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Oggetto: Impianto di coincenerimento rifiuti della società Laviosa Chimica Mineraria S.p.A. stabilimento di Villaspeciosa, Località Perda Bianca". Procedimento di valutazione di impatto ambientale (V.I.A.) "ex post". D.Lgs. 152/2006, e s.m.i. e Delib.G.R. n. 11/75 del 2021.

(altrimenti definito: "Impianto di produzione materiali assorbenti per animali domestici (lettiere) mediante la lavorazione di materiali inerti (argille bentonitiche) essicate utilizzando nell'installazione, quale combustibile, gli oli esausti (messa in riserva R13 e coincenerimento: R1 all'allegato "C" della parte quarta del decreto legislativo 3 aprile 2006, n. 152)" giusto "Avviso al Pubblico" in data 08.10.2024)

Osservazioni.

Con riferimento all'oggetto, obiettivo della presente è quello di Rappresentare all'Autorità Competente per il procedimento di Valutazione Impatto Ambientale alcune evidenze desumibili dal quadro documentale che sostanzia il progetto della Società "Laviosa Chimica Mineraria S.p.A.." *(nel seguito indicata come "Proponente")* unitamente ad alcuni ulteriori elementi conoscitivi e valutativi su base essenzialmente tecnica così come previsto dall' art. 24, comma 3 del del D. Lgs.152/2006, innovato e sostituito dall'art. 21, comma 1, lettera b), della legge n. 108 del 2021.

Trattasi in estrema sintesi di osservazioni in relazione alla documentazione progettuale depositata per la partecipazione del Pubblico e

sulle omissioni in essa chiaramente riscontrabili in relazione all'impatto del progetto proposto non solo sull'Ambiente Naturale ma anche e soprattutto in relazione alla Pubblica Sanità ed Incolumità delle Popolazioni residenti nel circondario del sito d'intervento.

Nello specifico:

- *Inattendibilità in relazione alle effettive caratteristiche delle materie prime elaborate.*
- *Omessa produzione di specifica valutazione in relazione agli impatti sanitari direttamente connessi all'implementazione del progetto, procedura FONDAMENTALE ed INDEROGABILE ogniqualevolta emerga la concreta ipotesi di rischi per la salute delle Popolazioni Residenti nelle aree interessate, in osservanza del Principio di Precauzione in materia ambientale, inserito a pieno titolo nell'Ordinamento Vigente (Art. 191 T.F.U.E. e Art. 3 ter del decreto legislativo n. 152/2006 e s.m.i.).*
- *Consequente NULLITÀ' dello Studio di Impatto Ambientale prodotto/ipotesi di improcedibilità dell'istanza di VIA in parola.*

La congruenza della tesi suesposta con il caso in specie è sinteticamente illustrata nel seguito con l'ausilio di Documentazione scientifica afferente la specifica problematica evidenziata ed idonea a supportare la stessa.

1) Argille bentonitiche: un materiale inerte?

Come chiaramente evidenziato già nel citato Avviso al Pubblico (e diffusamente richiamato in tutta la documentazione progettuale), secondo la valutazione del Proponente, le argille bentonitiche utilizzate come materia prima nell'Impianto di Villaspeciosa sarebbero un materiale inerte.

Appare alquanto d'uopo richiamare la definizione che della bentonite viene data a norma del Regolamento CLP dalla ECHA – European Chemicals Agency - (Agenzia europea per le sostanze chimiche).

Tale Ente collabora con la Commissione europea e i governi dell'UE per individuare le sostanze che suscitano "preoccupazioni" e prendere decisioni in materia di gestione dei rischi a livello Comunitario.

Esso disciplina le sostanze chimiche e i biocidi sul mercato dell'UE, tratta i fascicoli in materia di sostanze chimiche per l'industria e ne verifica la conformità alla Normativa Comunitaria.

Oltre ciò, circostanza di particolare rilevanza nel caso in trattazione, fornisce informazioni sulle sostanze chimiche e il loro uso sicuro attraverso un'unica banca-dati gratuita.

Dalla consultazione di tale banca-dati è stata rilevata un'univoca scheda (INFOCARD) relativa ai materiali denominati "bentonite" (che si riporta come **Allegato-1** in coda alla presente nota) nella quale si esplicita per tale prodotto l'obbligo di etichettatura con pittogramma GHS-07 – "Health Hazard" ("= pericolo per la salute") e l'apposizione della "significativa" avvertenza :

<i>"Warning! According to the classification provided by companies to ECHA in CLP notifications, this substance causes serious eye irritation, causes skin irritation and may cause respiratory irritation"</i>	<i>"Attenzione! Secondo la classificazione fornita dalle Aziende all'ECHA nelle notifiche CLP, questa sostanza provoca grave irritazione oculare, provoca irritazione della pelle e può causare irritazione delle vie respiratorie"</i>
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La bentonite (nel caso corrente essenzialmente l'urasite) non può pertanto essere assolutamente assimilata ad un materiale inerte ma deve essere considerata a tutti gli effetti un minerale industriale di caratteristiche alquanto insidiose, essenzialmente per la granulometria (d_{80} tipico - che può essere fatto coincidere con la cd. dimensione di liberazione - pari o inferiore a 0,002 mm, ovvero 2 micrometri).

Questo equivale a dover ammettere che la bentonite (e quindi l'urasite) ha un contenuto percentuale iniziale in polveri sottili ed in ultrafini (o polveri PM-2,5 denominate polveri respirabili in quanto sono in grado di penetrare anche nel tratto inferiore dell'apparato respiratorio, dalla trachea sino agli alveoli

polmonari) tendenzialmente molto elevato - valore che tende naturalmente e rapidamente a crescere in seguito, ad esempio, di trattamenti di manipolazione, in particolar modo meccanica come nel caso dei processi descritti dallo stesso Proponente che caratterizzano le attività che si realizzano nello stabilimento di Villaspeciosa.

Inequivocabilmente l'intero processo produttivo è caratterizzato da fenomeni di "perdita di massa materiale" che con evidenza non avviene unicamente dai punti di emissione indicati con la siglatura E1, E2, E3 E4 ed E8 come desumibile dalla Relazione Specialistica - Modelli Matematici di Dispersione in Atmosfera - (Documento: "Relazione Modello Climatologico_REV0") ma anche certamente dal Parco materie prime, (costituito da cumuli di bentonite vergine stoccata alla rinfusa) e dai piazzali dello stesso stabilimento di Villaspeciosa che, come ci si rende edotti dalla lettura della pagina 22 dello Studio di Impatto Ambientale (documento: "Studio di Impatto Ambientale_LCM.pdf") costituiscono:

...“L'alternativa all'utilizzo del olio esausto, quale combustibile, oltre all'olio di raffineria, è sostanziata dalla possibilità di effettuare l'essiccazione solare della bentonite. Tale procedura produttiva viene, allo stato attuale, già adottata, nei mesi estivi nei piazzali dello stabilimento di Villaspeciosa nel periodo estivo. Tuttavia, non può, totalmente, essere adottata per i restanti periodi dell'anno...”

Sia la movimentazione fra Parco materie prime e piazzali, sia quella fra gli stessi e la tramoggia di alimentazione all'essiccatore (vera "bocca d'impianto") avvengono ovviamente ed evidentemente con uso di mezzi meccanici.

Altrettanto ovvia appare pertanto la possibile generazione di "polveri" (per quanto "stagionalizzata") le quali con evidenza possono essere facilmente catturate dai flussi eolici e disperse in modo alquanto (se non del tutto) incontrollato (di dubbia o scarsa efficacia appaiono i riferimenti a

nebulizzazioni e/o spruzzature con acque piu' o meno addizionata di latte di calce, a fronte di eventuali nuvole di corpuscoli in balia dei flussi eolici).

Polveri la cui distribuzione granulometrica -c.d. "fuso granulometrico" - esattamente come quelle disperse dai punti di emissione indicati con la siglatura E1, E2, E3 E4 ed E8 e appunto genericamente e laconicamente indicate come "polveri" nello stesso Documento: "Relazione Modello Climatologico_REV0" non può assolutamente essere data per scontata.

Distribuzione granulometrica che altresì condiziona qualsivoglia ipotesi di congruità ed adeguatezza dello studio di Dispersione in Atmosfera e delle relative Tavole Grafiche, peraltro **gravato e con tutta probabilità inficiato dall'utilizzo, nella redazione, dei dati anemometrici e climatologici della Stazione di Decimomannu relativi ad 1 sola annualità (2013) e dello stesso utilizzo di un software (WindImula v. 3.x) che risulta alquanto datato (evidentemente antecedente a Settembre 2013, data di rilascio della versione v. 4.0) e presumibilmente obsoleto.**

Distribuzione granulometrica in relazione alla quali il "Proponente", anche in presenza di una "cospicua", "corposa" ma soprattutto "ponderosa" documentazione progettuale, parrebbe forse aver ritenuto (alquanto "inappropriatamente" come verrà illustrato al punto successivo) di non dover impegnare tempo e risorse quantunque si potrebbero fare considerazioni, anche solo a titolo di semplice esempio, in relazione alle caratteristiche di (lunga) persistenza in aria delle particelle ultrafini ed eventualmente considerare gli effetti cumulativi (la bentonite contiene generalmente minerali associati e silice libera cristallina nelle sue forme polimorfe – sostanza quest'ultima classificata come irritante, fibrogeno e/o sclerogeno e come sospetto cancerogeno per l'uomo (cat.A2 dall'A.C.G.I.H. (American Conference of Governmental Industrial Hygienists) e come cancerogena per l'uomo (gruppo 1) dalla I.A.R.C. (International Agency for Research on Cancer), con valore limite di esposizione indicato in 0,1 mg/mc per la frazione respirabile, ovvero PM_{2,5}).

Considerazioni che in questa Sede ma rischierebbero di apparire quasi ridondanti.

Un'ultimo ulteriore cenno si ritiene meriti l'evidenza che sono proprio le frazioni più fini ad avere la capacità di rimanere più a lungo in sospensione aerea e pertanto di coprire, per trasporto eolico, le maggiori distanze rispetto al punto d'emissione ma questo... collide nuovamente con la più volte citata Relazione Specialistica - Modelli Matematici di Dispersione in Atmosfera inclusa dal Proponente fra i documenti progettuali.

2) Polveri diffuse: distribuzione granulometrica e problematiche connesse.

Diversamente da quanto appena evidenziato in relazione all'operato dal Proponente, la rilevanza della distribuzione granulometrica del particolato (polveri) che verrà ineluttabilmente diffuso dalle attività in progetto e/o già in atto nello stabilimento di Villaspeciosa (trattandosi di una procedura di V.I.A. ex-post), emerge con cristallina chiarezza da tutta una serie di Documenti Scientifici, Studi ed Articoli portati all'attenzione della Comunità Scientifica Internazionale e del Pubblico da svariati Team Accademici di incontestabile Competenza Settoriale.

In questa sede si ritiene del tutto opportuno quanto sufficiente richiamare l'attenzione sul contenuto di un Lavoro pubblicato sulla prestigiosa Rivista Internazionale ***“Lancet Planet Health”*** – vol 7 – ***March 2023***, redatto da un Team coordinato dal Prof. Yu Wenhua del **“Climate, Air Quality Research Unit, School of Public Health and Preventive Medicine”** - **Monash University, Melbourne**, titolato: ***“Global estimates of daily ambient fine particulate matter concentrations and unequal spatio-temporal distribution of population exposure: a machine learning modelling study”***. Di tale Lavoro, che si allega alla Presente per farne parte integrante e sostanziale con la denominazione di **Allegato-3**, si ritiene assolutamente significativo già il semplice passo introduttivo:

Fine particulate matter (PM_{2.5}) is a leading risk factor for premature mortality and morbidity worldwide. According to a Global Burden of Disease Study, outdoor air pollution (including ambient particulate matter) was estimated to cause 6·67 million premature deaths in 2019.¹ In addition, no safe threshold for PM_{2.5} has been identified below which no damage to health is observed.^{2,3} An abundance of evidence has supported the adverse effects of short-term and long-term ambient PM_{2.5} exposure on human health, even at low PM_{2.5} concentrations.⁴

Ovvero:

“Il particolato fine (PM-2,5) è uno dei principali fattori di rischio-mortalità prematura e e morbilità in tutto il mondo. Secondo uno studio globale sulle cause di malattia l'inquinamento dell'aria esterna (compreso il particolato ambientale) esso è stato stimato come causa dai 6 ai 67 milioni di morti premature nel 2019. Inoltre, non è stata identificata una soglia di sicurezza per il PM-2,5 al di sotto della quale non si osservano danni alla salute. Numerose evidenze supportano l'evidenza di effetti negativi sulla salute umana dell'esposizione sia a breve sia a lungo termine al PM-2,5 anche a basse concentrazioni...”

Non si ritiene ci sia molto altro da aggiungere se non rimarcare che... a fronte della **certa produzione e diffusione** di particolato secondo le modalità illustrate al punto precedente, **la documentazione progettuale prodotta dal Proponente NON prende assolutamente in considerazione una problematica potenzialmente esiziale per il Circondario del sito dello stabilimento di Villaspeciosa e per la sua Popolazione** ed impedisce pertanto di poter avanzare anche soltanto di una pura e semplice interpunzione grafica nella conoscenza e definizione di una problematica di Sanità Pubblica che appare assolutamente quanto drammaticamente concreta e tangibile e pertanto da considerare come **autentica, inderogabile e vincolante base per qualsivoglia Valutazione che pur si voglia voglia aggettivare come “di Compatibilità Ambientale”.**

Conclusioni

Sebbene – in linea di principio – nell’ambito di un procedimento per il rilascio di VIA non risulti al momento obbligatorio procedere a specifica Valutazione di Impatto Sanitario, appare anche d’uopo ricordare che la sentenza del Consiglio di Stato, Sez. IV, 11 febbraio 2019, n. 983 ha stabilito che:

“...La valutazione di impatto sanitario (V.I.S.), normalmente necessaria per le centrali termiche, i grandi impianti di combustione (potenza termica superiore a 300 MW), gli impianti di raffinazione, gassificazione, liquefazione (punti 1 e 2 dell’Allegato II del decreto legislativo n. 152/2006 e s.m.i.), è comunque, necessaria ogni volta che emerga in sede istruttoria la concreta ipotesi di un rischio per la salute delle popolazioni interessate. Si tratta di espressione del principio di precauzione in materia ambientale, ormai inserito a pieno titolo nell’Ordinamento (art. 191 TFUE, art. 3 ter del decreto legislativo n. 152/2006 e s.m.i.). In tali situazioni, se l’Amministrazione Pubblica non vi desse seguito, incorrerebbe nel vizio dell’eccesso di potere sotto il profilo del mancato necessario approfondimento istruttorio.”

Appare pertanto come assolutamente Razionale, Ragionevole, e Giuridicamente Fondata l’ipotesi dell’inderogabilità di procedervi ogniqualvolta emerga in sede istruttoria la concreta ipotesi di un rischio per la salute delle popolazioni interessate. Ipotesi che non risulta poter essere “superata” e/o “bypassata” nemmeno in presenza di progetti alquanto opportunamente referenziati come “funzionali all’economia dell’intero Territorio Regionale” anche se forse lo risultano assai di più per gli Interessi (economici) del Proponente.

Si ritiene in ogni caso di aver fornito, con la presente breve Memoria, adeguati elementi di valutazione in relazione alla cogenza/inderogabilità della redazione/produzione nell’ambito del procedimento avviato di specifico studio di valutazione in relazione agli impatti Sanitari sulla Popolazione Residente nel Circondario territoriale su cui l’installazione di Villaspeciosa andrà a gravare, incluso soprattutto l’abitato del Comune di UTA, in relazione al quale i quartieri

residenziali di “Santa Porada”, “Bascus Argius” e “Pintulinu” risultano immediatamente esposti a poche centinaia di metri - come chiaramente desumibile dalla tavola grafica acclusa in coda alla presente come **Allegato-2** - sottovento rispetto ai flussi eolici dominanti da NW e da N-NW (Maestrale) - alle emissioni dello stabilimento di Villaspeciosa, unitamente alla zona P.I.P., agli impianti sportivi ed allo stesso sito in cui è attualmente in realizzazione il nuovo Plesso Scolastico Unificato, nell’Ambito del Progetto “ISCOLAS” finanziato dalla Regione Autonoma della Sardegna.

In assenza di tale fondamentale indagine non si vede alternativa all’AUSPICARE che le osservazioni formulate in questo documento siano considerate nell’ambito del presente procedimento di Valutazione d’Impatto Ambientale da parte delle Autorità Competenti, ai sensi degli artt. 9 e ss. della legge n. 241/1990 e s.m.i. e artt. 24 del decreto legislativo n. 152/2006 e s.m.i., e CHIEDERE che le stesse vogliano esprimersi con una FERMA, MOTIVATA E DEFINITIVA VALUTAZIONE DI NON COMPATIBILITÀ AMBIENTALE, ai sensi degli artt. 25 e ss. del decreto legislativo n. 152/2006 e s.m.i.

Uta li 08.11.2024

in fede



Allegato - 1
ECHA – European Chemicals Agency
Scheda (INFOCARD) relativa all’etichettatura obbligatoria
della sostanza denominata “bentonite”

Bentonite

A colloidal clay. Consists primarily of montmorillonite.

[Regulatory process names](#) 4 [Translated names](#) 23 [IUPAC names](#) 6 [Other names](#) 1 [Other identifiers](#) 13

Substance identity

EC / List no.: 215-108-5

CAS no.: 1302-78-9

Mol. formula:

⌈ ⌋

...

⌈ ⌋



Hazard classification & labelling



Warning! According to the classification provided by companies to ECHA in **CLP notifications** this substance causes serious eye irritation, causes skin irritation and may cause respiratory irritation.

Biocidal Uses

This substance is approved in the EEA and/or Switzerland for use in biocidal products more favourable for the environment, human or animal health.

Allegato - 2

*Inter - distanze reali Punti emissione macroinquinanti atmosferici
Impianto Laviosa – abitato di UTA*



Allegato – 3

Impianto di coincenerimento rifiuti della società Laviosa Chimica Mineraria S.p.A. stabilimento di Villaspeciosa, Località Perda Bianca". Procedimento di valutazione di impatto ambientale (V.I.A.) "ex post". D.Lgs. 152/2006, e s.m.i. e Delib.G.R. n. 11/75 del 2021. - Osservazioni



Global estimates of daily ambient fine particulate matter concentrations and unequal spatiotemporal distribution of population exposure: a machine learning modelling study

Wenhua Yu, Tingting Ye, Yiwen Zhang, Rongbin Xu, Yadong Lei, Zhuqing Chen, Zhengyu Yang, Yuxi Zhang, Jiangning Song, Xu Yue, Shanshan Li, Yuming Guo

Summary

Background Short-term exposure to ambient PM_{2.5} is a leading contributor to the global burden of diseases and mortality. However, few studies have provided the global spatiotemporal variations of daily PM_{2.5} concentrations over recent decades.

Methods In this modelling study, we implemented deep ensemble machine learning (DEML) to estimate global daily ambient PM_{2.5} concentrations at 0.1°×0.1° spatial resolution between Jan 1, 2000, and Dec 31, 2019. In the DEML framework, ground-based PM_{2.5} measurements from 5446 monitoring stations in 65 countries worldwide were combined with GEOS-Chem chemical transport model simulations of PM_{2.5} concentration, meteorological data, and geographical features. At the global and regional levels, we investigated annual population-weighted PM_{2.5} concentrations and annual population-weighted exposed days to PM_{2.5} concentrations higher than 15 µg/m³ (2021 WHO daily limit) to assess spatiotemporal exposure in 2000, 2010, and 2019. Land area and population exposures to PM_{2.5} above 5 µg/m³ (2021 WHO annual limit) were also assessed for the year 2019. PM_{2.5} concentrations for each calendar month were averaged across the 20-year period to investigate global seasonal patterns.

Findings Our DEML model showed good performance in capturing the global variability in ground-measured daily PM_{2.5}, with a cross-validation R² of 0.91 and root mean square error of 7.86 µg/m³. Globally, across 175 countries, the mean annual population-weighted PM_{2.5} concentration for the period 2000–19 was estimated at 32.8 µg/m³ (SD 0.6). During the two decades, population-weighted PM_{2.5} concentration and annual population-weighted exposed days (PM_{2.5} >15 µg/m³) decreased in Europe and northern America, whereas exposures increased in southern Asia, Australia and New Zealand, and Latin America and the Caribbean. In 2019, only 0.18% of the global land area and 0.001% of the global population had an annual exposure to PM_{2.5} at concentrations lower than 5 µg/m³, with more than 70% of days having daily PM_{2.5} concentrations higher than 15 µg/m³. Distinct seasonal patterns were indicated in many regions of the world.

Interpretation The high-resolution estimates of daily PM_{2.5} provide the first global view of the unequal spatiotemporal distribution of PM_{2.5} exposure for a recent 20-year period, which is of value for assessing short-term and long-term health effects of PM_{2.5}, especially for areas where monitoring station data are not available.

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Introduction

Fine particulate matter (PM_{2.5}) is a leading risk factor for premature mortality and morbidity worldwide. According to a Global Burden of Disease Study, outdoor air pollution (including ambient particulate matter) was estimated to cause 6.67 million premature deaths in 2019.¹ In addition, no safe threshold for PM_{2.5} has been identified below which no damage to health is observed.^{2,3} An abundance of evidence has supported the adverse effects of short-term and long-term ambient PM_{2.5} exposure on human health, even at low PM_{2.5} concentrations.⁴ Therefore, the latest version of the WHO global air quality guidelines published in 2021 has adjusted the recommended limit for outdoor PM_{2.5}

exposure from 10 µg/m³ to 5 µg/m³ for the annual mean exposure and from 25 µg/m³ to 15 µg/m³ for 24-h mean exposure.⁵

Estimates of global PM_{2.5} concentrations are a prerequisite for health risk assessments of global air pollution. Despite global expansion of the ground-based PM_{2.5} monitoring network (an example of which is the WHO ambient air pollution database), the frequently sparse spread and poor homogeneity in the distribution of monitoring stations make accurate measurement of global PM_{2.5} exposure challenging. Fortunately, with improvement of statistical and machine learning methods, the combination of emerging satellite products, chemical transport model simulations, and ground monitor

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For the WHO global ambient air pollution database see <https://www.who.int/data/gho/data/themes/topics/topic-details/GHO/ambient-air-pollution>

Research in context

Evidence before this study

Assessing the disparities in global exposure to $PM_{2.5}$ is crucial for population health risk assessment. However, little is known about the global short-term (daily) $PM_{2.5}$ exposure and its spatiotemporal variations. We searched PubMed and Google Scholar for articles in English published between Jan 1, 2011, and Dec 31, 2021, with the terms “fine particulate matter”, “ $PM_{2.5}$ ”, “global estimate”, and “short-term daily exposure”. We found that most previous studies estimated daily average $PM_{2.5}$ distribution at the city or national levels. Several studies have estimated global long-term trends in $PM_{2.5}$ concentrations, but few articles have reported daily $PM_{2.5}$ distribution worldwide. One article investigated the global daily $PM_{2.5}$ distribution from 1997 to 2014; however, it did not explore the inequality and spatiotemporal changes in daily $PM_{2.5}$ population exposure over decades.

Added value of this study

Our study provides a global perspective on the spatial and temporal distribution of daily $PM_{2.5}$ concentrations in 2000–19 and assesses the inequalities in global population exposure on the basis of the new $PM_{2.5}$ limits in the 2021 WHO air quality guidelines. We used a deep ensemble machine

learning (DEML) approach to estimate the daily mean $PM_{2.5}$ concentrations in each global grid cell ($0.1^\circ \times 0.1^\circ$ spatial resolution) for 20 years with use of ground-based $PM_{2.5}$ data from 5446 monitoring stations in 65 countries worldwide, combined with GEOS-Chem chemical transport model simulations, meteorological data, and geographical information on a global scale. Based on modelled estimates, only 0.001% of the global population was exposed to $PM_{2.5}$ at concentrations lower than the WHO annual limit ($5 \mu\text{g}/\text{m}^3$), and more than 70% of days of the year globally had a $PM_{2.5}$ concentration exceeding the WHO daily limit ($15 \mu\text{g}/\text{m}^3$) in 2019.

Implications of all the available evidence

Our grid-based daily $PM_{2.5}$ estimates could fill knowledge gaps with regard to the assessment of global $PM_{2.5}$ -attributable health burden and both short-term and long-term health effects, especially for areas where monitoring station data are not available. The study results offer a novel global perspective of daily spatiotemporal variations in exposure of the global population to ambient $PM_{2.5}$. Additionally, the DEML approach achieved good performance in estimating global air pollution concentrations.

expansion offers novel opportunities to accurately assess $PM_{2.5}$ concentrations globally.^{6–8} Global $PM_{2.5}$ estimation studies have typically focused on long-term (annual or monthly average) $PM_{2.5}$ estimates,^{7,8} whereas few studies have explored the short-term (from hours to days) exposure to $PM_{2.5}$ concentrations at a global level.⁹ Although increasing numbers of studies have estimated daily $PM_{2.5}$ concentrations at national and regional levels, such as in China,¹⁰ Europe,¹¹ and the USA,¹² few studies have assessed short-term $PM_{2.5}$ exposure and its spatiotemporal variations at the global level. The absence of uniformity in global training data and inconsistency in estimation methods create difficulties in comparing the previous regional estimates and in providing a global view of the spatiotemporal distribution in $PM_{2.5}$ exposure. Therefore, estimating global daily $PM_{2.5}$ concentration and its spatiotemporal variations with unified study designs, modelling approaches, and data sources is warranted.

Machine learning algorithms, especially ensemble machine learning technologies, have proven to be promisingly efficient for geospatial air pollution prediction¹³ and have been increasingly applied in the estimation of $PM_{2.5}$ concentration, offering high accuracy and the ability to handle large numbers of features with nonlinear associations.¹⁰ Ensemble learning algorithms use a collection of multiple machine learning algorithms to achieve an optimal combination of predictions.¹⁴ Multiple studies have indicated that ensemble machine learning could provide better estimations in environmental exposure assessment than a single machine learning model alone.^{15,16}

The deep ensemble machine learning (DEML) framework is a multilevel stacked ensemble machine learning algorithm that combines the advantages of several diverse base models and meta models to achieve an optimal prediction performance.¹⁷ Our previous study indicated the advantages of DEML modelling in environmental exposure assessment.¹⁷ In this study, we used a DEML model to assess the global daily $PM_{2.5}$ distribution at high spatial resolution ($0.1^\circ \times 0.1^\circ$) from 2000 to 2019. We also investigated regional and national population-weighted $PM_{2.5}$ concentrations and population-weighted exposed days at $PM_{2.5}$ concentration higher than the 2021 WHO daily limit ($15 \mu\text{g}/\text{m}^3$) to identify the spatiotemporal exposure distribution from 2000 to 2019.

Methods

Overview and data sources

In this modelling study, we utilised a DEML framework by integrating ground monitoring measurements of $PM_{2.5}$, chemical transport model simulations of all-source $PM_{2.5}$ concentrations, meteorological conditions, and geographical features. All data used in the modelling were for the period between Jan 1, 2000, and Dec 31, 2019.

Daily mean (24-h) concentrations of $PM_{2.5}$ recorded by government air quality monitoring stations in 2000–19 were obtained from multiple sources, including the national environmental protection agencies of the USA and China, the European Environmental Agency, and Australian state and territory government agencies.¹⁸ Details on the air quality station data sources are provided

For the US Environmental Protection Agency air data see https://aqs.epa.gov/aqswb/airdata/download_files.html

For the China National Environmental Monitoring Centre see <http://www.cnemc.cn/en/>

For the European Environmental Agency air quality database see <https://www.eea.europa.eu/data-and-maps/data/aqereporting-9>

For the Australian National Air Pollution Monitor Database see <https://osf.io/jxd98/>

in the appendix (p 4). Figure 1 shows the spatial distribution of the included 5446 monitoring stations in 65 countries. These countries covered 73% of the global population in 2019 according to WorldPop data, and 56% of the global land area in 2019 according to land cover data from the MCD12Q1.061 product. A summary of the sampled monitoring stations by country is presented in the appendix (pp 6–7).

We extracted simulations of global daily all-source $PM_{2.5}$ concentrations for the study period by using a three-dimensional (3D) GEOS-Chem chemical transport model (version 12.0.0) at a spatial resolution of $2.0^\circ \times 2.5^\circ$ as described by Yue and colleagues.¹⁹ Further details on the data sources for the GEOS-Chem model are provided in the appendix (p 1).

Satellite-based meteorological data with a spatial resolution of $0.1^\circ \times 0.1^\circ$ were collected from the ERA5 dataset (the fifth-generation European Centre for Medium-Range Weather Forecasts reanalysis set)²⁰ through the Google Earth Engine platform. For the study period, we collected ambient temperature, ambient dew point temperature (at 2 m above the land surface), wind speed at 10 m height above sea level, surface pressure, surface solar radiation, total precipitation, and total evaporation. All were collected as hourly measurements for the period under study. Daily mean data were calculated as an average of 24 h of observations, starting each day from 12 noon. Daily relative humidity was calculated with an algorithm in the humidity package²¹ in R (version 0.1.5) on the basis of daily mean ambient temperature and dew point temperature.

Satellite-based annual land cover data with a spatial resolution of 500 m were obtained from the MCD12Q1.061 product through the Google Earth Engine platform. Satellite-based annual digital elevation data with a spatial resolution of 90 m were extracted from the Shuttle Radar Topography Mission project. Global annual population counts at 1 km spatial resolution were obtained from the WorldPop platform.

All data variables and sources are summarised in the appendix (pp 4–5). Bilinear interpolation was used to upscale the GEOS-Chem simulations, land cover, ground elevation, and population density to $0.1^\circ \times 0.1^\circ$ spatial resolution for further grid cell estimation.²² Missing values in the collected datasets were excluded in the analysis to avoid the potential bias introduced by imputation. For ground measurements of $PM_{2.5}$, we removed extreme outliers higher than the 99.9th quantiles of the total dataset, and removed unsuitable data obtained from six stations in central eastern Asia (Kazakhstan, $n=1$; Uzbekistan, $n=1$; Tajikistan, $n=2$; and Afghanistan, $n=2$) due to data being constantly or extremely high.

Statistical analysis

We estimated global daily $PM_{2.5}$ concentration using the DEML framework,¹⁷ which has a three-stage structure. At the first stage, three base models (random forest, light

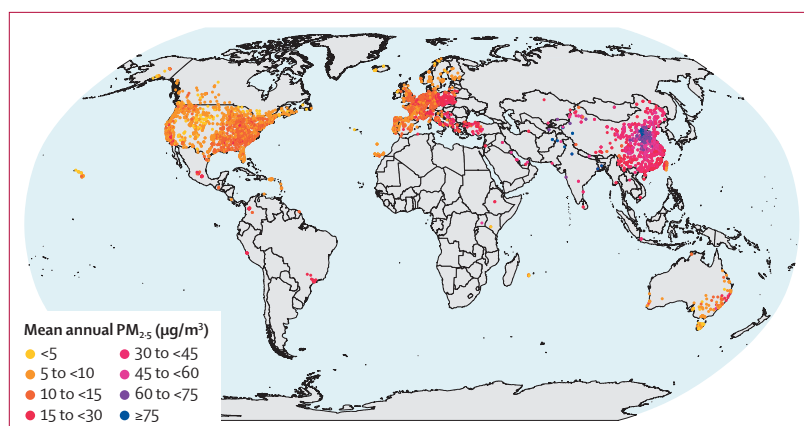


Figure 1: Global monitor station distribution and mean annual $PM_{2.5}$ concentration over two decades (2000–19)

gradient boosting machine [LightGBM], and extreme gradient boosting [XGboost])²³ were used to generate new $PM_{2.5}$ concentration data. At the second stage, two meta-models (random forest and generalised linear model)²³ were used to estimate daily $PM_{2.5}$ concentrations with the predictions from the first-stage models. At the third stage, a non-negative least squares algorithm was used to obtain optimal weights for $PM_{2.5}$ estimations. Details of the DEML framework are presented in the appendix (pp 1–2).

Spatiotemporal generalisation and reliability of the DEML framework were evaluated by a series of cross-validations, including a general 10-fold cross-validation, spatial cross-validations (based on monitor station sampling and grid cell sampling), a continent-stratified cluster cross-validation, and a temporal cross-validation by year. Details on the cross-validation strategies are provided in the appendix (p 3). Root mean square error (RMSE), mean absolute error, and coefficients of determination (R^2) were used to assess the prediction performance of the DEML model. Benchmark machine learning models (random forest, LightGBM, and XGboost) were used to compare the performance of DEML.

For observed and estimated daily values, we present both mean (SD), median (IQR), and range. We calculated the monthly and annual mean values (from daily mean values in each calendar month or year) for observed and predicted $PM_{2.5}$ concentration and used density scatter plots to assess DEML performance for the daily, monthly, and annual mean $PM_{2.5}$ estimations in the study period. Spearman's correlation analysis was used to compare consistency between the DEML-estimated and observed daily mean $PM_{2.5}$ concentrations at the global and regional levels. We also compared the variability in temporal trends of observed and predicted daily mean $PM_{2.5}$ in eight metropolises (Beijing, China; Delhi, India; Ho Chi Minh, Viet Nam; Milan, Italy; New York, NY, USA; Sao Paulo, Brazil; Sydney, NSW, Australia; and Toronto, ON, Canada), which were selected as being regionally representative and for comparison with

See [Online](#) for appendix

For **WorldPop** see <https://hub.worldpop.org/>

For the **MCD12Q1.061 product** see https://developers.google.com/earth-engine/datasets/catalog/MODIS_061_MCD12Q1#description

For the **GEOS-Chem chemical transport model** see <https://geos-chem.seas.harvard.edu/>

For the **ERA5 dataset** see https://developers.google.com/earth-engine/datasets/catalog/ECMWF_ERA5_LAND_HOURLY

For the **Shuttle Radar Topography Mission project** see <https://bigdata.cgair.org/srtm-90m-digital-elevation-database/>

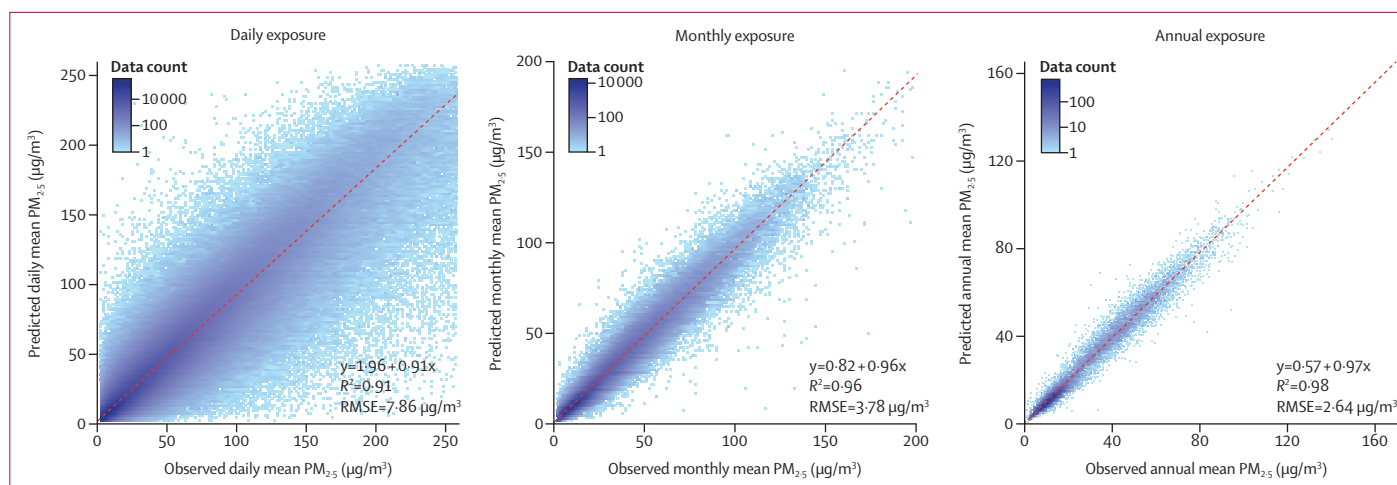


Figure 2: Comparison between observed and model-estimated PM_{2.5} concentrations in 2000–19

The x-axis indicates observed mean PM_{2.5} in the monitor stations; the y-axis indicates estimated mean PM_{2.5} by the deep ensemble machine learning model in 10-fold cross-validation analysis; the points represent the corresponding PM_{2.5} for both observed and predicted values. There are 9 289 613 datapoints for daily PM_{2.5}, 434 122 for monthly PM_{2.5}, and 38 488 for annual PM_{2.5}; the red line represents a regression line (simple linear regression) for the observed and predicted PM_{2.5}, R²=coefficient of determination for the unseen independent data in 10-fold cross-validation analysis. RMSE=root mean square error.

previous studies. Overall averages for the 20-year study period were also calculated.

We assessed the global spatial distribution of PM_{2.5} and the spatiotemporal changes per decade. We presented the global distribution of annual mean PM_{2.5} in 2000, 2010, and 2019, and measured the changes in annual mean PM_{2.5} per decade by multiplying 10 years with the coefficients of a linear regression model established from 20 annual mean PM_{2.5} values for each global grid cell (0.1°×0.1° spatial resolution). Additionally, the proportion of global and regional land area exposure and population exposure at the previous 2005 WHO limit (10 µg/m³) and updated 2021 WHO limit (5 µg/m³) for annual mean PM_{2.5} were assessed for the year 2019.

Population-weighted PM_{2.5} and population-weighted exposed days, when daily PM_{2.5} concentration was higher than the 2021 WHO-recommended limit (15 µg/m³), were identified to indicate both the levels of air pollution and the size of the affected population in each region. Regional mean population-weighted PM_{2.5} concentrations were identified as:

$$\text{Population-weighted concentration} = \sum \frac{p_i^w}{P} \times C_i$$

Where C_i denotes the daily mean PM_{2.5} concentration and p_i the annual mean population in a specific grid cell, i ; and, $P = \sum p_i$, which is the total population of grid cells in a specified region. Regional population-weighted exposed days (PM_{2.5} > 15 µg/m³) were identified as:

$$\text{Population-weighted exposed days} = \sum \sum \frac{p_i^w}{P} \times D_{ij}$$

Where D_{ij} is a Boolean value, indicating whether daily mean PM_{2.5} in a specific j day of a year in a grid cell i was higher than 15 µg/m³ ($D_{ij}=1$) or not ($D_{ij}=0$). This indicator

can be used to quantify the average exposed days to concentrations higher than the WHO guideline limit for the population in a specified region. Study regions were based on the country and regional groupings of the UN Statistics Division, and we provide estimates for 175 countries. The annual mean population-weighted PM_{2.5} concentrations and annual accumulation of population-weighted exposed days at the global and regional levels were used to assess global and regional PM_{2.5} exposure in 2000, 2010, and 2019. Countries with the highest exposures were ranked and labelled by income group (2019 World Bank classification). To investigate seasonal patterns, PM_{2.5} concentrations for each calendar month were averaged across the 20-year period for each global grid cell. We also assessed mean population-weighted PM_{2.5} values for each calendar month over the 20 years at the global population level and in eight specific countries (Australia, Brazil, China, India, South Africa, Thailand, the UK, and the USA), which were selected as regionally representative countries and for comparison with previous studies.

Role of the funding source

The funders of the study had no role in study design, data collection, data analysis, data interpretation, or writing of the report.

Results

Globally, across 175 countries, the mean annual population-weighted PM_{2.5} concentration for the period 2000–19 was estimated at 32.8 µg/m³ (SD 0.6). When estimating mean annual exposure across regions, the highest population-weighted PM_{2.5} concentrations were distributed in eastern Asia (50.0 µg/m³ [SD 2.2]) and southern Asia (37.2 µg/m³ [1.2]), followed by northern

For the country and regional classifications of the UN Statistics Division see <https://unstats.un.org/unsd/methodology/m49/>

For The World Bank income classification see <https://datahelpdesk.worldbank.org/knowledgebase/articles/906519-world-bank-country-and-lending-groups>

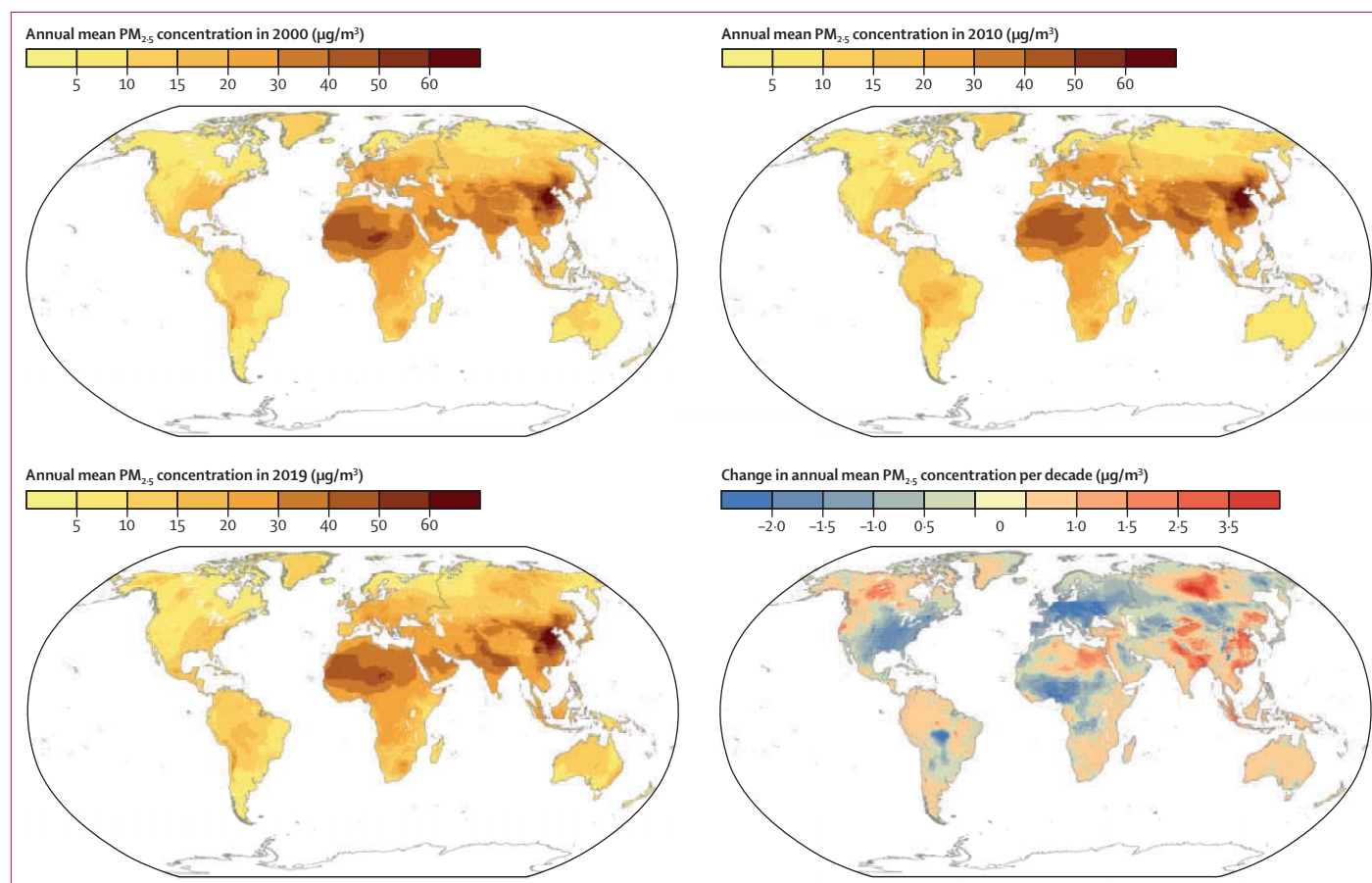


Figure 3: Annual mean PM_{2.5} in 2000, 2010, and 2019 and changes in annual mean PM_{2.5} per decade at 0.1° × 0.1° spatial resolution

The change in annual PM_{2.5} per decade was calculated by multiplying 10 years with the coefficients of a linear regression model established from 20 annual mean PM_{2.5} values for each global grid cell.

Africa (30.1 µg/m³ [0.7]). Australia and New Zealand (8.5 µg/m³ [0.9]), other regions in Oceania (12.6 µg/m³ [0.5]), and southern America (15.6 µg/m³ [0.3]) had the lowest annual population-weighted PM_{2.5} concentrations (appendix p 8). We observed the highest correlation between estimated and observed daily mean PM_{2.5} in eastern Asia (Spearman's $r=0.87$) and western Europe ($r=0.82$), and the lowest correlation in Australia and New Zealand ($r=0.59$). Estimated global daily mean PM_{2.5} concentrations had high correlation with the observed global mean ($r=0.91$; appendix p 9). The temporal trends between observed and predicted daily PM_{2.5} in eight metropolises are presented in the appendix (p 21).

Observed and DEML-predicted PM_{2.5} concentrations were compared (figure 2). DEML could accurately capture the global variability in ground-measured daily mean PM_{2.5} with a 10-fold cross-validation R^2 of 0.91 and RMSE of 7.86 µg/m³, and with a cross-validation R^2 of 0.96 (RMSE 3.78 µg/m³) for monthly mean PM_{2.5} and 0.98 (2.64 µg/m³) for annual mean PM_{2.5}. Results of other spatiotemporal cross-validations also indicated robust model performances (appendix pp 10–13).

We presented the spatial distribution of estimated annual mean PM_{2.5} in 2000, 2010, and 2019 and spatiotemporal changes per decade in 2000–19 (figure 3). Despite large differences between regions in PM_{2.5} concentrations, most areas with high PM_{2.5} were in eastern Asia, southern Asia, and northern Africa. Most areas in Asia, northern and sub-Saharan Africa, Oceania, and Latin America and the Caribbean had increases in PM_{2.5} concentrations during the 20 years, while decreases were estimated in Europe, some regions of northern America, and some regions of Africa (figure 3). Based on the 2005 WHO guideline limit,²⁴ 29.4% of the global land area and 1.8% of the global population were exposed to an annual mean concentration of PM_{2.5} lower than 10 µg/m³ in 2019. When restricting to the 2021 WHO guideline value of 5 µg/m³, only 0.18% of the global land area and 0.001% of the global population remained at an annual exposure lower than the guideline limit in 2019. Regional land area exposures and population exposures in 2019 are provided in the appendix (pp 14–15).

We assessed temporal changes in estimated annual mean population-weighted PM_{2.5} concentration and the proportion of annual population-weighted exposed days

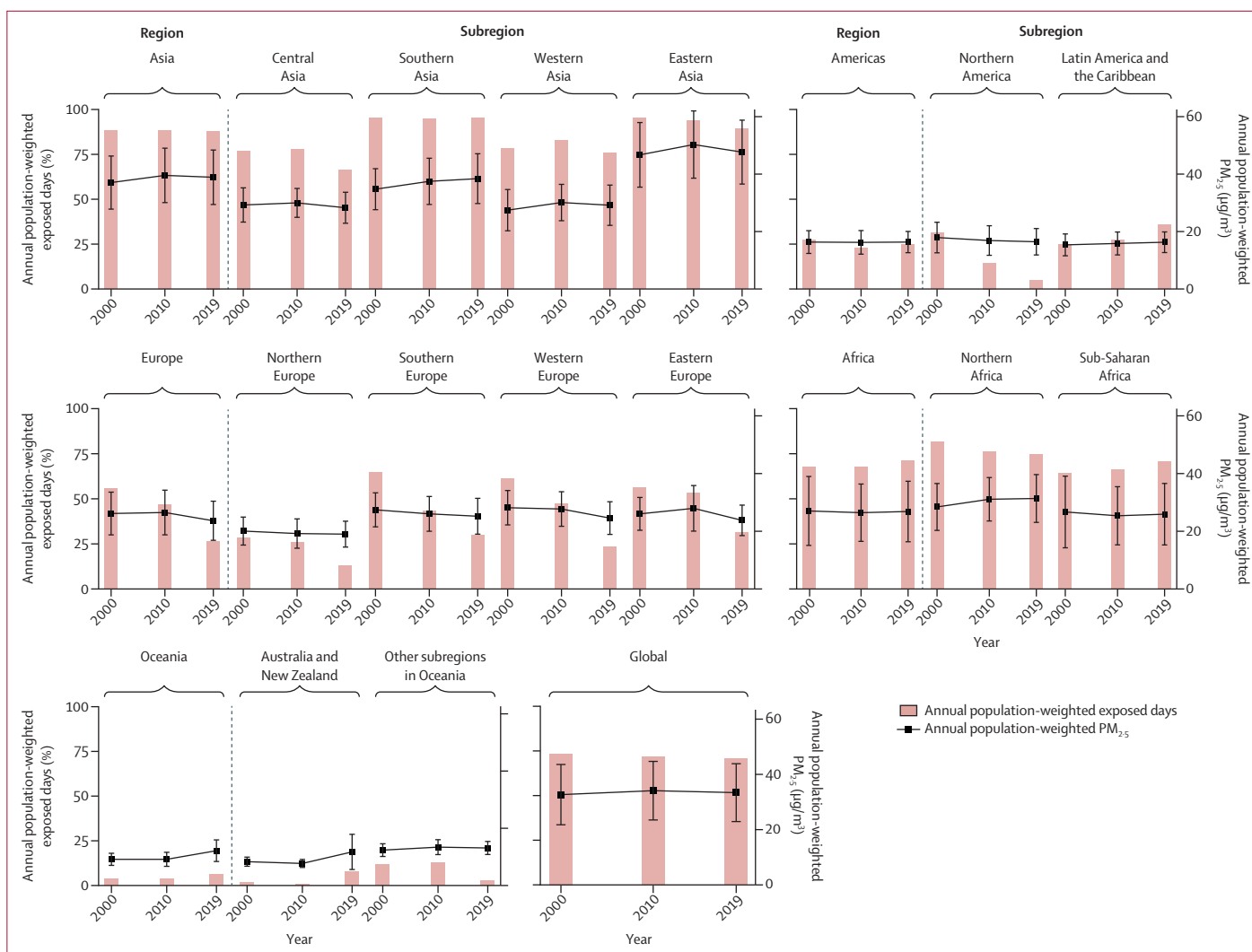


Figure 4: Changes in annual mean population-weighted $PM_{2.5}$ concentration and annual population-weighted exposed days by region in 2000, 2010, and 2019

The bar chart shows the proportion of population-weighted exposed days with $PM_{2.5}$ concentrations higher than the 2021 WHO air quality guideline limit ($15 \mu\text{g}/\text{m}^3$) in a year. Datapoints and connecting lines show the annual mean population-weighted $PM_{2.5}$ concentrations; error bars show standard deviation.

($PM_{2.5} > 15 \mu\text{g}/\text{m}^3$) in 2000, 2010, and 2019 (figure 4). Globally, we observed similar annual mean population-weighted $PM_{2.5}$ at each timepoint, from $31.6 \mu\text{g}/\text{m}^3$ (SD 10.5) in 2000, to $33.0 \mu\text{g}/\text{m}^3$ (10.3) in 2010, to $32.3 \mu\text{g}/\text{m}^3$ (10.1) in 2019 (figure 4). Geographically, population-weighted $PM_{2.5}$ concentration and annual population-weighted exposed days in Europe and northern America decreased over the two decades, whereas exposures increased in southern Asia, Australia and New Zealand, and Latin America and the Caribbean. Despite a slight decrease in population-weighted exposed days globally, by 2019 more than 70% of days still had $PM_{2.5}$ concentrations higher than the 2021 WHO daily limit ($15 \mu\text{g}/\text{m}^3$). In southern Asia and eastern Asia, more than 90% of days in 2000, 2010, and 2019 had daily $PM_{2.5}$ concentrations higher than

$15 \mu\text{g}/\text{m}^3$ (figure 4). Oceania, especially Australia and New Zealand, had a marked increase in the number of days with high $PM_{2.5}$ concentrations in 2019. Trends in annual mean population-weighted concentrations by continent are presented in the appendix (p 22).

We ranked countries with the highest estimates of population-weighted $PM_{2.5}$ exposure and exposed days above $15 \mu\text{g}/\text{m}^3$ $PM_{2.5}$ in 2000, 2010, and 2019 (figure 5). Despite a decrease in estimated $PM_{2.5}$ exposure between 2010 and 2019, China ranked first for annual mean population-weighted $PM_{2.5}$ concentration at all three timepoints. Additionally, increased country rank in the past two decades according to annual mean population-weighted $PM_{2.5}$ concentration was observed in southern Asian countries such as Bangladesh (from 11th in 2000 to 3rd in 2019), India (from 15th to 8th), and

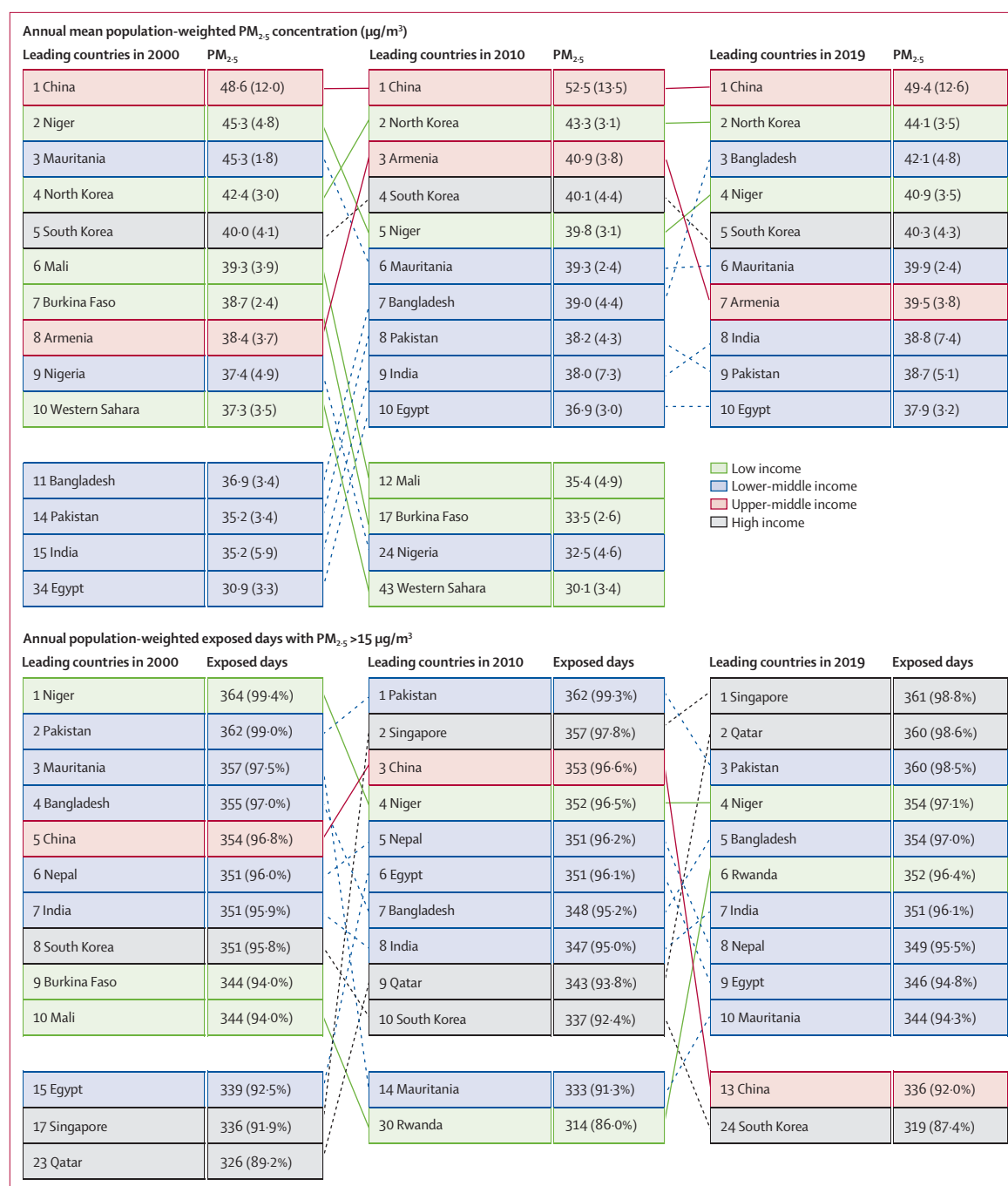


Figure 5: Leading countries for annual mean population-weighted PM_{2.5} concentration and population-weighted exposed days in 2000, 2010, and 2019
 Values in parentheses are standard deviation (for PM_{2.5} concentration) or % (for proportion of days out of 365). The 2021 WHO air quality guideline limit for daily PM_{2.5} (15 µg/m³) was the threshold for population-weighted exposed days. Income categories are based on The World Bank classifications for the year 2019.

Pakistan (from 14th to 9th). For annual population-weighted exposed days, all countries ranked within the top 10 in 2000, 2010, and 2019 had more than 90% of days of the year exceeding 15 µg/m³, including several high-income countries such as Singapore and Qatar in 2010 and 2019, and South Korea in 2000 and 2010.

Estimates of annual mean population-weighted PM_{2.5} and annual population-weighted exposed days in 2000, 2010, and 2019 by all countries and regions are listed in the appendix (pp 16–20).

Seasonal patterns in estimated PM_{2.5} concentrations for the 20-year period were investigated (figure 6). The

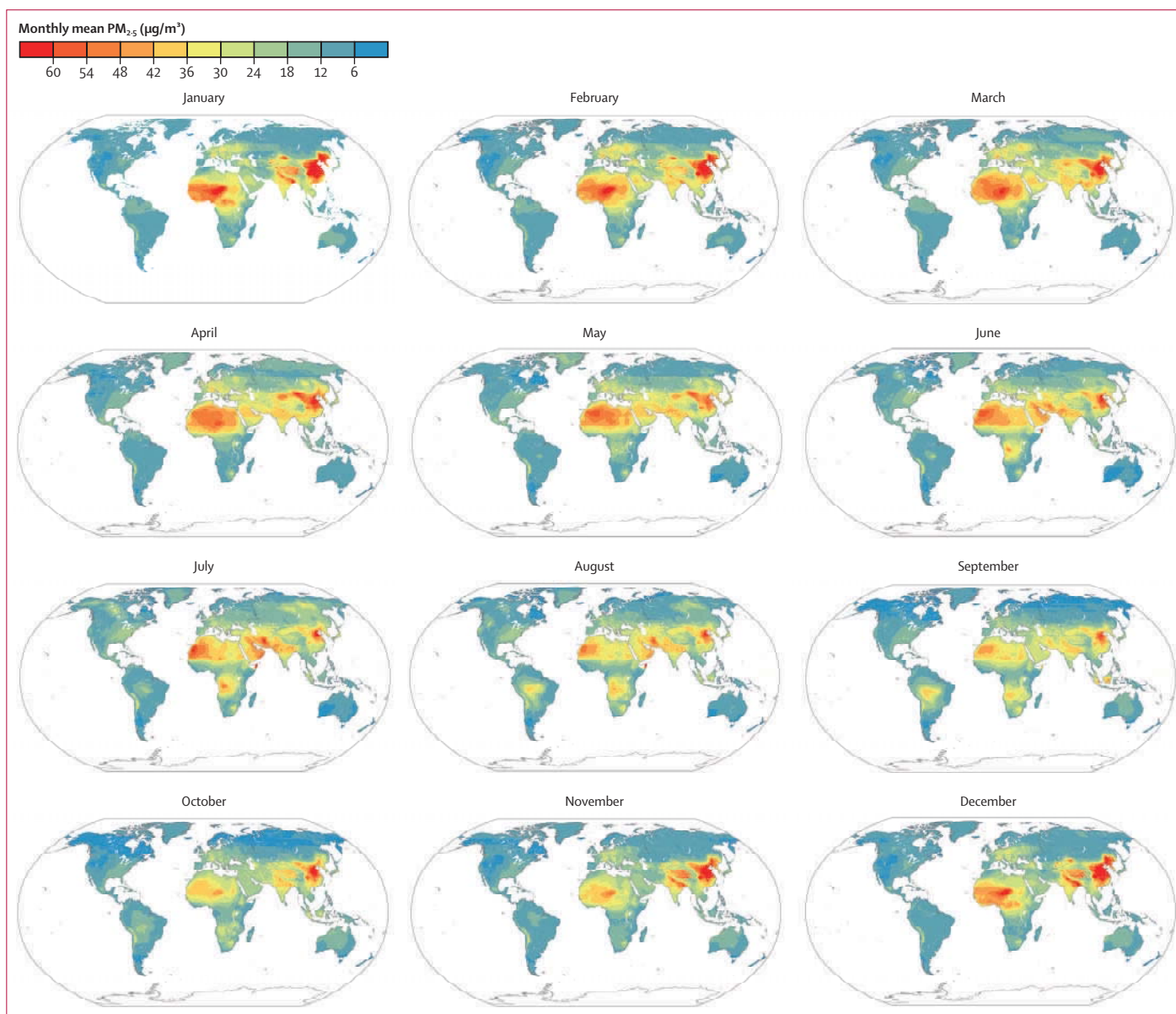


Figure 6: Global mean $PM_{2.5}$ concentrations for each calendar month in 2000–19 at $0.1^\circ \times 0.1^\circ$ spatial resolution
 Estimated daily mean $PM_{2.5}$ values were used to calculate mean $PM_{2.5}$ for each calendar month, and the monthly mean values were averaged over the 20 years.

estimates of $PM_{2.5}$ showed marked seasonal variations across particular regions and countries. For example, northeast China and north India had high $PM_{2.5}$ concentrations during their winter months (December, January, and February), whereas eastern areas in northern America had high $PM_{2.5}$ in its summer months (June, July, and August). Relatively high $PM_{2.5}$ air pollution was observed in August and September in South America and from June to September in sub-Saharan Africa. Seasonal patterns for eight national distributions are presented in the appendix (p 23).

Discussion

Our grid-based daily $PM_{2.5}$ estimates provide the first global perspective on the spatial and temporal variations of population exposure in 2000–19. We implemented a validated DEML model on the basis of $PM_{2.5}$ data from 5446 monitoring stations in 65 countries worldwide, combined with chemical transport model simulations, meteorological conditions, and other geographical information, and achieved strong model performance. Consistent with previous studies,^{6,8,10} our results for eastern and southern Asia indicated the highest $PM_{2.5}$ concentrations. Additionally, the estimates

of PM_{2.5} showed distinct seasonal patterns in many regions of the world.

In this study, the innovative DEML approach to estimate daily PM_{2.5} concentrations attained high prediction accuracy. Global estimation of PM_{2.5} concentrations has been described in many previous modelling studies.^{6,9,25,26} An estimation of global daily PM_{2.5} concentration in 1997–2014 used remote sensing and meteorological data, and ground-based observations of PM_{2.5} in 55 countries, to train a machine learning algorithm, achieving a correlation coefficient for each independent validation dataset of 0.52–0.75.⁹ Similarly, other studies have measured global monthly and annual mean PM_{2.5} exposure. Hammer and colleagues⁶ assessed global estimates of annual mean PM_{2.5} concentrations in 1998–2018 with data from satellite observations, a chemical transport model, and ground-based monitoring, obtaining an R^2 of 0.90–0.92. Shaddick and colleagues⁸ used a Bayesian hierarchical model with multiple data integration to estimate the global annual mean PM_{2.5} exposures in 2014 with an R^2 of 0.91. However, comparisons with previous global PM_{2.5} estimation studies are restricted given that few studies estimated the global daily (short-term) PM_{2.5} exposure, and further comparison analyses are required.

Although ambient PM_{2.5} concentrations vary substantially across the world, few global land areas and populations are exposed to PM_{2.5} at concentrations lower than the 2021 WHO guideline limit.⁵ Based on modelled estimates for 2019, we found that 0.18% of the global land area and 0.001% of the global population had an annual exposure to PM_{2.5} lower than the WHO limit of 5 µg/m³, and more than 70% of days in the year, even in some high-income countries such as Singapore and Qatar, had a daily mean PM_{2.5} exposure above 15 µg/m³. Additionally, the high spatiotemporal global PM_{2.5} estimates provide valuable air pollution information for areas not covered by monitoring stations. For example, we observed a notable seasonal trend with regularly high regional PM_{2.5} concentrations in August and September in the Amazon rainforest region, where monitoring stations are scarce.

The spatiotemporal variations in PM_{2.5} concentrations might be the result of different types and components of anthropogenic fuel combustion emissions²⁷ and the changes in natural sources due to extreme weather events such as bushfires and windblown dust.²⁸ For example, northeast China had increased estimated PM_{2.5} concentrations in winter, which might stem from conducive weather patterns²⁹ and winter heating-related fossil fuel combustion,³⁰ whereas southern American countries such as Brazil had increased estimated PM_{2.5} concentrations in August and September, which might be associated with anthropogenic emissions such as slash-and-burn cultivation.³¹ By contrast, the increasing frequency and scale of climate change-related air pollution events, such as windblown dust and bushfire

events in 2019, might have contributed to the elevated PM_{2.5} concentrations in south-eastern Australia in 2019.³²

This study has several limitations. Despite conducting spatial and temporal cross-validations to test the robustness of estimations with stable results, biases might exist in the DEML model due to the sparse ground station distribution in most regions, limited availability of station data before 2014, and uncertainties from some predictors, such as the fixed anthropogenic emissions assumption in the chemical transport model data after 2013. In areas with few monitoring stations, the estimates should be interpreted with caution because our model predictions in these areas were only validated by a small number of monitoring stations, and the air quality records from these location-based ground stations are not representative of overall exposure variations across a wide region. In addition, despite a high spatial resolution, uncertainties exist in the aggregation of gridded estimates and calculation of population-weighted exposure and exposed days. Our study also cannot account for personal exposure, and assuming equivalent population exposures within each grid cell might lead to exposure misclassification due to population migration and different activity patterns and behaviours, such as time spent indoors and outdoors.

This study is the first to apply an innovative DEML approach to estimate high-resolution global daily PM_{2.5} exposure over two decades. Our DEML model showed accurate performance in predicting global and regional short-term and long-term PM_{2.5} concentrations, and provided a global perspective on the spatial and temporal variations of PM_{2.5} concentrations between 2000 and 2019. The results highlight the inequality of global air pollution population exposure, particularly in southern and eastern Asia. Our findings are of importance for global air pollution mitigation strategies and for assessing the short-term and long-term health effects of global PM_{2.5} exposure.

Contributors

YG and SL obtained funding. YG, JS, and SL are the study guarantors. YG and WY conceived the idea for the study. WY wrote the original draft and reviewed and edited the manuscript under the supervision of YG, JS, and SL. WY also performed the literature search, established the model, and analysed the data. TY was responsible for results visualisation and parts of the results presentation. RX, YL, WY, ZY, and YuZ contributed to data collection, cleaning, and preparation. RX and YL also verified the analytical methods. XY, YiZ, and ZC contributed to the interpretation of parts of the results and code improvement. YG and SL supervised the project. YG and WY accessed and verified the data. All authors had full access to all the data in the study. All authors discussed the results, provided critical feedback, helped revise the final manuscript, and had final responsibility for the decision to submit for publication.

Declaration of interests

We declare no competing interests.

Data sharing

The global annual mean PM_{2.5} data will be freely available in Monash Planetary Health Data Hub (<https://deepearth.erc.monash.edu/>), which is under development and may become available in 2023. The daily PM_{2.5} datasets and related study documents including the monitoring

station dataset are available upon request from the corresponding authors.

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