, fitting to recent reported emissions, concentrations estimated from the emissions before 1960, and measured concentrations in Oslo since 1960. The concentrations marked in red (including orange) were used for the deposition estimates and are given in the Appendix. The legend entries are only included once for all the three diagrams. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

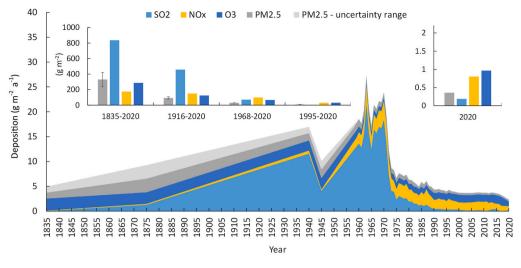


Fig. 2. Calculated accumulated dry deposition (neglecting removal processes) of air pollutants in the urban background centrally in Oslo since 1835. The ranges of the $PM_{2.5}$ dry deposition represents the variation from estimates with the low to high value of the $PM_{2.5}$ emissions factor (for wood, Table 1). The accumulated dry deposition and uncertainty ranges of $PM_{2.5}$ are reported from the middle of the displayed uncertainty range.

was found to be 83%, 37% and 19% of the O_3 deposition.

4. Discussion and uncertainty

4.1. Emissions factors

This work applied emission factors of air pollution from the combustion of major fuels and technologies in Norway, reported as time series back to 1990 (Informative Inventory Report (IIR), 2017). Some earlier estimates were found in the literature (Gram, 2005). It was however difficult to assess the earlier (than 1990) emission factors averaged over the fuels and combustion technologies. The uncertainty in these reported values (Informative Inventory Report (IIR), 2017) due to changing knowledge about for example the condensable fraction of the particulate matter was not evaluated. The uncertainty in the derivation of single averaged values of the emission factors for the fuel categories and combustion technologies in 1960 is considerable, and the uncertainty increases for earlier years. The uncertainty in the estimation of the particle emissions from wood combustion was evaluated to be the largest, both due to the relative magnitude of these historical emissions and the possible variations in the combustion. A suggested uncertainty range (of x 2 before 1945–1960) was therefore included for the PM_{2.5} emissions and dry deposition. The particle emission factors of other wood products, than fuel wood, such as waste and pellets are generally lower (Informative Inventory Report (IIR), 2017). It was considered that the combustion was of mainly fuel wood before 1960 with increasing use of different wood products in the years thereafter. There will, however, have been some amount of burning of, especially, wood waste also before 1960. The particle emission factors and total emissions from the combustion of coke and coal, and oil, were found to be much smaller than from wood. Due to the unavoidable proximate nature of this uncertainty range, it was suggested to illustrate the probable uncertainty in the total PM2.5 emissions from all the fuels. It was not possible to suggest the variation in the SO₂ emission factor of the mixes of sulfur containing fuels and combustion technologies, or the variations in the SO₂ fuel as compared to process emissions, before 1960. The pre-1960 NO_x emissions were small. The uncertainty range for PM2.5 was therefore presented together with a general notation about the uncertainty in all the estimated historical emissions, but without suggesting other ranges than for PM_{2.5}.

4.2. Concentrations

The major uncertainty is in the concentrations, and thus deposition values, from 1835 to 1960. The approximation from the reported national fuel consumption and assumed constant fuel category averaged emissions factors and constant fraction of the concentrations in Oslo to the national combustion emissions as in 1960, is, obviously, highly uncertain. Some reasons for historical variations from or around the presented long-term dry deposition trends in Oslo before 1960 are: changes in the fuel category emission factors, changes in the fraction of the emission of the fuels to process emissions, and changes in the distribution of the emissions between Oslo and the rest of the country (see also Methods section, General, and S1) including differences in the short- and long-range transport and formation of secondary aerosol, that will have affected the concentrations in Oslo. For example, it was reported that in 2015 the emissions of SO_2 , NO_x and PM_{10} from energy consumption were 35%, 86% and 65% of the total emissions (Informative Inventory Report (IIR), 2017). Since 1990, the reductions in the SO₂ emissions from energy combustion were 77% whereas the reductions from the metal industry, being the remaining largest emission source in 2015, were 54%. This illustrates historical variations in the emission source partitioning of SO₂ from fuel combustion and industrial processes. The trend of long-range transport of sulfureous emissions from western Europe (Oden, 1968) seems weak before 1960 and this transport was a small fraction of the urban SO_2 . This is thus probably a minor error. It seemed difficult to obtain more certain results for the dry deposition trends without a much larger data collecting and aggregating effort than was performed, considering also the historical and geographical distribution of the air pollution, that would be difficult or unfeasible to do today.

The minor uncertainty is in the concentrations after 1960 (as compared to those derived for the period from 1835 to 1960). The uncertainty in the concentrations (of SO_2 and NO_x) from the dispersion modelling was discussed by (Gram, 2005) who asserted that "it was difficult to report the uncertainty due to the many needed assumptions made", and that "the levels would be correct, but there could be geographical (local) displacements". The concentrations from the city measurement stations were reported as quality-controlled data (Air quality monitoring da, 2020) and should be less uncertain. The difference between the dispersion modelled and measured concentrations for SO_2 and NO_x (Fig. 1) may be explained by their different emission sources. Whereas the release of SO_2 was from chimneys at some elevation with spreading to the location of the roadside measurement

stations, the NO_x emissions was mainly from the close to station traffic emissions. Thus, the SO_2 measurements should represent the modelled dispersion to a larger urban (background) area, whereas the NO_x measurements would represent values close to the source before much dispersion.

4.3. Deposition velocities

Dry deposition velocities vary much depending on meteorological and surface characteristics and are typically reported for non-urban land surfaces. It is difficult to assess how well the lower value in the reported ranges of the deposition velocities used in this work represent the temporal average of the dry deposition velocities to the urban background or more specifically to buildings in Oslo. The typical double deposition velocity and concentration of PM₁₀ to PM_{2.5} (Rabl et al., 2014a), would imply an, on average, four times higher mass deposition of PM₁₀, than that estimated of PM_{2.5}. Validation of the results of the deposition with measurements and/or other estimations is needed. There will however be large local variations, depending on emission sources, meteorology, and landscape and building features, that may be more practically significant than average measures. Emission from buses and other vehicles can be very close to buildings. Turbulence and local air movements, building geometry and drainage patterns will be very important. The deposition to more prestigious or cultural heritage buildings may be deemed more critical and lead to higher maintenance costs. The perceived damage is thus both a physical and economic/social/political issue.

4.4. Comparison with London and Paris

Fig. 3 shows the concentrations in the centre of Oslo compared to

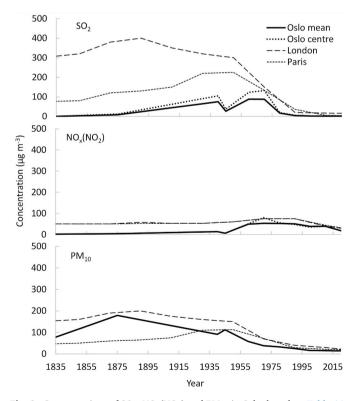


Fig. 3. Concentrations of SO₂, NO_x(NO₂) and PM₁₀ in Oslo, based on Table A1 and (Gram, 2005), compared to reports from London (Brimblecombe and Grossi, 2009) and Paris (Ionescu et al., 2012), since 1835. The PM₁₀ in Oslo was calculated as 2.2 x PM_{2.5}, as observed on Oslo measurement stations (see S3). The SO₂ and NO_x(NO₂) are given both for the Oslo centre and city average situations, as five years moving averages.

those reported in London (Brimblecombe and Grossi, 2009) and Paris (Ionescu et al., 2012), since 1835. The direct comparison of $NO_x(NO_2)$ assumes that the NO_2 reported in London and Paris includes full transformation of the main primary emission, NO, to NO_2 , to compare with the NO_x in Oslo.

The figure reflects the later industrialization in the smaller country of Norway than England and France, with a later and shorter period of, a lower, maximum SO₂ than in London and Paris. The SO₂ and NO₂ concentrations in Oslo are observed to be much lower than London and Paris until the peak years in the 1960s and 1970s, when the SO₂ concentration had already been falling in London since the end of the 19th century and in Paris since about 1950. The NO_x(NO₂) maximum appears at the same time in the three cities, which may be related to the urban traffic development. The PM10 concentrations in Oslo were found to be higher than in Paris until the 1920s and closer resemble the trend in London than Paris before about 1945, after when it resembles the falling trend in these cities at slightly lower concentrations. It was reported that the lower PM₁₀ in Paris than London was due to less coal burning until the beginning of the 19th century (Ionescu et al., 2012). The emissions from coal and coke burning were low also in Oslo until about 1875. It seems that in Oslo the emissions from wood burning in the 19th century may have resulted in higher PM₁₀ than in Paris, approaching the values due to the coal burning in London.

4.5. Comparison to the environmental Kuznets curves (EKC)

The emission curve for NO_x in Fig. 1 seems to reflect the relative and absolute turning points of the EKC in Norway described by (Grytten et al., 2018), with a reducing increase in the emissions from about 1975 and then a reduction from about 2000. This seems to follow from a strong correlation between the NOx emissions and traffic development and industrial fuel consumption (see S1 and S2), with little focus on emission reductions or cleaning of NO_x from flue gases before year 2000. The sharp decrease in the SO₂ emissions from 1970 seems to "shortcut" the development observed for NOx, with the relative and absolute changes happening at the same time. The strong awareness about the damages from SO₂ and the better reduction and cleaning options than of NO_x seems to explain the difference from the somewhat slower and more (directly) economically connected rationale behind the later increasing and then decreasing NO_x emissions. The gradual decrease in the $PM_{2.5}$ emissions from about 1945 seems to correspond with the decrease in energy intensity of combustion due to the substitution of wood with more energy efficient fuels from about this time, described by (Grytten et al., 2018). As the wood thereafter was the dominant source for these emissions this decrease has continued as the wood burning decreased.

5. Conclusion

The historical (1835-2020) dry deposition of major air pollutants (SO₂, NOx, O₃ and PM_{2.5}) in the urban background in Oslo, Norway, were approximated from reported fuel combustion, emission factors, air concentrations since 1960, and dry deposition velocities. Over these years the total accumulated dry deposition of the pollutants was found to have varied from 2.3 to 27 \pm 1 g m $^{-2}$ a $^{-1}$, with 1.6 \pm 0.1 kg m $^{-2}$ having deposited over all the years. The uncertainty was then only considering the PM_{2.5} deposition. The uncertainty in the estimate for each of the other pollutants is considerable but was not suggested. The PM_{2.5} deposition was found to increase to a maximum in 1875 and then slowly decrease. The "traditional" particle emissions from wood combustion seem to have dominated the loads until the second part of the 19th century. The total maximum in the 1960s was caused by high SO₂ emissions from the combustion of fuel oils. The SO₂ deposition decreased to a low value around 1990. The NO_x deposition had a maximum at the same time as the SO2 deposition, in the 1960s and 1970s, became the largest from the 1980s, and showed a clear decrease from about 2010. The O₃ deposition was lower in the years of the

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maximum total and NO_x deposition. The significant increase in the air pollution load in Oslo from the first part of the 19th century parallels, from a lower level, the increases reported from studies of London and Paris (Brimblecombe and Grossi, 2009; Ionescu et al., 2012), but is later, than especially London.

The deposition of the major air pollutants in Oslo seems today lower than it was since 1835, but the NO_x deposition is higher and could thus clearly be reduced, which should also be the case for the PM, as its present emission sources are partly like those of NO_x and related to the urban traffic development. As NO_x decreases, the ozone levels again approach those of pre-industrial times, which might be a conservation concern for outdoor and indoor sensitive inorganic, such as copper, and organic materials, such as paint coatings and diverse cultural heritage items. The influence of climate change on air pollutants like ozone and PM is an important and complex issue related both to long term effects and expected more frequent and dramatic episodes. Warmer climates could increase the emissions of volatile organic gases that are ozone precursors and thus the levels of tropospheric ozone and change the amounts and composition of primary and secondary aerosol (Brimblecombe and Lefèvre, 2021; Sollberg et al., 2008). The consequences of increasing wildfires, and of the soot emissions, is one serious issue. In the

present much improved air quality situation in Oslo the policy awareness of the "traditional" impacts and options for pollution reductions is still important. Increasingly assessment and preparation for new and possibly dramatic air pollution situations is expected to be needed. The status is different in other cities, countries, and parts of the world, but with a similar increasing need to assess climate change impacts.

CRediT authorship contribution statement

Terje Grøntoft: Conceptualization, Methodology, Estimations, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2021.118777.

Appendix

Table A1

Pollution concentrations in the Oslo centre urban background.

Year	Pollutant (µg m ^{-3}), i = interpolated					Year	Pollutant (µg m ^{-3}), i = interpolated					
	SO ₂	NO _x	O ₃	PM _{2.5} low	PM _{2.5} high		SO ₂	NO _x	O ₃	PM _{2.5} low	PM _{2.5} high	
1835	0.4	1.6	59	23	47	1985	12	58	27	18	18	
1840–1875	i	i	i	i	i	1986	9.5	52	29	17	17	
1875	12	4.2	57	54	107	1987	12	70	23	15	15	
1880-1935	i	i	i	i	i	1988	8.8	56	28	10	10	
1940	105	14	50	29	53	1989	6.2	51	30	15	15	
1945	37	6	55	34	67	1990	4.3	41	34	i	i	
1950-1955	i	i	i	i	i	1991	4.1	42	34			
1959						1992	4.3	47	31			
1960	124	51	30	29	29	1993	3.6	44	33			
1961	116	55	28	23	23	1994	3.3	45	32			
1962	140	58	27	24	24	1995	2.6	39	35	9.3	9.3	
1963	196	82	19	25	25	1996	3.5	48	31	11	11	
1964	143	64	25	20	20	1997	3.0	42	34	9.2	9.2	
1965	113	55	28	i	i	1998	3.0	39	35	8.5	8.5	
1966	149	67	24			1999	3.0	36	36	7.9	7.9	
1967	142	65	24			2000	2.7	33	38	7.6	7.6	
1968	154	75	21			2001	i	i	i	8.3	8.3	
1969	151	76	21			2002	1.7			i	i	
1970	167	85	18			2003-2004	i					
1971	124	90	17			2005	1.3					
1972	66	70	23			2006-2007	i					
1973	37	58	27			2008	2.0	39	35			
1974	36	48	31			2009	i	39	35			
1975	21	48	31			2010		40	35			
1976	28	59	27			2011	0.6	43	33			
1977	26	59	27			2012	i	33	38			
1978	23	57	27			2013		34	38			
1979	24	64	25			2014	4.2	26	42			
1980	18	54	29	16	16	2015	i	27	41			
1981	18	54	29	14	14	2016		29	40			
1982	14	48	31	17	17	2017	0.2	24	43			
1983	14	47	31	12	12	2018-2019	i	i	i			
1984	11	45	32	9	9	2020	2.0	18	47	7.0	7.0	

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