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Estimation of the historical dry deposition of air pollution indoors to the monumental paintings by Edvard Munch in the University Aula, in Oslo, Norway

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Abstract

The historical (1835–2020) deposition of major air pollutants (SO_2, NOx, O_3 and $PM_{2.5}$) indoors, as represented by the monumental Edvard Munch paintings (c. 220 m^2) installed in 1916 in the Oslo University Aula in Norway, were approximated from the outdoor air concentrations, indoor to outdoor concentration ratios and dry deposition velocities. The annual deposition of the pollutants to the paintings was found to have been 4–25 times lower than has been reported to buildings outdoors in the urban background in the centre of Oslo. It reflected the outdoor deposition but varied less, from 0.3 to 1.2 g m^{-2} a^{-1}. The accumulated deposition since 1916, and then not considering the regularly performed cleaning of the paintings, was found to have been 43 ± 13 g m^{-2}, and 110 ± 40 g m^{-2} in a similar situation since 1835. The ozone deposition, and the $PM_{2.5}$ deposition before the 1960s, were a relatively larger part of the accumulated total indoor (to the paintings) than reported outdoor deposition. About 18 and 33 times more O_3 than NO_x and $PM_{2.5}$ deposition was estimated to the paintings in 2020, as compared to the about similar reported outdoor dry deposition of O_3 and NO_x. The deposition of $PM_{2.5}$ to the paintings was probably reduced with about 62% (50–80%) after installation of mechanical filtration in 1975 and was estimated to be 0.011 (± 0.006) g m^{-2} in 2020.

Keywords: Urban air pollution, Historical indoor dry deposition, Soiling, indoor environment, Oslo University Aula, Edvard Munch monumental paintings.

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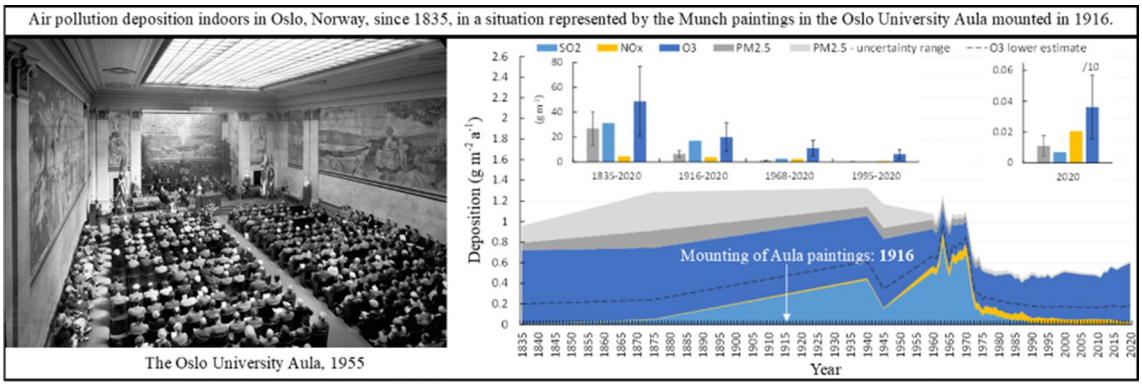
Graphical Abstract



Introduction

The topic of this paper is the indoor mass dry deposition of the major air pollutants, sulfur dioxide (SO_2), nitrogen oxides (NO_x = nitrogen dioxide (NO_2) + nitric oxide (NO)), ozone (O_3) and particles with mean aerodynamic diameter less than 2.5 μm ($PM_{2.5} \leq 2.5$ μm) indoors since 1835 in a situation like the grand paintings by Edvard Munch (c. 220 m^2) mounted in the Oslo University Aula in Norway since 1916. The understanding of the deposition mechanisms, rates and impacts of the air pollution on the paintings is important to determine the optimal conservation measures. The soiling by air pollution physically changes the surfaces of outdoor [1–3], and indoor cultural heritage such as paintings [4, 5], and can accelerate chemical deterioration [6, 7] for example by metal soap formation [5, 8] or iron ion catalysed oxidation, of paper [9]. Measurements of airborne particles in the Aula have not been performed for practical reasons related to its use and lack of resources. An evaluation of the historical and present loads by the available outdoor $PM_{2.5}$ data is therefore considered important. Measurements of airborne particles in Oslo have shown organic (OC) + elemental (EC) carbon concentrations of about 1–3 μg/m^3 with the largest amount in the fine fraction (< 1 μm in diameter) and an EC contribution up to about 1/3. Nearly all of which can penetrate indoors in residential buildings that may however, unlike the present situation in the Aula, also have indoor sources [10]. The EC may (since about year 2000) thus have been up to about 10% of the typical $PM_{2.5}$ concentration in Oslo (of ~ 8 μg/m^3 [11]). Since year 2000 the main sources of $PM_{2.5}$ were wood burning for domestic heating and vehicle exhaust [12]. Historically, especially industrial emissions and smoking indoors will also have contributed. The soiling of the paintings by the small and dark particles, but also those of different composition and size, is a serious concern. Besides the soiling, typical damaging surface reactions involving air pollution are: the acidification, dissolution, and corrosion reactions due to SO_2, sulfite (SO_3^{2-}) and sulfate (SO_4^{2-}) [13, 14]; the decomposition and reactions of O_3 [15–18] especially with double bonds in organic materials [19–21] such as rubber [22] and dyes [23]; and the oxidation by NO_2 of organic materials such as paper [24] and colourants [25], possibly after reaction with O_3 and formation of nitrate radicals ($NO_3\cdot$), and/or the surface reaction of NO_2 to volatile nitrous acid (HNO_2) and adsorbed reactive nitric acid (HNO_3) [26–28]. The long-term impacts of air pollution on buildings have been investigated [11, 29, 32]. The improvement in the condition and savings

Fig. 1 The Oslo University Aula with the monumental Munch paintings, seen during a concert in 1955. Uneven soiling can be observed, especially on the left, as dark horizontal and vertical stripes on the painting History. Image source: Oslo Museum

in conservation costs that could historically have been obtained by reduced air pollution and soiling of the Munch paintings have been reported [33]. The Munch paintings in the Oslo University Aula (Fig. 1) are an important case with some available information about the indoor environment, and where better understanding of the indoor air pollution conditions is needed.

Methodology

The historical indoor mass dry deposition of SO_2, $NO_x(NO_2)$, O_3, and $PM_{2.5}$ was estimated (modelled) from the indoor concentrations of the pollutants, calculated from the urban background concentrations around the Aula [11], and the expected I/O concentration ratios, and the assessed dry deposition velocities of the air pollutants to the paintings, from [34].

$$F = v_d \times C, \quad (1)$$

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.

Site description

The Aula is centrally located and is surrounded by a small green area to resemble an urban background situation. The closest major busy road is 80 m away. The paintings in the Aula were since the mounting in 1916 exposed to air pollution resulting in deterioration, especially by soiling [35]. A brief description of the urban development and air pollution in Oslo was given in [11]. The most radical plans that would directly have affected the environment around and in the Aula, foresaw the building of high rises and of motorways through the very centre of the nineteenth century city (Fig. 2).

Figure 3 shows the floor plan of the Aula, with the dimensions 28(l) m × 16(w) m × 10(h) m and an air volume of about 4500 m^3 (including the gallery area and somewhat narrower scene than room).

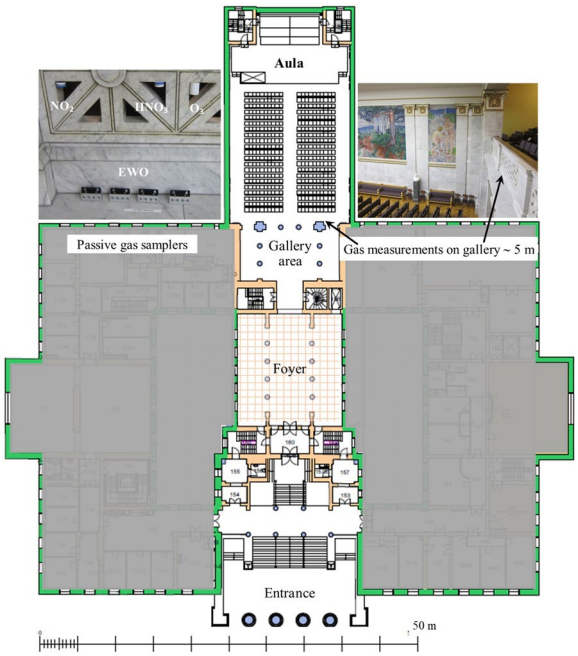


Fig. 3 The floor plan of the Oslo University Aula with the measurement location shown. The mounted passive gas samplers are noted. *EWO* early warning organic dosimeter (see text). Image source: University of Oslo

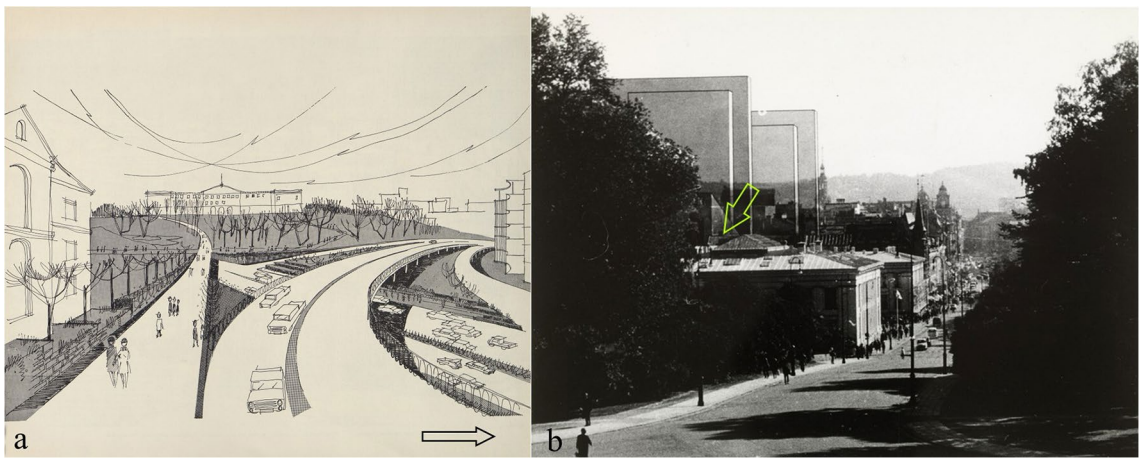


Fig. 2 Suggestions for the development of the area of the main promenade street, Karl Johan, in the centre of Oslo from the 1960s. The drawing (**a**) and photo (**b**) show opposite directions from the approximate same viewpoint. The location of the Oslo University Aula is marked with the arrows. Image sources: Oslo Byarkivet (Oslo City archive)/Oslobilder.no, and (**a**) [36, 37] and (**b**) [38]

Ventilation and air pollution transport to the paintings in the Oslo University Aula

The ventilation and infiltration of particles into the Aula is a main reason for the observed accumulated dark soiling on the paintings [33, 35, 39]. The cleaning of the sticky dark soiling from the unvarnished, fragile and water sensitive painting layers is a resource demanding conservation challenge [4, 33, 35, 40–42]. The soiling could not be fully removed by the cleaning [42], which was performed historically about every 14 years [33, 42–44]. Significant amounts of loose dust have at intervals also been removed from the paintings [33]. The possible deterioration due to reaction with gaseous pollutants is not immediately visible.

Mechanical ventilation with particle filtration was introduced in the Aula in the 1970s [33]. The exact year was not found in reports and this change was therefore set in this work to have happened in 1975. The Aula has however had significant infiltration of air besides the mechanical airflows, through the front door and foyer that have also on some days been kept open for the public, and other leaks in the historical building [45]. The indoor to outdoor concentration ratios (I/O ratios) of the air pollution will have depended on the ventilation regimes and fraction of naturally to mechanically ventilated air.

A major effort in the Aula renovation and paintings conservation campaign in 2009–2011 was to reduce the air transport of particles and pollutants to the paintings [46]. A new mechanical filtration system was installed [45]. Measures were taken to reduce the convection airflows along the walls and paintings. The overall ventilation rate in the Aula was reduced by Demand-Controlled Ventilation (DCV) [47]. The localized heating from under sofas along the walls was removed and most of it compensated with some degree of overheating (compared to the Aula) of the surrounding rooms including the attic. Water based underfloor heating was installed to give acceptable audience comfort at a lower room temperature. A stable slight temperature stratification has been observed (see below). All the paintings were thermally insulated with an underlying honeycomb structure to remove "cold bridges" [42] to the brick walls, which were found to result in downward induced airflow along and temperature induced transport and diffusion (thermophoresis) of particles to the paintings.

The rate of air pollutant transport along the paintings will depend on the airflow velocity including the turbulence intensity. As a comparison to the deposition velocities in Table 2 the approximate minimum average air velocity along the room surfaces (and paintings) was estimated to be from 0.3 to 0.4 cm s^{-1}, as follows. The average exchange time of the Aula air volume is today approximately 0.9 times per hour (or by volume, 4000 m^3 h^{-1}). The maximum possible capacity is 4.5 times per hour (20,000 m^3 h^{-1}) [48]. The ventilation is kept as low as possible to reduce airflows, but sufficient to keep a low overpressure to hinder particle/dust ingress. It is regulated by humidity, temperature, and carbon dioxide (CO_2) sensors. The ventilation is from inlets in the floor along the walls on the scene and in the hall, and from behind the seats for the public on the gallery. The outlets are on the edge of the glass roof in the hall (Fig. 1) and roofs of the gallery and scene. The gallery (Fig. 3) is somewhat enclosed from the hall and its paintings, and it could be expected that much of the ventilation air to the gallery leaves through its roof outlets. The airflow passing the paintings is expected to be mainly from the scene and hall outlets. The scene is narrower than the room and has a low podium (Fig. 1). The distance from the inlets to outlets on the scene is about 9.7 m, as the reported vertical distance between the inlets and outlets in the hall. The direct distance between inlets and outlets in the hall was reported to be 10.2 m. This gives (by using Pythagoras theorem) a distance along the two right-angled room surfaces (wall and roof) of 12.9 m, and air flow velocities of $9.7/0.9 = 0.3$ m h^{-1} and $12.9/0.9 = 0.4$ m h^{-1}.

Experimental and data

In addition to the derivations based on published data, a few experiments were performed to obtain data about the air pollution and the mechanical filtration efficiency in the Aula. A short description of the experimental methods and results are given below. The results from the experiments were used in the assessment of the modelling input data to the estimation of the indoor pollutant deposition (Sect. "Modelling data").

Experimental methods

Continuous (half-hourly) measurements of the air temperature (T) and relative humidity (RH) were performed with ClimaSpot (Profort engineering) loggers by three of the paintings (~3.5 m from the floor), and higher up on the gallery (~4.5 m from the floor), during 1 year (01/04–2014 to 31/03–2015) with a data coverage of 77%. The concentration of nitrogen dioxide (NO_2) was measured over one month (17/12–2019 to 17/1–2020), and of ozone (O_3) and nitric acid (HNO_3) over three months (17/01–2020 to 17/04–2020), with single passive badge diffusion samplers of the IVL type [49]. HNO_3 is a strong acid and its (low) value in air is interesting to report from a conservation perspective [26], although this result was not used in the estimations of the pollution deposition. In addition, the deteriorating impact of the environment was measured by so called MEMORI® EWO (Early warning organic)—PPO (poly-phenylene oxide) polymer film dosimeters [50] over the three months. The EWO films are spin coated on small glasses of dimension 1.0 cm × 0.5 cm × 0.1 cm. Four of them were exposed

in one of the aluminium holders seen in Fig. 3. They measure a photo-oxidizing response, which is compared with the deterioration risk of cultural heritage materials, in the MEMORI® system. MEMORI® EWO results should be interpreted as indications of (relative) risk in the context of conservators' evaluation of the specific situation of, the usually aged, cultural objects of interest [50–52].

Results are further reported of measurements of the content of water-soluble ions in the outer surfaces of particle filters mounted in the air inflow from outdoors and outflow from the Aula, in the mechanical ventilation system that was installed in the 1970s. The filters (HI-FLO, HFGS-10-F7) were reported to have an ePM1 filter efficiency of 60% according to ISO 16980 [53]. The filters had been mounted in the ventilation system for nearly 1 year (from 20.11.2007 to 9.10.2008) when they were measured. The ion content was determined with ion chromatography, and the indoor to outdoor fraction of the separate ions and its average for all the ions were calculated.

Experimental results

The climate measurements by the paintings showed annual temperature and RH averages of 19.6 °C and 40%, with 5 and 95 percentiles of 17.8 °C and 22.8 °C, and 25% and 59%. On the gallery the temperature was 21.2 °C, with 5 and 95 percentile values of 20.0 °C and 23.3 °C and the RH was 0.5% lower than by the paintings. This compares to a previously reported RH variation between 8 and 80% [40]. The measured concentrations of NO_2, O_3 and HNO_3 are shown in Table 1, together with the average of reported outdoor values over the same time periods, of NO_2 from seven urban measurement stations, and of O_3 from the one urban station in Oslo where it is measured [54], and the indoor to outdoor (I/O) ratios.

Figure 4 shows the results from the EWO exposures compared with risk levels for two materials relevant to the paintings, as assessed in the MEMORI® system.

It is seen in Fig. 4 that the response of the EWO in the Aula clearly indicated a risk for deterioration of sensitive organic materials and pigments, of relevance for the paintings.

Figure 5 shows the measured amounts of ions in the 2007–2008 ventilation filters and the I/O ratios. The bicarbonate was calculated from the ion-balance.

Table 1 Measured air pollution values in the Oslo University Aula and outdoors in the centre of Oslo in the winter 2019

Pollutant	Concentration (μg m^{-3})		
	Indoor (I)	Outdoor (O)	I/O
NO_2	18.1	30.7	0.59
O_3	25.3	53.5	0.47
HNO_3	0.03		

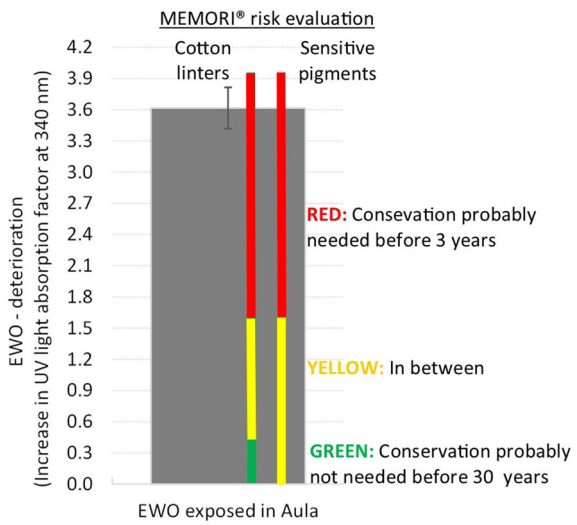


Fig. 4 EWO results and MEMORI® assessed risk levels for cotton linters and sensitive pigments. The standard deviation of four measurements is shown

For the outdoors, the large variation in the sodium and negative bicarbonate balance indicates (mainly) missing sodium ions to balance the measured chloride, sulfate, and nitrate. Indoors, there was little of these anions compared to the sodium which seems to be associated with the bicarbonate. It seems that the filtration removed most of these anions except the bicarbonate, which may have had an indoor source, possibly building dust, which might however also have been introduced from outdoors through other paths than the mechanical ventilation.

The measure of the indoor to outdoor ion fraction, $I/O_{average}$ in Fig. 5, does not directly represent the indoor to outdoor $PM_{2.5}$ fraction nor the filter efficiency in the removal of $PM_{2.5}$ or black carbon particles, which will have a different size distribution than the water-soluble ions. However, without a direct measurement of the $PM_{2.5}$ I/O ratio this value (of 0.2) is a good indication (to be discussed below).

Modelling data

The values used in the estimations of the historical indoor concentrations and deposition velocities of air pollutants in the Aula were obtained from assessment of the experimentally measured values in the Aula, including of the particle filtration efficiency after 1975, and from reported values from similar situations and laboratory measurements. They have the nature of a hypotheses that could be modified by future possible input from more parameter measurements or of the resulting deposition amounts. Deposition velocities vary with the surface humidity as affected by the air RH [56]. More humid situations will usually favour more adsorption and potentially increase

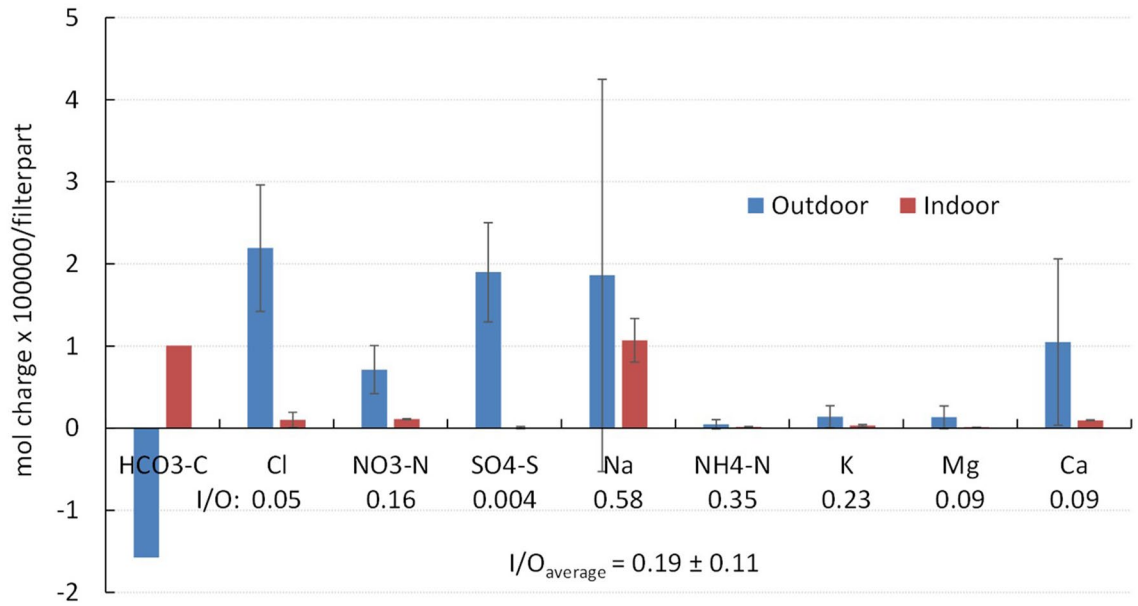


Fig. 5 Amounts of ions on outdoor and indoor filter surfaces in the mechanical ventilation system and the I/O ratio. Triplicate samples of the filter surfaces were analysed. The final average variation (±) was calculated by propagation [55] from the shown triplicate standard deviations of the measured outdoor and indoor amounts of the separate ions

the rate of deteriorating reactions. To represent the situation in the Aula, the calculations of the indoor deposition by Eq. (1) were made with deposition velocities of the gases assessed at the measured annual average RH 40% [56]. For the more reactive O_3 this value was in the upper range of other reported values [57, 58]. The outdoor concentrations (used to calculate the indoor concentrations, in Appendix Table 3, by the I/O ratios) were obtained from reports, of estimates from fuel consumption before 1960 and of measurements after 1960 [11]. The main primary nitrogen oxide, NO, emissions were assumed to have reacted with O_3 to NO_2, to give similar urban background values of NO_x and NO_2 before ventilation indoors in the Aula. The I/O ratios were assessed from the measured values in 2019 (Table 1) together with the reports of annual averages from monthly measurements with passive diffusive samplers in a large room in the (former) National Gallery of Oslo 100 m away from the Aula in each of the four seasons in 2010–2011 [59], and with other reports of I/O ratios of $PM_{2.5}$ [10, 60]. The historical situation and size of the different indoor sinks of $PM_{2.5}$ before and after installation of the mechanical filtration (~1975) were highly uncertain. Ranges of the O_3 deposition velocity and $PM_{2.5}$ I/O ratios were therefore applied (Table 2). Table 2 gives the values, explanations, and literature sources of the I/O ratios and indoor dry deposition velocities of the pollutants used in the deposition estimations.

A more detailed evaluation and rationale for the I/O ratios is provided in Additional file 1. The post 2009–2011 I/O ratio of $PM_{2.5}$ should be lower than it was since 1975 (Table 2), but measurements are needed to determine this. There is clearly considerable uncertainty, and historical variation, also in the other parameter values (than of O_3 and $PM_{2.5}$). To avoid excessive complexity, ranges for the calculations were however not suggested. The detailed derivations and uncertainties are discussed in Additional file 1, and more generally in the "Discussion" section.

Results

Figure 6 shows the approximated annual dry deposition since 1835, outdoors to buildings in the central urban background, adapted from [11], compared to indoors as represented by the Munch paintings mounted in the University Aula in Oslo since 1916. 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 maintenance and cleaning of façades and indoor surfaces (the Munch paintings), are not considered.

The temporal variation in the dry deposition of the pollutants, together, indoors and outdoors, is observed in Fig. 6 to be due to a maximum in the particle deposition in 1875 and then a subsequent gradual decrease, an increase in the SO_2 deposition until 1940 and then

Table 2 Expected indoor (Oslo University Aula) to outdoor (Oslo central urban background) concentration ratios and pollutant dry deposition velocities

Pollutant	Indoor/outdoor concentration ratio[a]	Indoor dry deposition velocity (cm s^{-1})
SO_2	0.23[b]	0.057[g]
NO_2	0.9[c]	0.004[h]
O_3	0.5[d]	0.021[i]–0.077[g]
$PM_{2.5}$ before 1975	0.5–0.8[e]	0.02[j]
$PM_{2.5}$ after 1975 (with mechanical ventilation filtration)	0.1–0.4[f]	0.02[j]

[a] See also Additional file 1

[b] Measured in the National Gallery of Oslo.

[c] Suggested probable value based on data from the National Gallery and the literature, allowing a slight reduction to the indoors.

[d] Measured in the Aula in the winter 2019.

[e] Based on [60] and the assumption that there will probably have been some reduction to the indoors.

[f] Based on evaluations of possible variations in the $PM_{2.5}$ I/O ratio from the measured average ion I/O ratio in the Aula (Table 1) and the considerable general uncertainty.

[g] Average of surface dry deposition velocity to paintings at RH = 30% and RH = 50% [56].

[h] A two times higher value (0.008 cm s^{-1}) was reported from measurements [56] at RH = 30% than from RH = 50–90% (0.004 cm s^{-1}) The value of 0.008 cm s^{-1} seems anomalous and it was decided to use the value of 0.004 cm s^{-1}, for the situation with RH = 40% in the Oslo University Aula.

[i] Obtained by modelling to measurements by [57, 58] report a typical value of 0.051 cm s^{-1} "based on studies by [61] and by [62]"

[j] Average experimental dry deposition velocity to the indoor building surfaces of the historic house museum of Apsley House in London [60]. Simulation of the mechanically ventilated room in the house (Waterloo Gallery) showed the same value [63]

a maximum 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 O_3 in the years of maximum NO_x emissions around 1970. The O_3 deposition was found to be a major part of the total indoor air pollution deposition since 1835. The total indoor deposition may have been the highest in the period of high outdoor SO_2 emissions in the 1960s, or from about 1875 to 1940 due to high ozone and particle deposition. This is, however, highly uncertain and will have depended on the historical wood combustion practises and technologies [11], and on the expected very variable infiltration and deposition of, especially, O_3 indoors. The displayed accumulated values reflect these trends. The fraction of the outdoor (to building façades) to the indoor (to the Munch paintings in the Aula) dry deposition probably increased from 5–12 in 1835, to 10–25 between 1900 and 1990 with the maximum of 20–25 in the 1960s, and then again decreased to about 4–12 in 2020. In 2020 the O_3 deposition was found to dominate the indoor deposition on the Munch paintings, with the NO_x, $PM_{2.5}$ and SO_2 deposition being a probable 4–13%, 2–7% and 1–5% of the O_3 deposition. The ranges are due to the assessed uncertainty in the indoor O_3 deposition. Figure 7 shows the estimated effect of the installation of particle filters after 1975 on the deposition of $PM_{2.5}$ to the paintings.

The figure shows a probable reduction in the deposition of $PM_{2.5}$ to the painting of 62% (50–80%) after the installation of the filters in 1975. It should be noted that in this case (Fig. 7) only the suggested I/O ratio limits were considered and not the most probable filtration efficiency (of 0.8). The larger reduction of 87.5%, from the upper uncertainty limit of the I/O ratio of 0.8 to the lower limit of 0.1, would only be possible if the filter efficiency was higher (than 0.8) (see "Discussion").

Discussion

The surface uptake of an air pollutant happens through a so-called quasi-laminar sublayer of more or less stationary air of thickness on the order of millimetres [34]. The slowest of the air transport, of the $PM_{2.5}$ to the surface by convection and diffusion through the sublayer, and the surface reactivity, will limit the deposition. With increasing airflow velocity up to the surface dry deposition velocity, the sublayer becomes narrower, the concentration gradient through it steeper, and the pollution transport to the surface increases, as more (a higher concentration of) pollution comes closer to the surface. At even higher airflow velocity above the surface dry deposition velocity, and constant room concentration, the surface uptake will be constant. Smooth and little reactive surfaces can quickly be saturated even at low airflows. However, indoor concentrations are usually below those outdoors, and increasing ventilation, and airflows, will increase the indoor concentrations, and the deposition to the surfaces (by Eq. (1)). Thus, to limit deteriorating surface reactions, it is essential to reduce the air pollution concentrations, their ventilation to the indoors, the pollution transport flow to below the (maximum) surface reaction rate, and to reduce this rate, as represented by the surface deposition velocity (v_s).

Concentrations

The outdoor concentrations estimated from fuel consumption before 1960 were reported to be considerably more uncertain than those from the dispersion modelling or measurements after 1960 [11]. There is then considerable additional uncertainty in the indoor concentrations due to estimation by the I/O ratios (see Additional file 1). Annual pollution measurement of, especially, O_3 and $PM_{2.5}$ in the Oslo University Aula are needed to validate the present (2020) concentrations (Appendix Table 3).

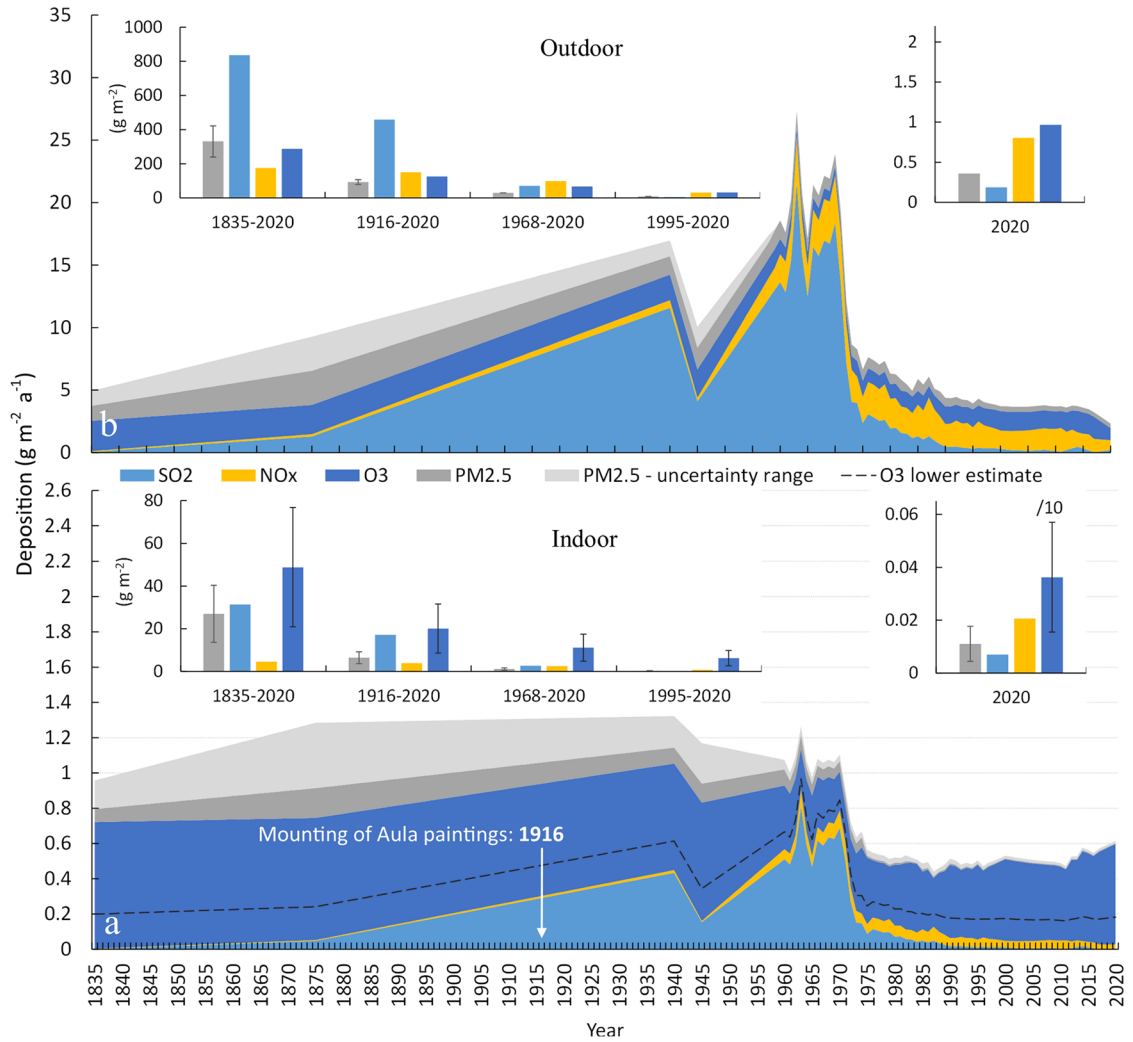


Fig. 6 Assessed indoor (**a**) and outdoor (**b**) [11] accumulated dry deposition (and not thus considering removal processes) of air pollutants to buildings in the urban background centrally in Oslo since 1835. The indoors is represented by the Munch paintings in the Oslo University Aula that were mounted in 1916. The ranges of the $PM_{2.5}$ and O_3 dry deposition represent the variation from estimates with the low to high values of the $PM_{2.5}$ emissions factors [11] and I/O ratios, and O_3 indoor deposition velocity (Table 2). The accumulated dry deposition and uncertainty ranges of $PM_{2.5}$, and of O_3 indoors, are reported from the middle of the displayed uncertainty ranges. Note the magnitude lower values of the indoor than outdoor dry deposition, and that the value of the indoor O_3 deposition shown in the bar chart for 2020 is divided with 10

Mechanical filtration and air pollution sinks

The mechanical air filtration in the Aula from 1975 reduced the I/O particle concentrations. The effect of the filtration depended on the fraction of the total ventilation air that passed the filters and the filtration efficiency. The natural and remaining mechanical particle ventilation (after filtration) subtracted the particle deposition in natural infiltration paths and to indoor surfaces, will have determined the I/O ratio due to ventilation (when not then considering other transport ways such as with visitors). It should be considered how the range of possible I/O $PM_{2.5}$ ratios with filtration (the I/O values after 1975 in Table 2) depend on the suggested natural (unfiltered) I/O ratio (the I/O values before 1975 in Table 2) and the filtration efficiency. The typical higher outdoor than indoor $PM_{10}/PM_{2.5}$ ratio indicates overestimation of the $PM_{2.5}$ filter efficiency by the ions I/O ratio measured in the Aula. On the other hand, the origin of some of the indoor particles, and ions, trapped in the filter, from indoor sources and/or natural particle transport and infiltration into the Aula indicates underestimation of the filter efficiency by the ions I/O ratio. Considering the opposing influences and the reported ePM1-60% (see Experimental) of the filters, a $PM_{2.5}$ filter efficiency of 80% (0.8) was used in the illustrating estimates below, based on

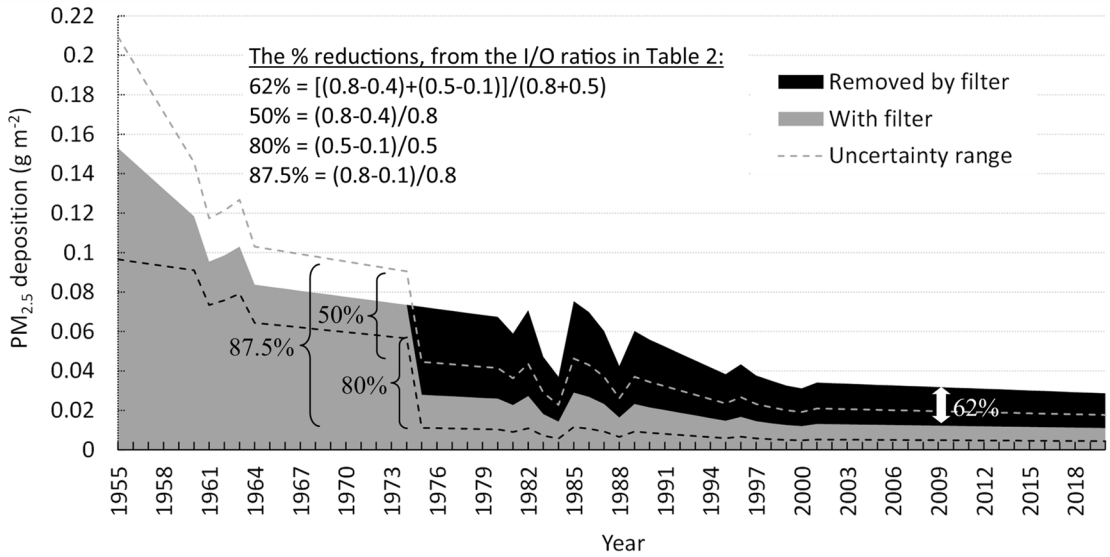


Fig. 7 Approximated deposition of $PM_{2.5}$ to the Munch paintings in the Oslo University Aula since 1955, with and (hypothetically) without particle filters installed in the ventilation system after 1975. The deposition is reported as the midpoint of the uncertainty range, to be comparable with the accumulated and year 2020 deposition values shown in Fig. 6. The uncertainty range represents the (similar span) variation of the I/O ratio scenarios, between 0.5–0.8 without filters and 0.1–0.4 with filters. The probable filter removal of 62%, and percentages removal between uncertainty range limits from before to after filtration are noted together with the calculation from the I/O ratios in Table 2

the measured ions I/O ratio of 0.2 (Fig. 5), but keeping the uncertainty in mind. Equations (2) to (5) give the indoor sink fractions of an outdoor pollutant that is ventilated and infiltrating into a building, depending on the filtration efficiency. The equations were derived from the simple mass balance of the sink fractions of the particles in the ventilation air (*F, d*) depending on the efficiency of the mechanical ventilation filters, in the steady state. More detailed derivations of a similar purpose can be found in for example [64, 65]

$$F_n = I_o - F_m, \tag{2}$$

$$F_m = \frac{(1 - I_o - d) \times (1 - f)}{f}, \tag{3}$$

$$F_d = (F_n + F_m) \times (d/(1 - d)), \tag{4}$$

$$F_f = 1 - F_n - F_m - F_d, \tag{5}$$

where I_o is the I/O ratio, F_n *and* F_m are the indoor fractions of the pollution transported in with the natural and mechanical ventilation, F_d is that deposited on the indoor surfaces, F_f is that captured by the filters in the ventilation system, d is the constant fraction of deposition to surfaces from the indoor air (in the steady state) and f is the constant filter efficiency. One should note here that the I/O ratio and fractions of pollution in the air will be larger on infiltration than exfiltration, the difference being the indoor deposition. Thus, the deposition happens from a concentration varying between that in the infiltration and exfiltration air, and the concentration and deposition calculated from the overall I/O ratio represents an average. Figure 8 gives the indoor sink fractions of $PM_{2.5}$, for the suggested I/O ratio scenarios in the Aula and a filter efficiency of 0.8, calculated from Eqs. (2) to (5).

Figure 8 shows the fractions of $PM_{2.5}$ that will have infiltrated naturally, still have been introduced through the mechanical ventilation system, that will have deposited on indoor surfaces, and the remaining part that will have been removed by the filters with a $PM_{2.5}$ removal efficiency of 0.8 installed in the mechanical ventilation system. The $PM_{2.5}$ fractions are shown for different possible I/O ratios below the lower (a, I/O ratio = 0.5) and upper (b, I/O ratio = 0.8) limits of the I/O ratio suggested for the situations without filtration before 1975. The amount of naturally infiltrated air (the "leakage") determines the position along the horizontal axis. It is seen in Fig. 8 that at the suggested filter efficiency (f) of 0.8, and I/O ratio of 0.8 before filtration, the lowest I/O ratio that could be reached with the filtration is 0.16 in a situation with no natural infiltration (the unfiltered I/O ratio $\times (1 - f) = 0.8 \times 0.2$). Figure 8 shows that the filtration in the Aula from 1975 (at f = 0.8) probably reduced the fraction of outdoor $PM_{2.5}$ that was ventilated indoors with

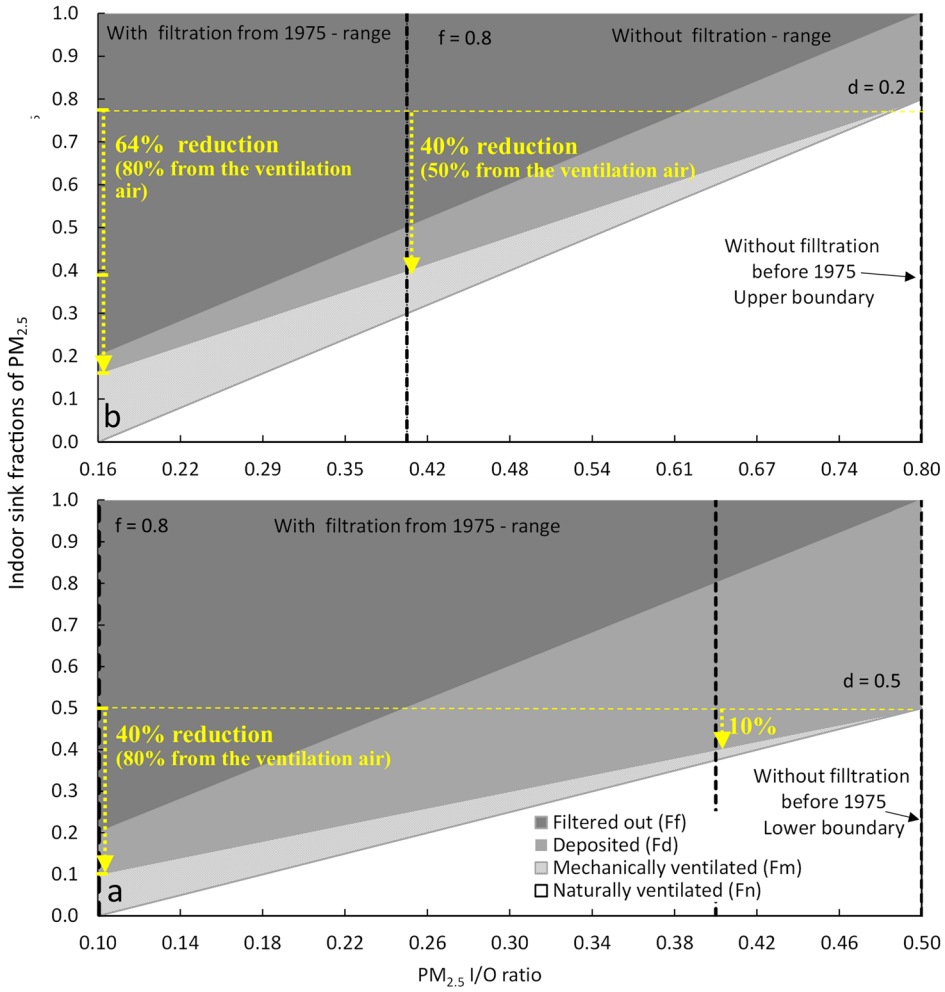


Fig. 8 Indoor sink fractions of $PM_{2.5}$ for filtration scenarios from the low (I/O = 0.5) (**a**), and high (I/O = 0.8) (**b**), I/O ratio scenario limits without filtration in the Aula before 1975, to the respective low (I/O = 0.1) and high (I/O = 0.4) I/O ratio scenario limits with filtration after 1975, calculated from Eqs. (2) to (5). The yellow arrows show the reductions in the fractions of outdoor $PM_{2.5}$ ventilated to the indoor air depending on the I/O ratios without filtration before 1975 (I/O = 0.5 (**a**) and 0.8 (**b**)) and with filtration after 1975 (I/O between 0.1 and 0.4)

between 10 and 64% depending on what the I/O ratio was without filtration (between 0.5 or 0.8) and became with filtration (between 0.1 or 0.16 and 0.4). The smaller $PM_{2.5}$ reduction of 10%, obtained by a change from the lower pre-filtering I/O ratio limit (of 0.5) to the upper filtering I/O ratio limit (of 0.4) seems however unrealistic. If assuming that the change was between the limits of the I/O ratio without filtration (0.8 or 0.5) to the lower limits with filtration (0.16 or 0.1) the reduction was between 64 and 40%. This is 80% (0.64/0.8 and 0.4/0.5) of the particles in the total ventilation air, rather than the 87.5% reduction shown in Fig. 7 for a change from an I/O ratio of 0.8 to one of 0.1. Thus, to obtain this reduction from the higher (possible) I/O ratio without filtration (of 0.8) to the lower suggested limit for the I/O ratio after filtration (of 0.1) a higher filter efficiency than 0.8 would be needed. The average probable reduction, of 62% (Fig. 7), is not affected by this consideration as this was calculated between the change from the high I/O limits from before to after filtration (0.8–0.4) and the change from the low I/O limits from before to after filtration (0.5–0.1).

Air pollution transport to the paintings

The deposition of air pollution ventilated into the Aula will be influenced by the air movements. If the transport of air pollution to indoors surfaces, such as the Aula paintings, is less than the surface reactivity (given by the deposition velocities in Table 2) the deposition could be less than estimated in this work. The average

non-disturbed laminar airflow velocity along Aula room surfaces was estimated to be about 0.3 – 0.4 cm s^{-1} (see Sect. "Ventilation and air pollution transport to the paintings in the Oslo University Aula"). These airflow velocities are about four to more than one magnitude higher, depending on pollutant, than the surface deposition velocities (Table 2). The low ventilation regime in the Aula thus probably reduces the deposition of pollution gases and $PM_{2.5}$ to the room and paintings, mainly by reducing the indoor concentrations, rather than the airflow transport to the surfaces.

There will however be variations from the average air velocity in the Aula, due to variations in the airflow path lengths along the room surfaces and residence time of the air, which can influence the deposition on the paintings. Variations can occur towards higher values due to mixing of compartment air and air circulation, and where there is disturbed air due to for example turbulence from air outlets, the room features and geometry, heat sources and gradients, and natural infiltration paths and visitors. Variations can occur towards lower airflow in possible pockets and zones of more stagnant air. There will be some, here undetermined, mixing of the inlet to outlet air between the three compartments, the scene, hall, and gallery. Little such mixed air probably flows along the paintings. The longer flow paths from the inlets to the outlets of such air could potentially result in somewhat higher (average) flow velocities. The airflow velocity is, generally, high(er) and often turbulent in the outlet ducts from ventilation systems, which can also induce turbulence along walls [66]. At low Reynolds numbers (Re) from the outlets, less than approximately 500, it has been found experimentally that the flow along the walls from outlet jets is probably transitional (turbulent to laminar) or laminar [67, 68]. Other reports [69] are of transition to turbulent flow along surfaces generally between $Re = 10^5$ and 3×10^6. The needed airflow rates at the outlets and their location can, potentially, increase the air mixing along and pollution transport to surfaces in the Aula even if the overall air exchange is low and, probably, usually mainly laminar. More detailed convection and turbulence measurements than was possible in this work would be of interest.

Deposition

Indoor dry deposition velocities vary much depending on the indoor air and surface characteristics. The uncertainty in the assessment for the Munch paintings (Table 2) is difficult to suggest. The surface deposition velocities could be considerably higher, than given in Table 2, to for example rough or soiled parts of the paintings. The surface deposition velocity may increase as the paintings get more soiled and deteriorated. New soiling particles might adsorb more easily and strongly to the established soiling than to the original painting surface. The soiling can enhance further soiling at an increasing rate. Thus, the regular cleaning and especially a tailor-made maintenance is important to reduce the accelerating damage [40]. What the temporal variation in the soiling rate may have been depending on the cleaning status of the paintings, is clearly an issue of interest.

Although the small and dark particles is the most critical, it is not the only concern. The total particle deposition will be larger than that of $PM_{2.5}$. Some amount of the $PM_{10-2.5}$ fraction will penetrate to the indoors by natural ventilation paths and deposit. A partial correlation between the outdoor an indoor $PM_{10-2.5}$ has been observed [60] that was stronger in the summer and for higher concentrations. This was explained by higher ventilation in the summer, and relatively more transport of coarser particles with visitors in the winter (which is not related to outdoor air concentrations). The larger extent of impaction and settling by gravitation of particles in the $PM_{10-2.5}$ range and above can lead to increased localized deposition [7].

In the National Gallery of Oslo, the indoor fraction of $PM_{10}/PM_{2.5}$ was measured to 1.5 [59]. This would imply, for the typical double deposition velocity of PM_{10} to $PM_{2.5}$ [70]), a three times higher mass deposition of $PM_{10,}$ than $PM_{2.5}$ to the paintings in the Aula (ignoring in this case the different deposition of PM_{10} and $PM_{2.5}$ to different surface orientations). This however probably better describes the historical situation before the installation of mechanical filtration in ~ 1975. The present (2020) deposition of PM_{10} is probably less than three times that of $PM_{2.5}$, estimated to 0.011 ± 0.006 g m^{-2} a^{-1}, but will also depend on the transport of particles into the Aula outside of the mechanical ventilation system. In three British museums the soiling rate and fraction of fibrous dust on the walls, was reported to be significantly higher up to about 150 cm on the wall, due to dust from the floor and visitors [71]. On the higher (than 3 m from the floor) mounted Munch paintings in the Aula (Fig. 1) deposited large particles from visitors and indoor activities, often of organic origin, are still observed [33, 35]. Thus, the total soiling rate is expected to be higher than that estimated for the $PM_{2.5}$, or even PM_{10}, fraction in this work. Still, the visual nuisance and deteriorating effect of the soiling by the smaller and expected darker $PM_{2.5}$ particles from combustion, are larger due to the particle's stronger adherence, deeper penetration into surface topography and pores, and more difficult cleaning [7, 35, 72, 73]. From values of the particle densities (and characteristics) and/or the soiling density, the expected developing surface coverage [74, 75] or soiling thickness could be suggested from the deposition rates.

The estimated effect of the particle filtration indoors in the Aula, of 62% (50–80%) reduction in the mass

deposition of $PM_{2.5}$, constitutes only a small fraction of the probable deposition before 1975 (Fig. 6a), but it will have resulted in a significant reduction in the development of the soiling since 1975 (Fig. 7). The renovation of the filtration system in 2009–2011 will have further improved the situation. If the present filter efficiency were known, or could again be measured, and PM measurements could be performed for a period including some (short) time without the particle filters installed, but otherwise under normal operating conditions, the fraction of the particles which still infiltrates naturally and the effect of the filtration in reducing the deposition could be determined.

The indoor deposition of O_3 may today be much higher than of other air pollutants (Fig. 6a). This result should be interpreted with caution as a risk indication and warning rather than a definitive risk. How much of the O_3 surface loss that will be due to its decomposition and how much due to chemical reactions, will clearly depend on the oxidizability of the surface material and to some extent on the indoor temperature, relative humidity (RH) and amount of surface adsorbed water [16, 76]. Homogeneous reactions are, in the situation with low, or no, expected indoor NO probably a small sink for the O_3 [57]. Although, some indoor loss of O_3 indoors by reaction with NO and formation of NO_2 is typical in cities in the summer [77, 78]. With the low ventilation in the Aula the residence time of any emitted VOCs (volatile organic compounds) and amount of their reaction products with O_3 could be higher than is common indoors. It seems important to further evaluate, including experimentally, the rates of and deterioration risks due to, O_3 deposition on the paintings, besides those of particles and NO_x. With access to annual values of the indoor concentration of O_3 and $PM_{2.5}$ and more information about possible reactions, of especially O_3, with the paintings, the indoor deposition, and present risk, due to these pollutants could be better assessed. For validation and comparison with the deposition estimates, experimental measurements of the soiling rate in the Aula are ongoing on canvas samples with white grounds [5] and Teflon filters [79] mounted on the wall close to two of the Munch paintings. It might also be possible to assess the present soiling amount on locations on the paintings or other surfaces in the Aula. Although, due to the past renovation campaigns, the frequent cleaning and maintenance, and many sensitive conservation concerns, this is challenging.

The models presented in this work are in the form generally and commonly used to describe indoor air pollution deposition. Their explanatory power depends on the correctness of the assessed historical values of the I/O concentration and the deposition velocities. Clearly, there is considerable uncertainty in these values and thus the estimated historical trends of the air pollution deposition to the Munch paintings. The estimates should, however, be regarded also as an example of a comparison case with the outdoors. We think this assessment provides useful values of the historical amounts and trends of the indoor sink fractions and mass deposition of air pollution to the paintings, that could be practically compared with site measurements.

Conclusion

The historical (1835–2020) dry deposition of the major air pollutants (SO_2, NOx, O_3 and $PM_{2.5}$) together, to the indoors as represented by the monumental Munch paintings in the Oslo University Aula (since 1916), was found over these years to have been from 4 to 25 times lower than outdoors. It reflected the outdoor deposition but varied less between the years, from about 0.3 ± 0.1 to 1.15 ± 0.3 g m^{-2} a^{-1}. The accumulated deposition since 1916 was found to be 43 ± 13 g m^{-2}, as compared to 107 ± 39 g m^{-2} in a similar situation since 1835. The removal processes, mainly by the regular cleaning, was then not considered. The $PM_{2.5}$ deposition increased to a maximum in 1875 and then slowly decreased. The total maximum in the 1960s was due to high SO_2 emissions from the combustion of fuel oils. The SO_2 deposition then decreased to a low value around 1990. The NO_x deposition had a maximum in the 1960s and 1970s, at the same time as SO_2, it became the largest from the 1980s and showed a clear decrease from about 2010. The O_3 deposition was reduced in the years of the maximum NO_x and total air pollution deposition. The O_3 deposition, and the $PM_{2.5}$ deposition before the 1960s, were found to be a relatively larger part of the total indoor than outdoor dry deposition. The indoor deposition of O_3 was found to vary from $16 \pm 9\%$ (in 1959) to $38 \pm 21\%$ (in 2020) of the outdoor dry deposition. Whereas in 2020 the outdoor dry deposition of O_3 and of NO_x were found to be about the same, an 18 ± 10 and 33 ± 19 times higher indoor deposition of O_3, than of NO_x and of $PM_{2.5}$, was found to the paintings in the Aula. The particle deposition is however the most critical observable effect on the paintings. The estimates indicate a 62% (50–80%) reduction in the deposition of $PM_{2.5}$ to the paintings after the installation of filtration in the mechanical ventilation system in 1975 and a present (year 2020) $PM_{2.5}$ deposition of about 0.011 (± 0.006) g m^{-2} a^{-1}. The renovation of the Aula in 2009–2011 may have reduced the deposition towards the lower end of this range. The variation in the deposition includes the assessed possible historical variations in the $PM_{2.5}$ concentration before 1960 and uncertainty in the I/O ratios of $PM_{2.5}$ and indoor O_3 deposition velocity. The uncertainty in the estimate for SO_2 and NO_x is considerable but was not suggested. The uncertainty is difficult to assess and, especially, the indoor concentrations

and deposition of O_3 are expected to vary much due the high reactivity of this pollutant. There seems to be mainly two present air quality concerns related to the deterioration risk of the Aula paintings. The soiling and physical and chemical alterations caused by the small mostly dark particles in the $PM_{2.5}$ range, and by the larger mass of bigger particles, is a continued concern. It is essential to continue the efforts to reduce the ingress in the Aula, keep the deposition low, further decrease its rate, and to implement innovative cleaning [72] and other possible protection measures. The other main concern is the deterioration risk due to the, potentially, relatively high amount of O_3 deposition (as compared to outdoors and other air pollutants), but also lower deposition of NO_x(NO_2). Experimental exposures of representative samples and/or mock-ups [80] will probably be needed to assess these risks, in addition to the validation of environmental assessments as provided in this work. The air pollution and soiling risk is a permanent attention.

Appendix

See Table 3.

Table 3 Pollution concentrations in the Oslo centre urban background, and indoors in the Oslo University Aula, used in the deposition modelling

Pollutant (μg m^{-3}), I = indoor, O = outdoor. i = interpolated											
	SO_2	NO_x	O_3	$PM_{2.5}$	$PM_{2.5}$		SO_2	NO_x	O_3	$PM_{2.5}$	$PM_{2.5}$
Year	I	I	I	I-low	I-high	Year	I	I	I	I-low	I-high
1835	0.1	1.5	30	12	37	1985	2.7	52	13	4.6	9.2
1840–1875	i	i	i	i	i	1986	2.2	47	15	4.3	8.5
1875	2.7	3.8	29	27	85	1987	2.7	63	11	3.7	7.4
1880–1935	i	i	i	i	i	1988	2.0	51	14	2.6	5.2
1940	24	13	25	14	43	1989	1.4	46	15	3.7	7.4
1945	8.5	5.8	28	17	53	1990	1.0	37	17	3.4	6.8
1950–1955	i	i	i	i	i	1991	0.9	38	17	i	i
1959				15	25	1992	1.0	43	16		
1960	28	46	15	14	23	1993	0.8	39	16		
1961	27	50	14	12	19	1994	0.8	41	16		
1962	32	52	13	12	19	1995	0.6	35	17	2.3	4.7
1963	45	74	10	13	20	1996	0.8	43	16	2.6	5.3
1964	33	58	12	10	16	1997	0.7	38	17	2.3	4.6
1965	26	49	14	i	i	1998	0.7	35	17	2.1	4.3
1966	34	61	12			1999	0.7	33	18	2.0	4.0
1967	33	58	12			2000	0.6	30	19	1.9	3.8
1968	35	68	11			2001	i	i	i	2.1	4.2
1969	35	69	10			2002	0.4			i	i
1970	38	77	9			2003–2004	i				
1971	28	81	9			2005	0.3				
1972	15	63	11			2006–2007	i				
1973	8.5	52	13			2008	0.5	35			
1974	8.2	43	15			2009	i	35			
1975	4.9	43	16			2010		36			
1976	6.4	53	13			2011	0.1	39			
1977	5.9	53	13			2012	i	30			
1978	5.3	52	14			2013		31			
1979	5.5	58	12			2014	1.0	23			
1980	4.0	48	14	4.1	8.2	2015	i	24			
1981	4.1	48	14	3.6	7.2	2016		26			
1982	3.3	43	15	4.3	8.6	2017	0.1	22			
1983	3.1	42	16	2.9	5.8	2018–2019	i	i			
1984	2.4	41	16	2.3	4.5	2020	0.5	16	23	1.8	3.5

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s40494-022-00673-x.

Additional file 1. Detailed evaluation and rationale for the selection of I/O ratios (in Table 1).

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Authors' contributions
TG developed the paper and made the estimations of the indoor air pollution concentrations and deposition. TG and TF planned and carried out the environmental site measurements assisted by persons acknowledged and did text editing. TF supplied the Aula site and Edvard Munch paintings conservation information. All authors read and approved the final manuscript.

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Availability of data and materials
All data generated or analysed during this study are included in this published article [and its Appendix and Supplementary information files], with references to the data sources.

Declarations

Competing interests
The authors have no competing financial interests.

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