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Published in:
Environment International

Link to article, DOI:
10.1016/j.envint.2021.107062

Publication date:
2022

Document Version
Publisher's PDF, also known as Version of record

Citation (APA):
Mainka, A., & Fantke, P. (2022). Preschool children health impacts from indoor exposure to PM2.5 and metals. *Environment International*, *160*, Article 107062. https://doi.org/10.1016/j.envint.2021.107062

Environment International 160 (2022) 107062

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Preschool children health impacts from indoor exposure to $PM_{2.5}$ and metals

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ARTICLE INFO

Handling Editor: Olga-Ioanna Kalantzi

Keywords:
Health impact assessment
Indoor air
USEtox
Kindergarten
Cancer
Non-cancer

ABSTRACT

To better understand the relation between children health and indoor air quality, we measured the concentrations of fine particulate matter ($PM_{2.5}$) and 11 metals (arsenic, cadmium, chromium, copper, iron, manganese, nickel, lead, antimony, selenium, and zinc) from air samples taken during both winter and spring, and focused on urban and rural area kindergartens of the Upper Silesia Region, Poland, typified by the use of fossil fuels for power and heat purposes. We combined related inhalation intake estimates for children and health effects using separate dose–response approaches for $PM_{2.5}$ and metals. Results show that impacts on children from exposure to $PM_{2.5}$ was 7.5 min/yr, corresponding to 14 μDALY/yr (DALY: disability-adjusted life years) with 95% confidence interval (CI): 0.3–164 min/yr, which is approximately 10 times lower than cumulative impacts from exposure to the metal components in the $PM_{2.5}$ fraction of indoor air (median 76 min/yr; CI: 0.2–4.5 $\times 10^3$ min/yr). Highest metal-related health impacts were caused by exposure to hexavalent chromium. The average combined cancer and non-cancer effects for hexavalent chromium were 55 min/yr, corresponding to 104 μDALY/yr, with CI: 0.5 to 8.0 $\times 10^4$ min/yr. Health impacts on children varied by season and across urban and rural sites, both as functions of varying $PM_{2.5}$ metal compositions influenced by indoor and outdoor emission sources. Our study demonstrates the need to consider indoor environments for evaluating health impacts of children, and can assist decision makers to focus on relevant impact reduction and indoor air quality improvement.

1. Introduction

Among compounds that influence indoor air quality (IAQ), the World Health Organization (WHO) emphasizes risks associated with exposure to particles arising from the combined effects of chemical composition and particle size. The particle fraction most strongly associated with negative human health effects is fine particulate matter ($PM_{2.5}$), since it penetrates deeply into the lungs and causes a variety of negative health outcomes (Fantke et al., 2014; Taner et al., 2013; Wichmann et al., 2010). Among non-organic compounds, metals comprise 1–2% of atmospheric $PM_{2.5}$ (Rogula-Kozłowska et al., 2014) but cause different health effects. Several metals detected in $PM_{2.5}$ show high toxicity and/or carcinogenic potential even at low concentrations, which includes arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), and nickel (Ni) (IARC, 2015). Some metals are essential for human development but cause acute adverse health effects at high concentrations, such as copper (Cu), iron (Fe), manganese (Mn), and zinc (Zn) (Carter et al., 1997; Majestic et al., 2007; Tong and Lam, 1998). Yet other metals, namely antimony (Sb) and selenium (Se) are classified as "possible carcinogen in humans" (group 2B) (IARC, 2015).

Health effects vary based on the metal oxidation state, affecting absorption, membrane transport, excretion, and toxicity at the cellular or molecular target (Huggins et al., 2000; WHO, 2006). Among populations susceptible to $PM_{2.5}$ exposure and related metal constituents, children pose a particular concern. They have higher inhalation rates per unit body weight than adults (U.S. EPA, 2002) and their lungs are in a developmental stage, which means inhaled pollutants can interfere with normal lung function development (American Thoracic Society, 2004; Kulkarni and Grigg, 2008). Furthermore, children have narrower airways compared to adults; in adult airways, $PM_{2.5}$ may cause mild irritation but may lead to more substantial obstructions in children (Bruce et al., 2013). The heavy metal absorption rates and hemoglobin sensitivities to these metals in children are much higher than for adults. Finally, long-term, low-level exposure to metals results in various

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https://doi.org/10.1016/j.envint.2021.107062
Received 28 September 2021; Received in revised form 11 December 2021; Accepted 21 December 2021
Available online 24 December 2021

metabolic and cognitive disorders, decreases in school performance, learning disabilities, neuropsychological deficits, decreased intelligence, behavioral and developmental impairment, and growth disturbances (Tong and Lam, 2000). Because metals accumulate in our body and affect our central nervous system, they may be deposited in our circulatory system, disrupt normal internal organ function, and act as cofactors in other diseases. As a result, there is an increasing global concern regarding childhood exposure to metals (Gouveia et al., 2018).

As indoor exposure often dominates overall human exposure to $PM_{2.5}$ (Fantke et al., 2017; Hodas et al., 2016) and children spend most of their time indoors, it is important to evaluate adverse health effects in children resulting from inhaling pollutants that occur in indoor air. For preschool children, the kindergarten building is the second most common indoor environment after home regarding chemical exposure. Buildings in Poland are typically ventilated naturally, resulting in poor IAQ from two sources. Approximately 40% of $PM_{2.5}$ comes from outdoor sources, particularly from combustion of fossil fuels used for heating and transport, and the remaining 60% comes from indoor activities (Fromme et al., 2008; Zwoździak et al., 2013). For the latter, studies link respiratory illnesses with indoor features, such as new painting and wall covers or gas appliances (Adaji et al., 2019). Regular renovations conducted during the summer (Mainka et al., 2017, 2015a) and kitchens using gas stoves are common in Polish kindergartens. Except for typical indoor $PM_{2.5}$ sources, such as cooking, kindergartens frequently suffer from (1) insufficient ventilation (especially in winter), (2) infrequently and incompletely cleaned indoor surfaces, and (3) a large number of children relative to the room area and volume, with a constant resuspension of particles from the indoor surfaces in the room (Fromme et al., 2008).

Many studies evaluate children health effects of ambient $PM_{2.5}$ (Bose et al., 2018; Hao et al., 2020; Liu et al., 2017; Sanders et al., 2018; Ward and Ayres, 2004). However, very few study exposure of children to indoor $PM_{2.5}$ and its constituents (Oliveira et al., 2016; Sánchez-Soberón et al., 2019), with no available studies that combine $PM_{2.5}$ and metal exposure in kindergartens. To address this gap, the present study aims to evaluate the health impacts on kindergarten children associated with exposure to $PM_{2.5}$ and metals contained with the $PM_{2.5}$ fraction in air samples taken from kindergarten classrooms. We thereby focus on the following objective: based on measured $PM_{2.5}$ and metal levels in spring and winter at four kindergartens located in the Silesia Province, Poland, we estimate inhalation exposure of different kindergarten groups to measured $PM_{2.5}$ and metal concentration levels to characterize the health impacts on children and provide policy recommendations for improving children health.

2. Material and methods

2.1. Experimental setup for measuring $PM_{2.5}$ and metal concentrations

Winter and spring campaigns (from 9th December 2014 to 23rd May 2015) were conducted at four kindergartens located near Gliwice, in the Silesia Province of southern Poland. The sampling locations, schedules, and instrumentation for $PM_{2.5}$ sampling and metals analysis have been previously reported (Mainka et al., 2017, 2015b; Mainka and Zajusz-Zubek, 2019). Briefly, indoor $PM_{2.5}$ samples were collected in classrooms of 'younger children' (on average between 3 and 4 years of age) and 'older children' (on average between 4 and 6 years of age) at each kindergarten. PM samples were collected by using of Dekati® PM_{10} cascade impactor (Finland) with the air flow of 1.8 m^3/h. This impactor provides samples for PM fractions of $<$ 1; 1–2.5; 2.5–10 and $>$ 10 μm. Polycarbonate filters (Nuclepore, Whatman Int. Ltd., Maidstone, UK; diameter 25 mm) were used to collect samples in the first, second and third stage of the impactor. In the fourth stage, PM was collected with the use of Teflon filters (Pall Teflo R2PJ047, diameter of 47 mm, Pall, Int. Ltd., New York, NY, USA), which deposits particles with diameters of $\leq$ 1 μm (Zajusz-Zubek et al., 2018). Only in kindergarten no. 3, there was a separate bedroom used for an afternoon nap, which was occupied by a mixed group of older and younger children. The height of the sampling head was at the breathing zone of the children (~0.8–1.0 m above the floor). Before and after sampling, the filters were conditioned (temperature 20 ± 1 °C, relative humidity 50% ± 5%) for 48 h and weighed using a microbalance (±1 μg; MXA5/1, RADWAG, Poland). After digestion in ultrapure (Sigma Aldrich TraceSELECTultra®) HNO_3 (8 cm^3) and H_2O_2 (2 cm^3) according to PN-EN 14,902 standard (PN-EN14902, 2010), the concentrations of 11 metals (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, and Zn) in $PM_{2.5}$ were determined using atomic absorption spectroscopy with an acetylene-air flame (Avanta PM) and a graphite furnace (Avanta GM; GBC Scientific Equipment Pty Ltd., Melbourne, Australia). Replicate analyses were performed to ensure reliability. The limits of detection for the method, found by analyzing blanks (clean filter substrates) were 0.3 ng/m^3 for As, 0.25 ng/m^3 for Cd, 3.1 ng/m^3 for Cr, 1.4 ng/m^3 for Cu, 5.8 ng/m^3 for Fe, 1.5 ng/m^3 for Mn, 0.25 ng/m^3 for Ni, 0.5 ng/m^3 for Pb, 0.45 ng/m^3 for Sb, 1.0 ng/m^3 for Se and 5.0 ng/m^3 for Zn. The accuracy and precision of the extraction protocol were checked using Standard Reference Material - SRM NIST 1649a Urban Dust and NIST 1648 Urban Particulate.

2.2. Approach for estimating exposure

In order to derive children exposure to $PM_{2.5}$, we translated the measured $PM_{2.5}$ levels in kindergarten classroom air into mass intake by all children in each classroom. The indoor air concentration of $PM_{2.5}$ in each kindergarten classroom, $C_{PM_{2.5}}$ [g/m^3], is directly derived from the measured $PM_{2.5}$ mass and sample air volume. From $C_{PM_{2.5}}$, the cumulative annual intake mass of $PM_{2.5}$ by all children in a given kindergarten classroom, $I_{PM_{2.5}}$ [g/yr], can be expressed as:

$$I_{PM_{2.5}} = C_{PM_{2.5}} \times \sum_a \left(\overline{BR}_{c,a} \times t_a\right) \times t_{tot} \times n_c \quad (1)$$

where $\overline{BR}_{c,a}$ [m^3/capita-hr] is the average individual breathing rate of child c in a given classroom during a specific activity a (sleep/nap, sedimentary/passive, lightly and moderately intensive) based on the US-EPA Exposure Factors Handbook (U.S. EPA, 2011), t_a [hr/d] is the daily time of a particular kindergarten activity, t_{tot} is the annual number of days children spend in kindergartens [d/yr], and n_c [capita] is the number of children in the classroom. With that, we can define a classroom-specific overall effective daily breathing rate, BR_{eff} [m^3/d], as $BR_{eff} = \sum_a \left(\overline{BR}_{c,a} \times t_a\right) \times n_c$.

Similar to $PM_{2.5}$, we translated the experimentally derived metal levels in kindergarten air into the intake of children per classroom to derive metal exposure levels. We distinguished between different metal oxidation states, since health effects may differ widely across oxidation states. Data on the total concentration of $PM_{2.5}$-bound metals collected in different parts of the world are provided in Supplementary Table S1. Studies on metal oxidation state contributions to $PM_{2.5}$ required Scopus literature screening. Relevant identified studies are provided in Supplementary Table S2, presenting key features of oxidation state distributions for 11 metals (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, and Zn) in $PM_{2.5}$. Chromium toxicity is attributed to its hexavalent state, its concentrations are often scaled by a factor of 1/6 (Srithawirat et al., 2016), while the mean mass percentage of Cr(VI) to its total content in $PM_{2.5}$ collected in the Upper Silesia region is ~30% (Widziewicz et al., 2016). The concentration of oxidation state s of metal m in indoor air for each kindergarten classroom, $C_{m,s}$ [μg/m^3] is derived as:

$$C_{m,s} = C_{PM_{2.5}} \times fr_m^{PM_{2.5}} \times fr_{s \in m}^{air} \quad (2)$$

where $C_{PM_{2.5}}$ [g/m^3] is the $PM_{2.5}$ concentration in room air, $fr_m^{PM_{2.5}}$ [μg/g] is the fraction of metal m in the $PM_{2.5}$ fraction determined as the ratio of measured $PM_{2.5}$ mass to measured metal mass in the kindergarten classroom air, and $fr_{s \in m}^{air}$ [μg/μg] is the fraction of oxidation state s of metal m in air obtained from literature studies. From $C_{m,s}$, the cumulative annual intake mass of a metal species by all children in a given kindergarten classroom, $I_{m,s}$ [μg/yr], is expressed as:

$$I_{m,s} = C_{m,s} \times \sum_{a} \left(\overline{BR}_{c,a} \times t_a \right) \times t_{\text{tot}} \times n_c \tag{3}$$

where $\overline{BR}_{c,a}$, t_a, t_{tot}, and n_c are defined as in Eq. (1).

2.3. Health impact assessment

To arrive at potential health impacts of children from exposure to $PM_{2.5}$ and metal constituents, we combine cumulative intake rates with dose–response information. We follow two approaches, using for $PM_{2.5}$ an exposure–response model for children effects, and for metals a dose–response model for effects in adults or in tested animals, since no children-related dose-response functions for metals are available to date.

The approach for $PM_{2.5}$ follows the methodology used to estimate the population attributable fraction from exposure to $PM_{2.5}$ used in the Global Burden of Disease (GBD, 2019). In the GBD integrated exposure–response model (IER), presumed toxicity differs with regard to inhaled mass and without specification of $PM_{2.5}$ composition, due to currently inconclusive evidence on differential toxicity for $PM_{2.5}$ (Burnett et al., 2014; Fantke et al., 2019).

In our study, the IER model is applied to acute lower respiratory infections (ALRI) in children under the age of five as relevant cause associated with $PM_{2.5}$ indoor air concentration above C_0 as the theoretical minimum-risk exposure level. With that, we define a relative risk RR, for any $C_{\text{PM}_{2.5}} > C_0$ (else, $RR(C_{\text{PM}_{2.5}}) = 1$), where the exposure concentration $C_{\text{PM}_{2.5}}$ (in g/m^3) is:

$$RR\left(C_{\text{PM}_{2.5}}\right) = 1 + \alpha \times \left(1 - e^{-\beta \times \left(C_{\text{PM}_{2.5}} - C_0\right)^{\delta}}\right) \tag{4}$$

where $(1+\alpha)$ is the maximum relative risk, β is the ratio of relative risk at low-to-high $PM_{2.5}$ exposures, and δ is the power of $PM_{2.5}$ exposure concentration. Coefficients α, β, δ and C_0 are averages across 1000 equally likely estimates for each coefficient (Burnett et al., 2014). From RR, we can define the attributable risk fraction for ALRI as $ARF = (RR-1)/RR$, which we combine with the current individual mortality, M [deaths/capita], due to ALRI in Poland in 2015 based on GBD data. Combining ARF with M yields a marginal, individual exposure–response factor, ERF [deaths/capita per μg/m^3], per increment of $PM_{2.5}$ concentration, ΔC, at the indoor $PM_{2.5}$ level $C_{\text{PM}_{2.5}}$:

$$ERF = \left(\frac{\left[ARF\left(C_{\text{PM}_{2.5}} + \Delta C\right) - ARF\left(C_{\text{PM}_{2.5}}\right) \right] \times M}{\Delta C} \right) \tag{5}$$

Next, we combine ERF with the classroom-specific overall effective breathing rate, BR_{eff} [m^3/d]. Then, we transform units to 10^{-9} kg/μg and 365.25 d/yr, and multiply the result by a severity factor, SF [DALY/death], linking disability-adjusted life years (DALY) to mortality in Poland in 2015 based on GBD data. With that, we yield a marginal effect factor per kindergarten classroom for ALRI effects in children due to $PM_{2.5}$ exposure, $EF_{\text{PM}_{2.5}}$ [DALY/kg], as (Fantke et al., 2019):

$$EF_{\text{PM}_{2.5}} = \frac{ERF}{BR_{\text{eff}} \times \frac{\text{kg}}{\mu\text{g}} \times \frac{\text{d}}{\text{yr}}} \times SF \tag{6}$$

Finally, we combine the annual $PM_{2.5}$ exposure mass ($I_{\text{PM}_{2.5}}$ from Eq. (1)), corrected for 10^{-3} kg/g, with effects per exposure unit to yield an impact characterization factor for annual exposure, $CF_{\text{PM}_{2.5}}$ [DALY/yr]:

$$CF_{\text{PM}_{2.5}} = \left(I_{\text{PM}_{2.5}} \times \frac{\text{kg}}{\text{g}} \right) \times EF_{\text{PM}_{2.5}} \tag{7}$$

The approach for metals follows the methodology proposed in the scientific consensus model USEtox (Rosenbaum et al., 2011) to evaluate health impacts from chemical exposure, differentiating between cancer and non-cancer effects. Due to the lack of epidemiological data for children for the various considered metals, we rely on average human data extrapolated from animal test studies (Fantke et al., 2018, 2021a). Resulting effects and impact characterization factors, hence, show higher uncertainties for metals than for $PM_{2.5}$. Effect factors for metals, $EF_{m,s}$ [incidence risk/kg intake], are derived from $ED50_{m,s}$ [kg/lifetime] as human lifetime dose for a population response of $\alpha_{\text{response}} = 50\%$, and combined with a generic severity factor of $SF = 11.5$ DALY/incidence for cancer effects and $SF = 2.7$ DALY/incidence for non-cancer effects (Huijbregts et al., 2005):

$$EF_{m,s} = \frac{\alpha_{\text{response}}}{ED50_{m,s}} \times SF \tag{8}$$

For cancer effects, $ED50_{m,s}$ correspond to $TD50_{m,s}$ as human lifetime tumor dose, which are presented in the Supplementary Table S3, and non-cancer $ED50_{m,s}$ are presented in Table S4. Such data are in priority derived from human-based tests when available, or otherwise extrapolated from animal test (Huijbregts et al., 2010). Human inhalation exposure test data for cancer effects are available for As, Cd, Cr(VI) and Ni, while extrapolations from oral exposure animal tests are applied for all other metals. Non-cancer effect data are evaluated for all metals, with human inhalation exposure test data available only for Cr(VI) and Mn, and extrapolations from oral exposure animal tests applied elsewhere. An age-adjustment factor for children between 2 and 14 years is used to scale cancer lifetime slope-factors to the considered exposure duration (US EPA, 2005). Finally, we combine the annual exposure mass ($I_{m,s}$ from Eq. (3)), correcting for 10^{-9} kg/μg, with cancer and non-cancer effects per exposure unit, and aggregate over all relevant metals to yield total impact characterization factors for the annual exposure to all considered metals, CF_{metals} [DALY/yr]:

$$CF_{\text{metals}} = \sum_{m,s} \left[\left(I_{m,s} \times \frac{\text{kg}}{\mu\text{g}} \right) \times \left(EF^{\text{cancer}}_{m,s} + EF^{\text{non-cancer}}_{m,s} \right) \right] \tag{9}$$

2.4. Exposure duration

To evaluate the exposure of children to $PM_{2.5}$ and metals, we used the average presence of children in each kindergarten based on information provided by the staff. On average, children spend seven hours a day in the building and attend kindergarten 200 days per year. $PM_{2.5}$ samples were collected only when children occupied the building. Separation into younger and older groups is based on the number of children and time-activity patterns. Younger groups of children include 3-year-olds as well as older children. For better comparability across exposure scenarios, we present results in terms of a year spent per child in each kindergarten group.

2.5. Assumptions and uncertainties

Uncertainties in our results consider both uncertainties related to measured air concentrations and uncertainty associated with health impact approaches. The standard deviation of effect factors for exposure to $PM_{2.5}$ (Fantke et al., 2019) and effect factors for exposure to metals (Rosenbaum et al., 2011) were used. While health effects from $PM_{2.5}$ exposure specifically account for an effect endpoint derived for children (age ≤ 5 yr) based on epidemiological studies, uncertainties associated with effects from metal exposure is primarily related to extrapolation from generic non-cancer human or animal toxicity test data. This was due to a lack of epidemiological studies that derive children-specific exposure–response information for metals.

Uncertainties in our results consider both uncertainties related to measured air concentrations and related exposure estimates, and uncertainty associated with health effect approaches. Uncertainties in the measured air concentrations mainly relate to measurement method, which varies across metals, and assuming that the study sites are representative for other sites and settings. We focused on spring and winter as kindergartens are closed most of summer, and assumed that measured indoor concentrations are representative of typical long-term pollution levels, which is supported by reported PM_{10} levels from outdoor air monitoring (see SI, Figure S2). In our analysis, we did not include exposure to $PM_{2.5}$ or metals during potential outdoor activities of the children, to most accurately link potential health risk to exposure to indoor concentrations.

Related uncertainties propagate into exposure estimates, where additional assumptions are made, such as assuming equal breathing rate and body weight across individual children and over the considered exposure period. Uncertainty in exposure estimates related to assumptions and measurement errors are typically within a factor two to six (Hodas et al., 2016).

Several additional assumptions have to be introduced to derive related health effect estimates, leading to considerable uncertainty, particularly for metals. We applied reported standard deviations of effect factors for exposure to $PM_{2.5}$ (Fantke et al., 2019) and effect factors for exposure to metals (Rosenbaum et al., 2011). Several sources of uncertainty are reflected in these standard deviations. First, health effects from $PM_{2.5}$ exposure specifically account for a health effect derived for children (age $\leq$ 5 yr) based on epidemiological studies (Cohen et al., 2017). In contrast, health effects from exposure to metals are based on extrapolations from chronic non-cancer human or animal toxicity test studies, including exposure route-to-route extrapolation for almost all metals in our analysis where inhalation effect data were effectively missing (Fantke et al., 2021a; Rosenbaum et al., 2011). In such studies, human and animal tests are performed on adults. This does not necessarily reflect children characteristics (including inter-individual toxicological susceptibility towards different pollutants, which can vary greatly) or limited children exposure duration (chronic test durations prescribe lifetime exposure assumptions), which can lead to over- or underestimation of related effect estimates. Furthermore, actual health effects are usually unknown in such studies, and linear dose–response models are assumed due to the lack of background exposure levels in children for individual metals and a general lack of non-linear models that apply to low-dose exposures.

Finally, loss of particle-bound metals due to interception, impaction, and gravitational settling processes in the human respiratory tract might be relevant and are usually not considered when effects are extrapolated from oral exposure tests. In these cases, effect results might overestimate bioaccessible metal fractions available in children that can effectively induce health effects, with bioaccessibility ranging from $<$ 1 to 93% depending on study conditions and metal (Kastury et al., 2017). This aspect is not relevant for $PM_{2.5}$ effects or for metals where we derived effect factors from inhalation test results, as body-internal loss processes are usually considered when directly linking inhalation exposure to measured health outcomes. With these assumptions, we expect health impact results that are more reliable and expected to be in the correct order of magnitude for $PM_{2.5}$, while being less reliable and typically overestimated by up to around two orders of magnitude for metals. This strongly reflects the lack of epidemiological studies to derive metal-specific exposure–response information for children via inhalation.

3. Results

3.1. Indoor air concentrations of $PM_{2.5}$

Average $PM_{2.5}$ concentrations are 69.7 μg/m^3 and 75.1 μg/m^3 during winter and spring, respectively and exceeded the annual limit of 20 μg/m^3 as established by the national regulation for air quality in Poland (Mainka et al., 2015b; Polish Journal of Laws, 2012). We find (Fig. 1) higher $PM_{2.5}$ concentrations in rural areas with an average level of 76.6 μg/m^3 relative to urban areas (67.2 μg/m^3). Inside both rural kindergartens (no. 3 and no. 4), higher $PM_{2.5}$ concentrations are observed during the winter. During the winter, the $PM_{2.5}$ concentrations in the bedroom are higher than in the classrooms of younger children. Increases in the bedroom $PM_{2.5}$ concentrations were connected to the bedcovers needed for the afternoon nap, and agree with results reported elsewhere (Ferro et al., 2004; Yen et al., 2019), noting that making the bed lead to significant increases of $PM_{2.5}$ levels in indoor air.

Among urban kindergartens, $PM_{2.5}$ concentrations are higher in older children classrooms (I) except during the spring for kindergarten no. 2. Assuming the physical activity of five-year-olds exceeds that of three-year-olds in all kindergartens, the process of $PM_{2.5}$ resuspension confirms the increased concentration levels in the classrooms of older children (I) (Mainka et al., 2018, 2015a). However, during the spring in kindergarten no. 2, the direct access of younger children (II) to the playground through the terrace door plays a dominant role for the increased $PM_{2.5}$ levels. Children were changing shoes inside the classroom, and the classroom was not equipped with an entry mat, which is recommended to remove most of the related dust and associated pollutants (Hall et al., 2003).

3.2. Indoor air concentrations of selected metals

Fig. 2 summarizes the concentrations of selected metals measured in the $PM_{2.5}$ samples collected in urban and rural kindergartens during winter and the ratio between winter and spring seasons. The lowest concentration was found for Se, while Fe had the highest concentration. The differences between the metal concentrations in $PM_{2.5}$ according to the urban–rural location varied strongly between seasons according to local emission sources (see SI: Section S-1). Significant differences between seasons are observed for As, Cd, Cr, Sb, and Zn. During winter, higher concentrations are observed for As, Sb, and Zn ($p < 0.03$). The average concentrations for winter and spring seasons were: for As, 5.7 and 0.9 mg/m^3, for Sb, 2.1 and 0.8 mg/m^3, and Zn, 233.7 and 196.5 mg/m^3. During spring, higher concentrations of Cd and Cr are observed as compared to winter ($p < 0.006$), and their average concentrations during spring and winter are 14.5 and 18.5 mg/m^3 for Cd, and 64.7 and 346.7 mg/m^3 for Cr, respectively.

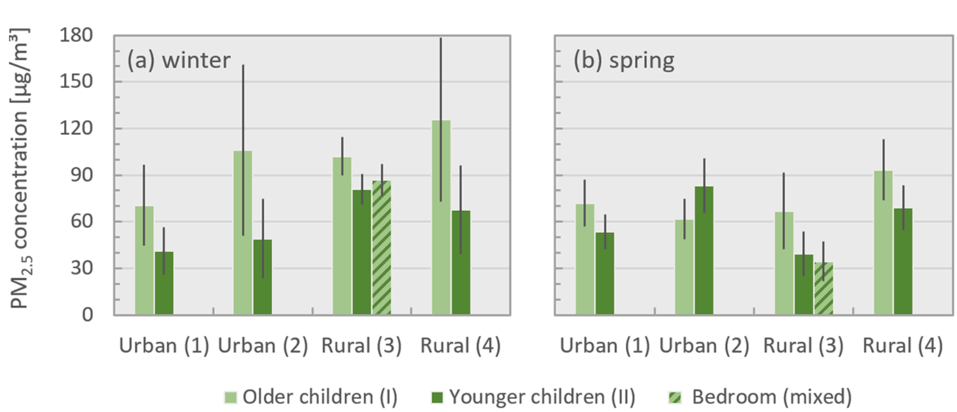

Fig. 1. $PM_{2.5}$ concentrations in kindergarten classrooms during (a) winter, and (b) spring seasons.

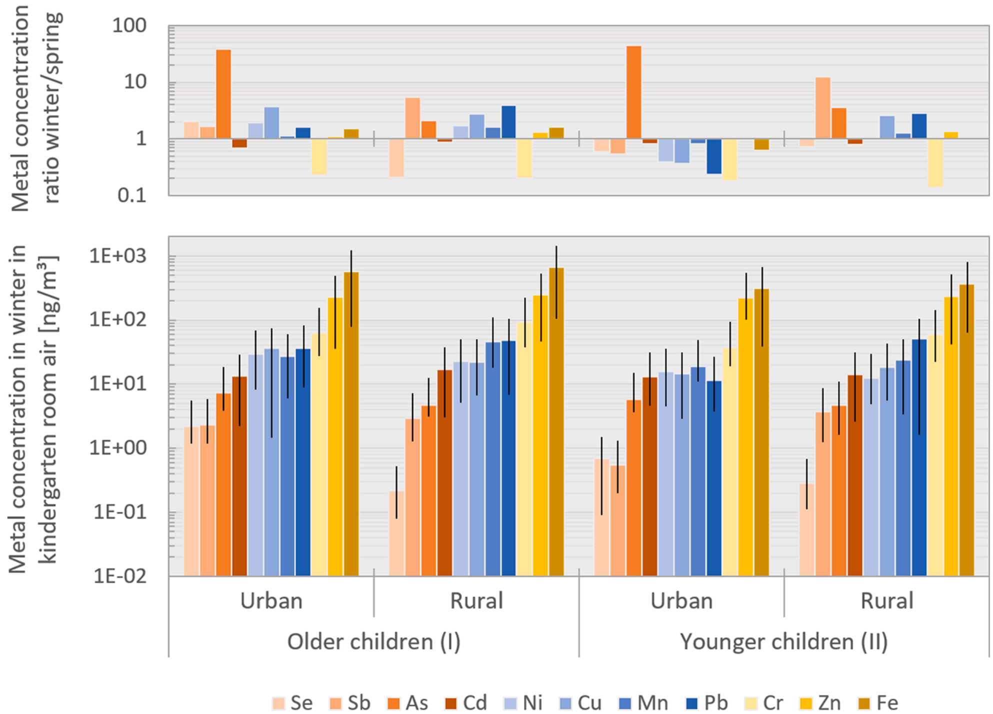

Fig. 2. Metal concentrations in $PM_{2.5}$ in kindergarten classrooms during winter and spring seasons.

The three elements with significantly higher levels in winter (As, Sb and Zn) are characteristic of fossil fuel combustion and production of non-ferrous metals (Zajusz-Zubek et al., 2019), which can migrate indoors after being attached to outdoor $PM_{2.5}$. Pesticide use results in arsenic emission (Šerbula et al., 2010), while refuse and sludge combustion emits Sb (Tylenda et al., 2014), and Zn comes from tires, motor oil, and the use of motor vehicle brakes (Latif et al., 2014). Research performed in Polish schools underscores the high infiltration of $PM_{2.5}$-bound Zn and Pb from outdoor emission sources such as coal combustion or vehicular emission (Zwoździak et al., 2013). Pearson correlation coefficient analyses point to positive correlations between As and Zn (0.62) as well as Sb and Zn (0.50) in $PM_{2.5}$ collected inside schools. Moreover, a principal component analysis (PCA) of outdoor $PM_{2.5}$ samples (Mainka et al., 2015b) points to common sources of As and Zn (defined as mixed anthropogenic sources with predominantly coal combustion for domestic heating purposes), while Sb in ambient air is primarily related to a variety of vehicular emissions.

The correlation coefficient (0.61) points to common sources of Cd and Cr – incineration of rubbish and sewage sludge (Zajusz-Zubek et al., 2019). According to the PCA, Cd, and Cr in ambient air come from the component characteristic for re-suspension of soil dust (Mainka et al., 2015b). The fact that Cr levels are approximately six times higher during spring and that residents complained about the dumping of sewage sludge on rural agricultural fields that took place at this time suggest that this may have been the primary Cr source related to indoor levels.

Fig. 3 presents the distribution of each metal in $PM_{2.5}$ according to oxidation state (SI, Table S2). The highest median amount in $PM_{2.5}$ is found for Fe(III) (3879.3 µg/g), while the lowest for Sb(III) (1.4 µg/g).

3.3. *Exposure to $PM_{2.5}$ and metals in the $PM_{2.5}$ fraction*

In every group of children attending kindergarten, the average annual $PM_{2.5}$ intake is 1.8 ± 0.6 g/yr and the corresponding daily intake vary from 4.9 to 15.2 mg per day per classroom. The daily $PM_{2.5}$ intake is higher for older children (10.9 ± 2.7 mg/d and 7.2 ± 2.1 mg/d per classroom for older and younger children, respectively). Considering the number of children present in classrooms during measurements (14–22 children), an older child inhales on average 0.6 ± 0.2 mg of $PM_{2.5}$ per daily occupancy at kindergarten, comparable to a younger child with a 0.4 ± 0.1 mg/d average daily intake (for details, see SI, Table S5). Higher daily intakes for older children (I) are observed for As, Cd, Cr, Fe, Mn, Sb, and Se; however, the difference was statistically significant (p = 0.03) only for Mn (other metals p = 0.11–0.67). For Cu, Ni and Pb, daily intakes are comparable for both groups of children (p = 0.89–0.96).

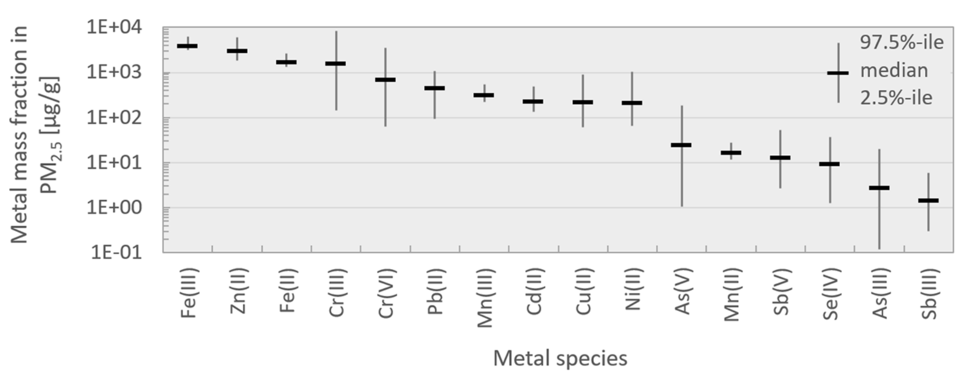

Fig. 3. Metal mass fractions (in µg/g $PM_{2.5}$) in air of kindergartens across seasons and classroom settings.

3.4. Health impacts due to indoor $PM_{2.5}$ and metal exposure

Linking exposure to $PM_{2.5}$ inside the kindergarten classrooms with relevant health effects yielded an average impact of 14 μDALY/yr (95% CI: 0.6–310) across classrooms, corresponding to 7.5 (95% CI: 0.3–164) minutes of healthy lifetime lost per year (min/yr). The difference between older (I) and younger (II) children is not significant, (95% CI: 0.3–162 and 0.4–167 min/yr, respectively).

For evaluating the health effects associated with exposure to metals in the $PM_{2.5}$ fraction, we distinguished cancer and non-cancer effects. Cancer effect data are available and were calculated for As, Cd, Cr(VI), Ni, and Pb. Test data for Cr(III), Sb, Se, and Zn show no clear carcinogenic effects, while cancer test data for Cu, Fe, and Mn are not available. Non-cancer effect data were evaluated for all metals. Ranking metals according to their increasing cumulative incidence risk for carcinogenic effects, we obtain Cr(VI) > Cd > As(V) > Ni > As(III) > Pb. For non-carcinogenic effects, cumulative risk per year decrease in the following order: Cr(VI) > Pb > Mn(III) > Cd > Fe(III) > Zn > As(V) > Fe (II) > Mn(II) > Se > As(III) > Sb(V) > Ni > Sb(III) > Cu > Cr(III).

Fig. 4 presents the annual lifetime loss for combined cancer and non-cancer effects based on older/younger children, season, and location. The older children (I) group showed slightly higher impacts. During winter, the highest lifetime loss is caused by As, particularly in urban areas. Highest daily impacts in spring, particularly in rural areas, are due to Cr(VI) exposure. The average combined cancer and non-cancer effects for Cr(VI) are 55 min/yr, corresponding to 104 μDALY/yr. Dominated by Cr(VI), lifetime losses were highest during the spring in rural areas and lowest in winter at urban sites.

When comparing the approaches for estimating health impacts from $PM_{2.5}$ and from metal exposure, the difference is substantial yet with overlapping uncertainty ranges (Fig. 5). For both groups of children, $PM_{2.5}$ inhalation during one year spent in kindergarten resulted in the loss of approximately seven minutes of healthy lifetime per year or 13.3 μDALY/yr (7.4 and 7.6 min/yr for older and younger children, respectively, corresponding to 14.1 and 14.4 μDALY/yr). Compared to that, metals contained in the $PM_{2.5}$ fraction in kindergarten air cause higher lifetime loss per group of children (95% CI: 0.2–5.0 × 10^3 and 0.2–4.1 × 10^3 min/yr for older and younger groups of children, respectively). There was a statistically insignificant difference between averages per children group (84 and 68 min/yr, corresponding to 160 and 129 μDALY/yr per children group, $p = 0.54$). However, a significant difference is observed between the spring and winter seasons. Combined exposure to metals during winter cause a loss of 41 lifetime minutes per year and children group (79 μDALY/yr) and a four-fold higher loss 111 (210 μDALY/yr) during spring ($p = 0.0005$). The averages translate into 2.3 and 6.2 min/yr (4.4 and 11.7 μDALY/yr) per child during the winter and spring, respectively, so during one year in a kindergarten located in a region based on fossil fuel combustion, each child loses, on average, approximately 4 min of healthy lifetime per year (8 μDALY/yr). During winter, the highest lifetime loss was caused by As, particularly in urban areas. As is characteristic for coal and solid fuels combustion and in kindergartens it mainly infiltrates from highly polluted ambient air. In contrast, highest daily impacts in spring, particularly in rural areas, are due to Cr(VI) exposure. The source of Cr is mainly the agricultural utilization of sewage sludge, of which residues can be transported indoors by children via for example playing in kindergarten backyards.

4. Discussion

4.1. Pollution concentrations

The problems of high $PM_{2.5}$ levels exceeding WHO daily and annual recommendations (25 and 10 μg/m^3, respectively) in naturally ventilated buildings and related possible health effects on kindergarten children agree with findings from other studies (see SI, Table S6.). In our study, rural kindergartens yielded higher concentrations than urban sites due to the use of low-quality coal, biomass, or even waste (PET bottles, clothing, furniture, etc.) for heating during the winter in Poland. Rural areas feature many more individual heating sources and often show lower air quality than urban areas (Mainka et al., 2015b). In contrast, inside Swedish preschools equipped with mechanical ventilation, $PM_{2.5}$ concentrations were significantly lower, between 3.2 and

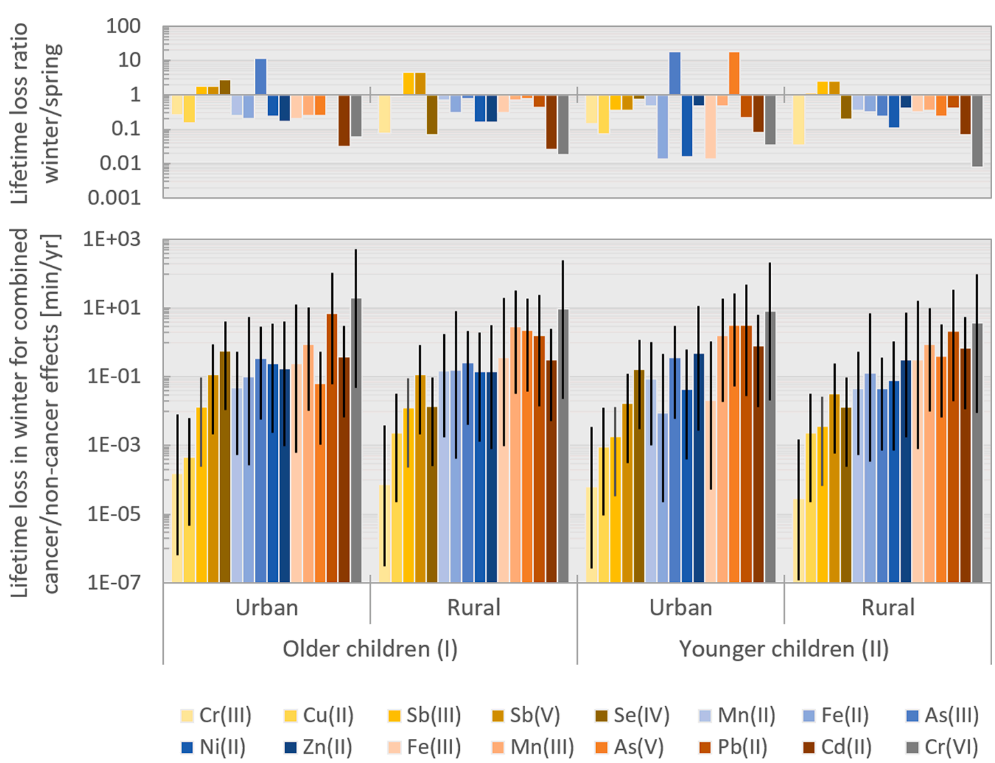

Fig. 4. Annual lifetime loss [min/yr] including cancer and non-cancer effects from exposure to metals in $PM_{2.5}$ depending on season and location.

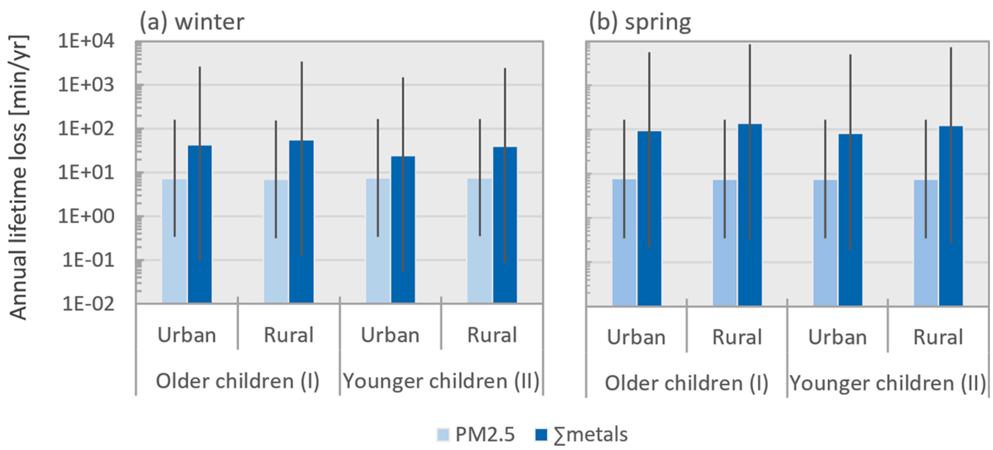

Fig. 5. Annual lifetime losses [min/yr] from exposure to $PM_{2.5}$ as compared to cumulative exposure to the loading of metals in the $PM_{2.5}$ fraction inside kindergarten classrooms as a function of the group (older/younger) and season a) winter, b) spring.

9.3 μg/m^3 (Wichmann et al., 2010). $PM_{2.5}$ samples in the present study were collected only when children were present; their physical activity is most likely the cause for high $PM_{2.5}$ levels, especially on the carpet placed in every classroom. For comparison, the effect of children jumping on the bed may result in $PM_{2.5}$ levels above 350 μg/m^3 (Yen et al., 2019). In this case, the use of an air purifier can improve indoor air quality substantially. For example, Gayer et al. (Gayer et al., 2018) reported the average air purification efficiency of 40% of various air purifiers tested in four selected kindergartens in Warsaw, Poland. The authors recommended running those devices continuously in all rooms, in particular during the heating season when outdoor $PM_{2.5}$ concentrations were high. More efficient but expensive and work-intensive methods include modernizing ventilation systems.

When comparing metal concentration levels with other kindergartens, there is shortage of data on metal concentrations in $PM_{2.5}$. Researchers often encounter problems accessing kindergartens, and installing the necessary measuring equipment to avoid disturbances during measurements and to limit the curiosity of children (Mainka et al., 2015a). Hence, metal levels in kindergartens are often determined using the dust embedded on horizontal surfaces and the floor (SI, Table S7). Compared with $PM_{2.5}$ indoor air metal concentrations (SI, Table S6) for As, Fe, Mn, Pb, and Zn, our results agree with results from schools in Wroclaw, Poland. For Fe and Sb, our levels are comparable with results from Spanish schools, while our concentrations of Cu and Se fell within the range reported for schools in Chile. For Cd, Cr and Ni, our results exceeded other indoor environments but the levels are comparable to ambient air results in Chinese industrial regions (SI, Table S1).

4.2. Children intake exposure levels

$PM_{2.5}$ intake is ~ 2 g/yr per group of children, which corresponded to approximately 100 mg of $PM_{2.5}$ per child annually. Among metals with extrapolated non-carcinogenic effects, the highest intake (>3.0 mg/yr) is observed for Fe(III) > Zn(II) > Fe(II) > Cr(III). For Zn, only exposure to high doses comes with toxic effects for most people, making acute zinc intoxication a rare event (Plum et al., 2010).

Intake of Cr(VI) exceed all other carcinogenic metals (1.5 mg/yr; 8.4 μg/d for older children). Studies from Europe and Mexico report daily Cr intakes of 30–35 μg/d in adults (Langård and Costa, 2014); hence, our results (Cr(VI) + Cr(III) ≈ 25 μg/d) are in accord with cumulative exposure levels, although we do not consider a source other than inhalation exposure. Intake levels of Cr reported here may lead to several health effects such as nasal septum mucous membrane perforations (nasal itching and soreness) and impaired respiratory functioning including acceleration of asthma (Al Osman et al., 2019).

Other carcinogenic metals that exceeded the intake level for children (2 μg/d) are Cd, Ni, and Pb, comparable with the load in a cigarette containing up to 2 μg of Cd (Nordberg et al., 2015), 0–0.51 μg of Ni (Klein and Costa, 2014) and 3–12 μg of Pb (Skerfving and Bergdahl, 2014). Inhalation of Cd during smoking is ~ 10% and assuming a 50% uptake of inhaled Cd, a daily uptake of 2 μg Cd corresponds to 20 cigarettes (Nordberg et al., 2015).

Despite the low daily intake value (<1 μg/d), As(V) contributes substantially to the daily lifetime loss, particularly during the winter due to emissions from heating systems. Arsenic exposure generates free radicals and changes the methylation state of cellular DNA, which is associated with several types of cancer such as skin, lung, and urinary bladder. Moreover, As(V) reduces *in vivo* to As(III), which comes with increased toxic effects (Sanchez-Rodas et al., 2012).

4.3. Children health impact estimates

Considering the $PM_{2.5}$ concentration inside the kindergarten classrooms examined, related health impacts led to an average lifetime loss of approximately 7.5 min per year (14 μDALY/yr) regardless of the age group. In contrast, metal exposure in the $PM_{2.5}$ caused an approximately 10-fold higher annual lifetime loss in children. Considering children attending kindergartens in the Upper Silesia region (145,152 children in 2014/2015), kindergarten children in the region lose ~ 42 days of healthy life per year due to ALRI-related effects considered in the GBD study series as $PM_{2.5}$-related causes related specifically to children. Compared with extrapolated age-unspecific carcinogenic and non-carcinogenic effects of the 11 metals studied in the $PM_{2.5}$ air fraction, the health impacts on children in the region result in the loss of approximately 400 days of healthy lifetime.

This seems surprising, as the cumulative impacts of all $PM_{2.5}$-related chemical constituents should not exceed the impacts associated with $PM_{2.5}$ as a whole. However, this assumption only holds when using children-related exposure–response information from epidemiological studies across both $PM_{2.5}$ and the metals studied, the latter of which is currently unavailable. Instead, exposure–response information for certain metals is available for specific, age-independent causes, such as respiratory effects (laryngitis, tracheae bronchitis, rhinitis, pharyngitis, shortness of breath, chest sounds, nasal congestion, and perforation of the nasal septum), cardiovascular abnormalities, anemia (normochromic normocytic, aplastic and megaloblastic), and leukopenia (granulocytopenia, thrombocytopenia, and myeloid, myelodysplasia) associated with exposure to As or Pb (Lim et al., 2012; Mandal and Suzuki, 2002). Selected studies link specific effects in children (e.g. IQ loss) to environmental exposure, based on directly linking blood concentrations to effect prevalence (Al Osman et al., 2019; Lanphear et al., 2005). To make use of such information, relevant exposure routes, as well as blood concentration levels from intake exposure, are required as conducted in other studies to compare biomonitoring and modeled exposures (Csiszar et al., 2017). Overall, since children-related epidemiological data associated with inhalation exposure are currently missing for most of the metals studied, effect data used in the present study for metals came from adults or were extrapolated from animal test studies. This introduces significantly higher uncertainties as compared with effect data for $PM_{2.5}$, which likely overestimates the associated lifetime loss due to metal exposure.

4.4. Limitations of the presented approach

We focused on assessing indoor air quality, since children spend most of their day indoors, including kindergartens. Although the measurements were performed during 2014 and 2015, the situation in the region including $PM_{2.5}$ concentrations and kindergarten occupancies are consistent with current exposure conditions. Poland is located in a moderate climate zone where spring is similar to autumn, and both seasons (spring and autumn) cover 50% of a kindergarten year with the rest occurring during winter. The measurements were performed during colder months, since in summer kindergartens in Poland remain mostly closed.

Among the main limitations of our study is limited pollutant coverage, i.e. we do not include other $PM_{2.5}$ compounds that may affect vital children body organs, such as mercury and polycyclic aromatic hydrocarbons (PAH). Another simplification is the assumption that the entire regional kindergarten children population is exposed to the same average pollutant concentrations. Moreover, our study focuses on inhalation as a major route of entry for small particles.

The USEtox model currently relies on linear dose–response models for non-cancer effects, mainly due to lack of background exposures and population group-specific vulnerability. With that, effect factors are scaled to reflect 3 years kindergarten exposure.

4.5. Future research needs

Public demand to improve the educational achievement of children is strong. For kindergarten children, there is a general shortage of data on relationships between air pollutants and the possible health effects of kindergarten-age children. More specifically, children-specific health effects from exposure to individual metals (including metals with different oxidation states) remain largely underdeveloped, and future studies should focus on closing this important data gap to reduce uncertainties associated with related effect factors, building for example on increased uptake of digitalization methods for characterizing chemical exposure and risk (Fantke et al., 2021b).

One promising approach to evaluate exposure is biological monitoring – an important tool in the prevention of diseases related to those exposed chemicals on a regular basis, particularly when multi-route exposure (inhalation, dermal, ingestion) or abnormal exposure takes place (Oliveira et al., 2017).

Future work should focus on further improving our understanding of toxicity effects particularly for children from indoor exposure, in support of building an environment that is sustainable in absolute terms for current and future generations (Fantke and Illner, 2019). The baseline of a health risk assessment for indoor environments could provide the necessary information upon which policymakers, as well as building managers, can make priority decisions or take remediation actions (Bruinen de Bruin et al., 2022). Another strategy worth exploring is assembling an electronic database to accumulate and preserve data and results from health risk assessment case studies in educational buildings. Such a collective experience in indoor risk estimation could become an invaluable resource for characterization and exposure assessments in indoor spaces. Such a database guides future risk assessment and provides accurate data for use in model testing.

We strongly suggest to update the dose–response model in USEtox, in line with global recommendations proposing non-linear dose–response models (Chiu et al., 2018; Fantke et al., 2021a), which needs to be developed also for metals. We further recommend that future studies also provide metal effect information for children a specific health endpoint in order to move away from the need to use generic effect data.

5. Conclusions

We provide insights into potential risks of children associated with inhalation exposure to $PM_{2.5}$ and several metals in kindergarten air. Our results indicate that season and location influence $PM_{2.5}$ and metal concentrations, with fossil fuel and solid waste combustion as important sources in winter and secondary emissions from sewage sludge amendment in agriculture as relevant source in spring. When combining measured children exposure estimates with health effect information, our results illustrate on the one hand that exposure to metals in the $PM_{2.5}$ air fraction can be a substantial contributor to overall impacts on children from inhaling kindergarten air. On the other hand, our analysis also suggests that effect estimates for metals show large uncertainties. These uncertainties emphasize the need for improved dose–response models and underlying data for individual metals, differentiated by cause and between children and adults as well as between exposure routes, in order to derive impact estimates that are better aligned between approaches for $PM_{2.5}$ and approaches for metals. Overall, our study contributes to a better understanding of exposure and related risk levels of kindergarten children in Poland, but also highlights the need for broader monitoring to allow generalization beyond our studied locations, and the need for improved dose–response modelling, especially across the wider range of relevant metals. With that, our study provides initial recommendations for policy makers to reduce exposure of children to air pollutants in kindergarten air, and for the research community to improve health impact assessment for children.

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.

Acknowledgment

This work was supported by the Safe and Efficient Chemistry by Design (SafeChem) project, funded by the Swedish Foundation for Strategic Environmental Research (grant no. DIA 2018/11), and by statutory research by the Faculty of Energy and Environmental Engineering, Silesian University of Technology. A. Mainka also expresses gratitude to the National Agency for Academic Exchange of Poland (under the Academic International Partnerships program, grant agreement PPI/APM/2018/1/00004) for financial support of the internship at the Technical University of Denmark in 2019.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2021.107062.

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