Ambient Air Pollution Exposure Estimation for the Global Burden of Disease 2013

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Abstract

Exposure to ambient air pollution is a major risk factor for global disease. Assessment of the impacts of air pollution on population health and the evaluation of trends relative to other major risk factors requires regularly updated, accurate, spatially resolved exposure estimates. We combined satellite-based estimates, chemical transport model (CTM) simulations and ground measurements from 79 different countries to produce new global estimates of annual average fine particle ($PM_{2.5}$) and ozone concentrations at 0.1° × 0.1° spatial resolution for five-year intervals from 1990-2010 and the year 2013. These estimates were then applied to assess population-weighted mean concentrations for 1990 – 2013 for each of 188 countries. In 2013, 87% of the world's population lived in areas exceeding the World Health Organization (WHO) Air Quality Guideline of 10 $\mu g/m^3$ $PM_{2.5}$ (annual average). Between 1990 and 2013, decreases in population-weighted mean concentrations of $PM_{2.5}$ were evident in most high income countries, in contrast to increases estimated in South Asia, throughout much of Southeast Asia, and in China. Population-weighted mean concentrations of ozone increased in most countries from 1990 - 2013, with modest decreases in North America, parts of Europe, and several countries in Southeast Asia.

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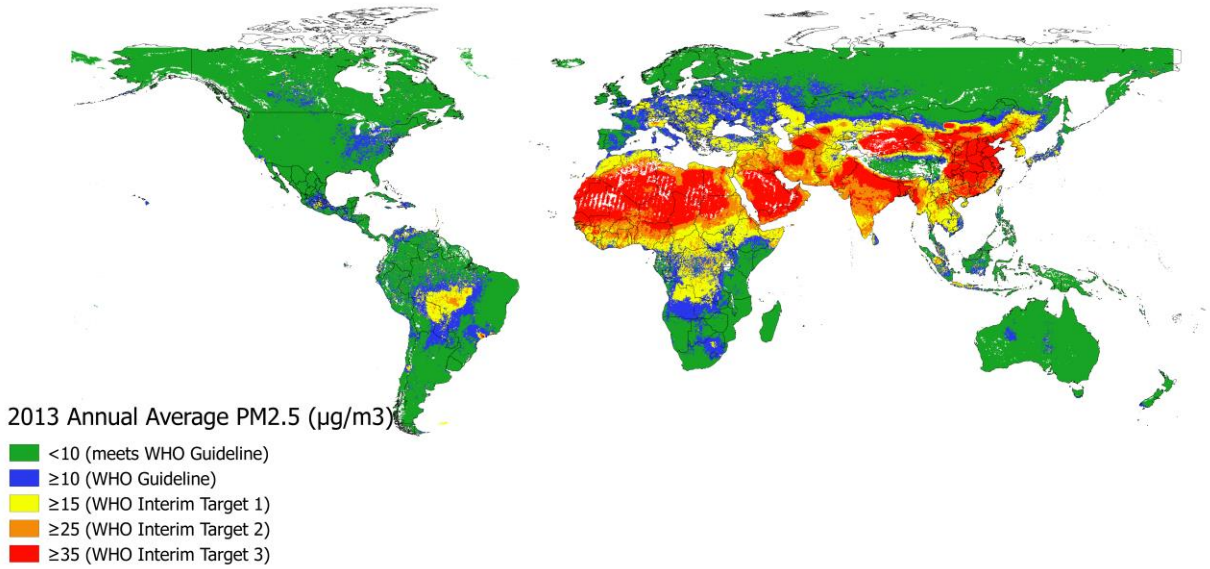

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Introduction

The Global Burden of Disease (GBD) 2010 provided important new estimates of the global health impacts attributable to ambient air pollution. Ambient particulate matter air pollution ($PM_{2.5}$, particulate matter with aerodynamic diameter 2.5 μm or smaller) was identified as a leading risk factor for global disease burden with an estimated 3.2 million attributable deaths in the year 2010. An additional 152,000 deaths in 2010 were attributable to long term exposure to ozone[1]. These two pollutants were selected as indicators of exposure to ambient air pollution based on extensive epidemiologic and mechanistic evidence indicating independent adverse health impacts[2]. These disease burden estimates incorporated novel high resolution global air pollution exposure estimates for 1990, 2005, and 2010 that included both urban and rural areas and that merged data from ground measurements, satellite retrievals and chemical transport models. In addition to the application of these exposure estimates to the entire global population, their high spatial resolution minimized biases due to spatial misalignment between exposure to air pollution and population information[3]. Beyond their direct use in the GBD 2010, the $PM_{2.5}$ exposure estimates were combined with chemical transport model simulations to estimate the sector-specific contributions to disease burden from motor vehicle transportation[4,5], household cooking with solid fuels[6] and household heating[7]. Additionally, these exposure estimates served as the basis of the World Health Organization (WHO) air pollution mortality estimates[8] and were used by the World Bank as indicators of sustainable development[9] and to estimate the economic damages attributable to air pollution[10]. The estimates have also been used in the U.S. Environmental Protection Agency's (USEPA) BenMap tool[11] which allows users to estimate health impacts and economic value of changes in air quality, and figured prominently in the exposure description of the International Agency for Research on Cancer Monograph on the

carcinogenicity of outdoor air pollution[12]. Given the lack of ground measurements in many regions of the world, the above analyses were not previously possible prior to development of consistent, globally applicable exposure estimates.

In the context of the most recent update of the Global Burden of Disease (GBD 2013)[13] we developed updates and revised global exposure estimates for $PM_{2.5}$ and ozone (O_3) with specific emphasis on evaluation of trends between 1990 – 2013. We also incorporated improvements in satellite-based estimation of $PM_{2.5}$[14], the availability of internally consistent emissions inventories spanning this entire period and a substantially increased number of ground measurements of particulate matter, compared to those utilized previously. Here we describe the methodology, provide descriptive information on year 2013 air pollution concentrations and illustrate trends since 1990, at the country level.

Methods

$PM_{2.5}$

The overall approach taken to estimate annual average $PM_{2.5}$ concentrations uses the mean of gridded values of satellite-based and chemical transport model estimates, calibrated to available ground measurements. In the previous GBD (2010) we used satellite-based estimates for $PM_{2.5}$ for the year 2005[15] combined with simulations from the TM5 (Tracer Model, version 5) chemical transport model[16] based on year 2005 emissions, which were the most current update available at the time of analysis. To estimate exposures for 1990 we used a TM5 simulation based on 1990 emissions but with the same "standard" meteorology as in the 2005 simulation. For the 1990

satellite-based estimates we used the same spatial pattern from the year 2005 estimates and adjusted this to 1990 based upon the ratio of 2005:1990 simulations from the GEOS-Chem chemical transport model[17] that used anthropogenic emissions from these years (while again maintaining consistent meteorology and natural emissions). For year 2010 estimates, we used the 2005 TM5 and satellite-based estimates and extrapolated these to 2010 based on the 1990 – 2005 trend following an assumption that the change proportional to the time period length from 2005 to 2010, relative to 2005 was 1/3 as large as the change from 1990 to 2005, relative to 1990 as described by the formula:

$$(PM_{2.5}\_2010-PM_{2.5}\_2005)/PM_{2.5}\_2005 = 1/3 * (PM_{2.5}\_2005 - PM_{2.5}\_1990)/PM_{2.5}\_1990. \quad (1)$$

In addition, we compiled available $PM_{2.5}$ (and where no $PM_{2.5}$ measurements were available, PM_{10}) annual average measurement data for ~2005 from a variety of sources[2]. These measurements were used to calibrate the average of the TM5 and satellite-based estimates for 2005 at the 0.1° x 0.1° grid-cell resolution with a simple prediction equation based upon agreement between the average of TM5 and satellite-based estimates for those locations with measurements.

For GBD 2013, we followed a similar approach but used updated inputs. Specifically, we used a new series of satellite-based estimates for $PM_{2.5}$ that included year-specific (3 year averages of retrievals centered on the specific year of interest) estimates for 1998 – 2012 and an improved estimation algorithm[14]. Briefly, satellite-based $PM_{2.5}$ estimates used aerosol optical depth (AOD) retrievals from satellites to estimate near-surface $PM_{2.5}$ by applying the relationship of $PM_{2.5}$ to AOD simulated by the GEOS-Chem chemical transport model. These updated $PM_{2.5}$ estimates make use of both "unconstrained" (as used on GBD 2010) and "optimal-estimation"

AOD retrievals in combination with the MODIS[18], MISR[19,20] and SeaWiFS[21] satellite-borne instruments. In the optimal estimation approach, AOD retrievals using observed MODIS top-of-atmosphere reflectance are constrained by simulated AOD based on their relative uncertainties. The $PM_{2.5}$ estimates produced by optimal estimation additionally used vertical profile information from the CALIOP[22] satellite instrument to inform about the relationship between column AOD and ground-level concentrations. These two (optimal estimation and unconstrained) sets of estimates were then combined with information on temporal variation based on SeaWiFS and MISR to estimate global $PM_{2.5}$ estimates (50% RH) at 0.1° x 0.1° for 2000, 2005, 2010 and 2011 (based on 3 years of retrievals, centered on the year of interest). Estimates for 1995 and 1990 were based on the ratio of GEOS-Chem simulations that used anthropogenic emissions (from EDGAR[23]) between 2005 and the respective year of interest, but constant meteorology[24].

We also included new TM5-FASST[25,26] (FAst Scenario Screening Tool, a reduced-form version of TM5) simulations for 1990, 2000, and 2010, using an updated set of emissions inventories, and constant meteorological inputs and emissions from dust and sea salt. TM5-FASST is a reduced form version of the TM5 chemical transport model that was used in the GBD 2010 exposure estimates. TM5 is a nested 3-dimensional global atmospheric chemistry transport model, which simulates ozone and aerosol components at 1° × 1° resolution[16]. TM5 FASST emulates the full TM5 chemical transport model with a set of linear relationships between emissions in 56 geographically defined source regions, and pollutant concentrations in receptor regions. TM5 FASST simulations are at a resolution of 1° x 1° grid cells that are sub-allocated based on population density (using the Gridded Population of the World, version 3, GPWv3[27]). Here we used a consistent set of emissions estimates [ECLIPSE (IEA) 2010, 2000, 1990

emissions; RCP: international aviation and shipping (2010); GFED v3.1: Forest fires and savannah burning (2000, 2010)] and "typical" (year 2001) meteorology, dust, and sea salt contributions with TM5-FASST to simulate ambient (50% RH) $PM_{2.5}$ concentrations at 0.1° x 0.1 ° for 1990, 2000 and 2010. Estimates for 1995 and 2005 were generated by fitting a natural cubic spline to the 1990, 2000 and 2010 estimates. Estimates for 2011 were estimated by fitting a natural spline to the 1990, 2000 and 2010 estimates, then extrapolating from the 2009 and 2010 fitted values. Full TM5 simulations for 1990 and 2005 (as used in GBD 2010) were also available.

To collect updated measurement data for 2010-2013, we used a variety of information sources including those used previously[28], as well as new data, especially from China and India, where available. We sought input from an international group of GBD collaborators, conducted targeted searches for data and included measurements compiled from a literature search[14] and from the WHO ambient air pollution in cities database[8]. A final database was constructed including measurement values, year of annual average (2010-2013 data were targeted – other years were used only if no other data were available), site coordinates (if available, or city centroid coordinates if not available), site type (if available), iso3 country code, data source and whether $PM_{2.5}$ was measured directly or estimated from a $PM_{2.5}$:PM_{10} ratio. All data sources are listed in the Supporting Information. Given the spatial biases in availability of ground measurements, differences in measurement approaches between jurisdictions, and absence of details regarding measurement data in some instances, the ground measurements were not used in evaluation of the exposure assessment methodology but rather incorporated within the approach as an additional source of information. For locations where daily values were obtained or where measurement completeness was available we retained all sites with >70% valid measurements,

as reported in the source databases. For locations where data were obtained for multiple years at the same location we retained one value per location, between the years 2008-2013 in the following order of preference: 2010, 2011, 2009, 2012, 2008, 2013 to best match the temporal scale of the satellite-based and TM5-FASST estimates.

If multiple monitoring types available at the same site were included in parent databases, these values were averaged. Industrial and roadside sites were excluded (where indicated in source databases; except in India where these were retained upon the advice of local experts who indicated that these sites adequately represent population exposure in that country). In the USEPA database, sites indicated as "background surveillance" were designated as to background, those indicated as "continuous air monitoring program", "population-oriented monitoring", or "exposure studies" were designated as population sites. Those sites identified as "episode monitoring" or "complaint investigation", were excluded. All others were identified as unspecified and retained. Measurements flagged as “events,” in the USEPA database were also excluded.

In locations where no $PM_{2.5}$ measurements were available, we estimated $PM_{2.5}$ from measurements of PM_{10}[2,29]. We preferentially used $PM_{2.5}$:PM_{10} ratios that were locally derived. In these cases ratios from any sites within 50 km where both PM_{10} and $PM_{2.5}$ were measured were used to estimate $PM_{2.5}$ from PM_{10} measurements. These local ratios were only accepted and used to derive $PM_{2.5}$ estimates for nearby sites if they were between 0.2 and 0.8. If local ratios were not available we used a country and monitor-specific average if available, followed by a country-specific average. Otherwise, we assumed a ratio of 0.5 to estimate $PM_{2.5}$ from PM_{10} measurements.

In total we included 4073 data points from 3387 unique locations (1,854 [46%] from direct measurements of $PM_{2.5}$) in 79 countries (Figure S1; Supporting Information). Of the 2219 data points in which $PM_{2.5}$ was estimated from PM_{10}, 1,151 (30% of 4073 total) were estimated using a ratio derived from monitors within 50 km, 590 (15%) were estimated using in-country monitors of the same type, and 309 (8%) were estimated with other in-country monitors. Finally, the remaining 169 (4%) were estimated with a ratio of 0.5, as used for GBD 2010. The mean ratio for the estimation was 0.61.

We then used a regression calibration approach to combine the mean of the satellite-based estimates and the TM5-FASST simulations with the measurements to produce final global estimates at 0.1° x 0.1 °grid-cell resolution. For the regression calibration, we initially evaluated a simple regression model:

$$\text{Measured } \ln(PM_{2.5}) = \beta_0 + \beta_1 * \ln(\text{fused}) \quad (2)$$

where fused is the mean of the satellite-derived and TM5 estimates for each grid cell. We first tested whether to apply a single global calibration function or one that varied by the 21 GBD regions. Modeling with a random effect by 7 aggregated “super-regions” (due to the complete absence of measurements in multiple regions) indicated some regional variation in slopes but also some very poor fits in some regions, so a single global calibration function was chosen. Evaluation of model residuals indicated no association with population density and addition of population density to the model only minimally improved fit. We then evaluated the impact of including available information on the measurement values and measurement sites including whether the exact site coordinates were known, whether $PM_{2.5}$ was directly measured or estimated and whether the monitoring site classification was known or unspecified. Inclusion of

these variables slightly improved the model R^2 while slightly reducing the residual standard error.

These two candidate global calibration models (Simple model: Intercept= 0.82, ln(fused)= 0.73, residual standard error = 0.43, Multiple R-squared: 0.60, Adjusted R-squared: 0.60; "Advanced" model with additional site parameters: Intercept= 0.42, ln(fused)=0.87, residual standard error = 0.41, Multiple R-squared: 0.64, Adjusted R-squared: 0.64) were further evaluated by a cross-validation procedure in which 10% of the measurement sites were randomly selected for model evaluation. This procedure was repeated for a total of 3 separate sets of 10% testing sites. In all cases the model with additional site parameters ("Advanced", Figure S2; Supporting Information) had a lower RMSE (12.17 vs 11.04; 12.17 vs 11.04; 10.21 vs 9.15) as well as lower Akaike's Information Criterion and Bayesian Information Criterion. Therefore, we selected the model with additional site parameters that included variables to indicate approximate location, unspecified monitor type, and $PM_{2.5}$ calculated from PM_{10}. A plot of these global calibration functions against the data from ground measurements and the mean of the satellite-based and TM5 estimates for the corresponding grid cells is provided in the Supporting Information (Figure S2; Supporting Information). The relevant terms from this calibration function were then used to adjust the gridded values of the mean of the satellite-based and TM5 estimates ("fused") as follows:

$$\text{Calibrated } PM_{2.5} = \exp[0.41765+(0.86953*\ln(\text{fused}))] \quad (3)$$

Estimates for 2013 were generated by extrapolating from the trend between the 2010 and 2011 fused and calibrated values. Specifically, for these extrapolations we assessed the rate of change

in concentrations between 2010 and 2011 and applied this growth rate to an exponential growth function to estimates concentrations in 2013, with the following function:

$$PM_{2.5}\_2013 = PM_{2.5}\_2010*\exp(\text{rate.of.change} *(2013-2010)) \quad (4)$$

where, (annual) rate.of.change = $\ln(PM_{2.5}\_2011/ PM_{2.5}\_2010)$. The final $PM_{2.5}$ estimates used in the burden of disease estimation and presented in the Results section are the fused, calibrated, and (where applicable) extrapolated values. National level population-weighted mean and 95% uncertainty interval concentrations were estimated by sampling 1000 fused grid cell concentrations from each country in combination with the calibration parameters and the standard error of the calibration function. These draws were then weighted by the corresponding grid cell population value (using the Gridded Population of the World, version 3, GPWv3[27]).

Ozone

As in GBD 2010, we calculated a running 3-month average (of daily 1 hour maximum values) for each grid cell over a full year and selected the maximum of these values. This metric was chosen to align with epidemiologic studies of chronic exposure which typically employ a seasonal (summer) average, and to account for global variation in the timing of the ozone (summer) season[28]. As described above, these estimates were simulated with TM5-FASST at 0.1° x 0.1° for 1990, 2000 and 2010 using the same emissions datasets and meteorological inputs as for the $PM_{2.5}$ simulations. Estimates for 1995, 2005, and 2011 were generated by fitting a natural cubic spline in the same manner as described previously for $PM_{2.5}$. As described above for $PM_{2.5}$, an exponential growth model for ozone concentrations was used to estimate 2013 concentrations from the 2011 estimates. Population weighted mean concentrations and 95% uncertainty intervals for each country were estimated as described above for $PM_{2.5}$ and assuming

a normal distribution with a standard deviation calculated by assuming an uncertainty interval of ± 6% of the estimated concentration. As in the GBD 2010 exposure estimates and given the scarcity of surface ozone measurements throughout the world and the challenges in accessing hourly data from available monitoring sites to develop the desired metric, we did not utilize surface ozone measurements for developing the global estimates.

Results

Of the ground measurements, while 79 countries were represented, more than half were from high income countries in North America (25%), Western (32%) and Central (12%) Europe. Because of a major expansion of China's air quality monitoring network, 10% of the ground measurements were from East Asia, and 5% were from South Asia. Countries in eleven regions (Andean Latin America, Australasia, Central Asia, Eastern Europe, High-income Asia Pacific, North Africa and Middle East, Southeast Asia, Southern Latin America, Southern Sub-Saharan Africa, Tropical Latin America, Western Sub-Saharan Africa) each contributed less than 3% of the measurement data, with no measurements at all from the other four regions (Caribbean, Central Latin America, East and Central sub-Saharan Africa) of the world. The highest measured annual average $PM_{2.5}$ concentration in the assembled measurement database was 194 $\mu g/m^3$ in Shijiazhuang, the capital of Hebei Province in China, while the lowest was <1 $\mu g/m^3$, measured in Soldotna, Alaska, USA.

Year 2013 gridded estimates of annual average $PM_{2.5}$ and seasonal hourly maximum ozone concentrations are displayed in Figures 1 and 2. The highest concentrations of $PM_{2.5}$ were evident in northern Africa and the Middle East due to emissions of windblown mineral dust, and

256 in South and East Asia, especially in northern India and eastern China due to combustion
257 emissions from multiple sources including household solid fuel use, coal fired power plant
258 emissions, agricultural burning, industrial and transportation-related emissions. Ozone
259 concentrations were less variable spatially, but relatively higher in parts of the U.S., the Amazon
260 Basin, sub-Saharan Africa and throughout much of southern Europe, the Middle East and Asia.

261 The relationship between the spatial distribution of ambient concentrations and that of the
262 population is particularly relevant to health burden assessment (Figure 3). Based upon the grid-
263 cell concentration estimates and corresponding population data, 35% of global population
264 resided in areas with concentrations above the WHO Interim Target 1 of 35 $\mu g/m^3$ annual
265 average $PM_{2.5}$ with nearly all of the most extreme (> 65 $\mu g/m^3$) concentrations experienced by
266 populations in China and India. Fully 87% of the global population resided in areas above the
267 WHO guideline of 10 $\mu g/m^3$, with essentially none of the population of China (0.4%) or India
268 (0.01%) living in areas meeting this level.

269 Changes in estimated concentrations between 1990 and 2013 at the 0.1° x 0.1° grid cell level are
270 shown in Figure 4. Large relative decreases were evident in the Eastern US., Europe, Russia and
271 in parts of Southeast Asia. In contrast, large relative increases were apparent in Western Canada,
272 parts of South America, the Middle East, India and China. Somewhat similar patterns were also
273 evident for ozone (Figure 5)

274 The trends in $PM_{2.5}$ are examined in more detail in Figures 6a-c which display population-
275 weighted mean (95% uncertainty interval) distributions of concentrations at the country level for
276 selected regions. Plots including all other countries are presented in the Supporting Information
277 (Figures S3-S6), along with all of the country-level population-weighted exposure data (mean,

95% uncertainty interval) for both $PM_{2.5}$ and ozone for 1990, 1995, 2000, 2005, 2010 and 2013) (File S1; Supporting Information). By way of example, Table 1 presents population-weighted estimates for $PM_{2.5}$ and ozone for 1990 and 2013 for the world's ten most populous countries. Large proportional increases in mean population-weighted $PM_{2.5}$ concentrations were apparent in India, China, Brazil, Bangladesh, India, China and Pakistan, with decreases observed in the U.S., Indonesia, Russia, Japan, and Nigeria.

Between 1990 and 2013, decreases in population-weighted mean concentrations of $PM_{2.5}$ were evident in most of the high income countries (Figure 6a), in contrast to consistent increases in South Asia (Figure 6b), much of Southeast Asia, and especially in China (Figure 6c). At the country-level, the highest population-weighted mean concentration estimated for 2013 was Mauritania (70 $\mu g/m^3$), followed by China (55 $\mu g/m^3$), Saudi Arabia (54 $\mu g/m^3$), Kuwait (49 $\mu g/m^3$), Bangladesh (48 $\mu g/m^3$), India (47 $\mu g/m^3$), Pakistan and Nepal (46 $\mu g/m^3$). The lowest country-level population-weighted estimates were for several Pacific and Caribbean island nations, Australia and Norway (≤ 6 $\mu g/m^3$). Population-weighted ozone levels also increased throughout most of the most heavily populated countries from 1990 - 2013, although to a lesser degree than seen for $PM_{2.5}$. There were modest decreases in population-weighted ozone levels in the U.S., Mexico and Canada as well as parts of Europe and several countries in southeast Asia (Indonesia, Malaysia, Singapore) (Table S1; Supporting Information).

We also compared estimates using the updated methodology described in this manuscript for the year 2010 to those reported previously from GBD 2010[2] for the same year (Figure S7; Supporting Information). This is a comparison of methodology, in contrast to the description of temporal changes described above. Our updated estimates resulted in lower levels in areas of North Africa, the Middle East and the Gobi Desert, all areas that were impacted by high levels of

windblown mineral dust. Slightly higher levels were estimated with the updated methodology in Brazil and elsewhere in South America with noticeably higher levels in India, Pakistan and Bangladesh. Somewhat lower levels were also estimated for some areas of eastern China. These differences may be due to additional temporal coverage that is incorporated into the current satellite-based and ground measurements. In North Africa the lower levels reflect changes in mineral dust emissions that were driven by changes in local meteorology, and the higher levels in India, Bangladesh and Pakistan likely reflect increases in emissions that may not have been represented in the earlier methodology that extrapolated 2005 estimates to 2010. In China, the somewhat lower levels were likely affected by the inclusion in the calibration of substantially more ground measurements of high concentrations from China which tend to reduce the slope of the calibration function (see Figure S2; Supporting Information).

At the country-level, population-weighted mean estimates for $PM_{2.5}$ for the year 2010 were very similar for most countries between those estimated for GBD 2010 and GBD 2013, and discrepancies reflect the same patterns described above. Estimates for China were noticeably lower for GBD 2013 (54.8 $\mu g/m^3$) compared to GBD 2010 (72.6 $\mu g/m^3$), as were several countries with high contributions from windblown mineral dust (Saudi Arabia: 53.8 vs 61.7 $\mu g/m^3$; Qatar: 40.1 vs 69.0 $\mu g/m^3$; United Arab Emirates: 40.9 vs 79.5 $\mu g/m^3$ for GBD 2013 vs GBD 2010, respectively). Higher levels were estimated in GBD 2013 for countries in South Asia (India: 43.4 vs 32.0; Bangladesh: 45.7 vs 31.1 6; Pakistan: 43.4 vs 38.1; Nepal: 41.5 vs 32.7 $\mu g/m^3$ for GBD 2013 vs GBD 2010, respectively).

Discussion

We have extended previous global estimates of long-term average exposure to $PM_{2.5}$ and ozone at 0.1 x 0.1° resolution to support the Global Burden of Disease 2013. Specifically, we applied improved satellite-based estimates that also incorporated additional information on temporal trends, as well as chemical transport model simulations incorporating internally consistent emissions trends from 1990 – 2013. Further, we have incorporated a substantially larger number of available surface measurements of $PM_{2.5}$ to calibrate the estimates based on satellite retrievals and chemical transport model simulations. Given the increasing emphasis with in the Global Burden of Disease on country-level (and sub-country) reporting, we also provided population-weighted estimates of exposure to $PM_{2.5}$ and ozone for 188 different countries for the years 1990, 1995, 2000, 2005, 2010 and 2013. As such, these data represent one of the most extensive collections of global air pollution concentration estimates produced to date. Given the advances in our methodology used to develop these estimates, compared to those previously reported[2], we consider these estimates to be more accurate.

Although these estimates incorporated recent advances in satellite-based estimation, newly developed emissions inventories and substantially more ground measurements, they are not without limitations. Given the need to produce similar estimates in future years in support of regular updates to the Global Burden of Disease[30] and other assessments, we anticipate further enhancements to the methodology to address these limitations. Specifically, we note that there is still poor agreement between these estimates and ground-based measurements in some locations, notably in parts of South America (e.g. Chile), southern Poland and Turkey, and in specific urban areas with high levels of ambient $PM_{2.5}$ such as Ulaanbaatar, Mongolia (Figure S8; Supporting Information) where ground measurements were underestimated by our approach.

The same underestimation of ground level measurements in southern Poland and Ulaanbaatar was identified by van Donkelaar et al, who suggested that higher wintertime (and in the case of Ulaanbaatar also nighttime) emissions when satellite retrievals are more limited due to more frequent winter cloud cover (or unavailable at night) are likely to be responsible[14]. A similar phenomenon may also be contributing to poor agreement in Chile where winter, nighttime wood burning is a major contributor to elevated $PM_{2.5}$ concentrations[31,32]. We do note however, that these same discrepancies and general underestimation of ground measurements in specific locations are not restricted to the satellite-based estimates as they are also evident in TM5-FASST simulations suggesting that both approaches may fail to accurately estimate ground-level $PM_{2,5}$ in relatively small areas having very high levels.

In addition, we used available PM_{10} ground measurements to estimate $PM_{2.5}$ concentrations in locations where $PM_{2.5}$ measurements were not available. Our use of $PM_{2.5}$:PM_{10} ratios for the measurement calibration dataset represents a balance between measurement specificity and spatial representativeness of the ground measurements. For example in North Africa and the Middle East, South Asia, and high-income Asia Pacific countries, less than 20% of available ground measurements were of $PM_{2.5}$. Despite increases in the number of available $PM_{2.5}$ measurements used in our calibration (4073 in this analysis compared to 679 in that reported previously[2]), more than 50% of the ground measurements that were used were estimated from PM_{10} measurements. Given the importance of these estimates in the calibration, we placed additional importance on using location-specific $PM_{2.5}$:PM_{10} ratios to estimate $PM_{2.5}$ concentrations, for example using ratios from measurements within 50km in 30% of cases and in-country ratios for an additional 23% of cases. Further, our regression calibration included a

term to account for the use of direct vs estimated $PM_{2.5}$ measurements. Van Donkelaar et al., using the same satellite-derived estimates included here, reported 58%, 53% and 65% of explained variability in ground measurements of $PM_{2.5}$ in North America, Europe and elsewhere, respectively[14]. In comparison, our candidate simple regression model, which treated directly measured and estimated ground $PM_{2.5}$ measurements equally, explained 60% of variability in ground measurements, while the (advanced) calibration model that was ultimately used explained slightly more variability (64%). Overall, there is a need for additional $PM_{2.5}$ measurements and greater global coverage. The establishment of a new global network to address these shortcomings and to improve the capability of satellite-based estimates of global particulate matter levels offers the promise of enhanced accuracy and representativeness[33]. The need for additional air pollution measurements was also specifically highlighted in the recent World Health Assembly Resolution on Air Pollution[34].

High levels of uncertainty in our estimates exist in regions of elevated windblown mineral dust. The latter is partially driven by TM5-FASST use of standard dust contributions that do not align with a specific year and the temporally variable levels of resuspended mineral dust in affected regions. As we observed variability between regions in the function used to calibrate the fused estimates ground measurements, there is a need to improve the incorporation of measurement information. For example, in future work we aim to make use of the increasing number of surface measurements, especially those in China, and to implement advanced approaches to incorporate ground measurements and other information more directly into the estimates. Future availability of additional ground measurements may also increase the feasibility of allowing spatially varying calibration functions. This might be achieved by geographically weighted

392 regression[35] or hierarchical modelling. For the latter, calibration models for different regions may
393 include a global calibration together with regional random effects. Such models sit naturally
394 within a Bayesian framework. Recent advances in computational methods for inference, for
395 example those based on Integrated Nested Laplace Approximations[36,37], have increased the
396 feasibility of implementing such complex models in this setting. Harmonization of measurement
397 approaches between jurisdictions would also be beneficial.

398 Because we estimated year 2013 concentrations based upon year 2010 concentrations and trends
399 in satellite-based estimates from 2010 to 2011, differences between estimates for 2010 and 2013
400 from this analysis may be overly influenced by short-term variability in meteorology rather than
401 longer-term trends in emissions, the latter being more stable over the 5-year periods included in
402 our estimates. However, as noted in Methods, the annual satellite-based estimates are 3-year
403 moving averages, centered on the year of interest (for example 2010 includes retrievals from
404 2009-2011) which should partially mitigate this instability.

405 The updated global exposure estimates presented here represent a further advance in
406 characterizing global population exposure to ambient air pollution for use in burden of disease
407 assessment and other impact analyses. The global coverage allows for estimation of
408 concentrations in areas without extensive ground monitoring, including for example, rural areas
409 with large emissions from household use of solid fuels[6]. Like our previous GBD 2010 estimates,
410 these are based on $PM_{2.5}$ mass concentrations. Although there is considerable interest in, and
411 active research concerning, the effects of the myriad constituents of ambient air pollution, ozone
412 and $PM_{2.5}$ mass concentrations remain the most robust and consistent indicators of health-
413 damaging air pollution from combustion and other major sources[38,39]. Use of these estimates in
414 combination with chemical transport model simulations can provide information on sector-

specific contributions to ambient concentrations and disease burden to inform air quality management[4–7]. Improvements in the quality of these estimates and the application of a consistent methodology to evaluate temporal trends in exposure over a 23 year period should inform the choice of air quality management strategies and other approaches to mitigate the health impacts of air pollution exposure. Accordingly, we are committed to making these estimates available for others to use and have provided the associated files of country-level population weighted (File S1) and gridded estimates (linked to population data and urbanicity indicators, File S2) and a data dictionary (File S3) in the Supporting Information. Given the evidence indicating the importance of ambient air pollution to global disease burden[1,8], we anticipate a need for regular updating and improving of these estimates and their use in policy assessments and comparative analyses.

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Country	1990 $PM_{2.5}$	2013 $PM_{2.5}$	% Change	1990 Ozone	2013 Ozone	% Change
China	39.3	54.3	38.0	57.0	64.5	13.2
India	30.2	46.7	54.3	61.5	74.0	20.2
United States	16.4	10.7	-34.5	70.3	67.0	-4.7
Indonesia	21.0	14.8	-29.7	47.3	39.6	-16.1
Brazil	9.7	16.5	70.4	43.4	51.0	17.3
Pakistan	36.5	46.2	26.3	59.0	68.8	16.5
Nigeria	31.0	29.5	-4.7	66.3	67.5	1.9
Bangladesh	29.9	48.3	61.6	59.4	72.0	21.3
Russia	19.7	14.2	-27.6	48.6	48.3	-0.6
Japan	19.4	16.0	-17.5	56.8	60.5	6.7

438

439 **Table 1**. 1990, 2013 and percent change since 1990 in annual average $PM_{2.5}$ ($\mu g/m^3$) and
440 seasonal mean 1 hour daily maximum ozone (ppb) concentrations for the world's ten most
441 populous countries.

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