234   a normal distribution with a standard deviation calculated by assuming an uncertainty interval of ±

235   6% of the estimated concentration. As in the GBD 2010 exposure estimates and given the scarcity

236   of surface ozone measurements throughout the world and the challenges in accessing hourly data

237   from available monitoring sites to develop the desired metric, we did not utilize surface ozone

238   measurements for developing the global estimates.

239

240   **Results**

241   Of the ground measurements, while 79 countries were represented, more than half were from

242   high income countries in North America (25%), Western (32%) and Central (12%) Europe.

243   Because of a major expansion of China's air quality monitoring network, 10% of the ground

244   measurements were from East Asia, and 5% were from South Asia. Countries in eleven regions

245   (Andean Latin America, Australasia, Central Asia, Eastern Europe, High-income Asia Pacific,

246   North Africa and Middle East, Southeast Asia, Southern Latin America, Southern Sub-Saharan

247   Africa, Tropical Latin America, Western Sub-Saharan Africa) each contributed less than 3% of

248   the measurement data, with no measurements at all from the other four regions (Caribbean,

249   Central Latin America, East and Central sub-Saharan Africa) of the world. The highest

250   measured annual average $PM_{2.5}$ concentration in the assembled measurement database was 194

251   $\mu g/m^3$ in Shijiazhuang, the capital of Hebei Province in China, while the lowest was <1 $\mu g/m^3$,

252   measured in Soldotna, Alaska, USA.

253   Year 2013 gridded estimates of annual average $PM_{2.5}$ and seasonal hourly maximum ozone

254   concentrations are displayed in Figures 1 and 2. The highest concentrations of $PM_{2.5}$ were

255   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,

14

278   95% uncertainty interval) for both PM$_{2.5}$ and ozone for 1990, 1995, 2000, 2005, 2010 and 2013)

279   (File S1; Supporting Information). By way of example, Table 1 presents population-weighted

280   estimates for PM$_{2.5}$ and ozone for 1990 and 2013 for the world's ten most populous countries.

281   Large proportional increases in mean population-weighted PM$_{2.5}$ concentrations were apparent in

282   India, China, Brazil, Bangladesh, India, China and Pakistan, with decreases observed in the U.S.,

283   Indonesia, Russia, Japan, and Nigeria.

284   Between 1990 and 2013, decreases in population-weighted mean concentrations of PM$_{2.5}$ were

285   evident in most of the high income countries (Figure 6a), in contrast to consistent increases in

286   South Asia (Figure 6b), much of Southeast Asia, and especially in China (Figure 6c). At the

287   country-level, the highest population-weighted mean concentration estimated for 2013 was

288   Mauritania (70 µg/m^3), followed by China (55 µg/m^3), Saudi Arabia (54 µg/m^3), Kuwait (49

289   µg/m^3), Bangladesh (48 µg/m^3), India (47 µg/m^3), Pakistan and Nepal (46 µg/m^3). The lowest

290   country-level population-weighted estimates were for several Pacific and Caribbean island

291   nations, Australia and Norway ($\leq$ 6 µg/m^3). Population-weighted ozone levels also increased

292   throughout most of the most heavily populated countries from 1990 - 2013, although to a lesser

293   degree than seen for PM$_{2.5}$. There were modest decreases in population-weighted ozone levels in

294   the U.S., Mexico and Canada as well as parts of Europe and several countries in southeast Asia

295   (Indonesia, Malaysia, Singapore) (Table S1; Supporting Information).

296   We also compared estimates using the updated methodology described in this manuscript for the

297   year 2010 to those reported previously from GBD 2010[2] for the same year (Figure S7;

298   Supporting Information). This is a comparison of methodology, in contrast to the description of

299   temporal changes described above. Our updated estimates resulted in lower levels in areas of

300   North Africa, the Middle East and the Gobi Desert, all areas that were impacted by high levels of

15

301   windblown mineral dust. Slightly higher levels were estimated with the updated methodology in

302   Brazil and elsewhere in South America with noticeably higher levels in India, Pakistan and

303   Bangladesh. Somewhat lower levels were also estimated for some areas of eastern China. These

304   differences may be due to additional temporal coverage that is incorporated into the current

305   satellite-based and ground measurements. In North Africa the lower levels reflect changes in

306   mineral dust emissions that were driven by changes in local meteorology, and the higher levels in

307   India, Bangladesh and Pakistan likely reflect increases in emissions that may not have been

308   represented in the earlier methodology that extrapolated 2005 estimates to 2010. In China, the

309   somewhat lower levels were likely affected by the inclusion in the calibration of substantially

310   more ground measurements of high concentrations from China which tend to reduce the slope of

311   the calibration function (see Figure S2; Supporting Information).

312   At the country-level, population-weighted mean estimates for $PM_{2.5}$ for the year 2010 were very

313   similar for most countries between those estimated for GBD 2010 and GBD 2013, and

314   discrepancies reflect the same patterns described above. Estimates for China were noticeably

315   lower for GBD 2013 (54.8 µg/m^3) compared to GBD 2010 (72.6 µg/m^3), as were several

316   countries with high contributions from windblown mineral dust (Saudi Arabia: 53.8 vs 61.7

317   µg/m^3; Qatar: 40.1 vs 69.0 µg/m^3; United Arab Emirates: 40.9 vs 79.5 µg/m^3 for GBD 2013 vs

318   GBD 2010, respectively). Higher levels were estimated in GBD 2013 for countries in South Asia

319   (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

320   µg/m^3 for GBD 2013 vs GBD 2010, respectively).

321

322   **Discussion**

16

323   We have extended previous global estimates of long-term average exposure to $PM_{2.5}$ and ozone

324   at 0.1 x 0.1° resolution to support the Global Burden of Disease 2013. Specifically, we applied

325   improved satellite-based estimates that also incorporated additional information on temporal

326   trends, as well as chemical transport model simulations incorporating internally consistent

327   emissions trends from 1990 – 2013. Further, we have incorporated a substantially larger number

328   of available surface measurements of $PM_{2.5}$ to calibrate the estimates based on satellite retrievals

329   and chemical transport model simulations. Given the increasing emphasis with in the Global

330   Burden of Disease on country-level (and sub-country) reporting, we also provided population-

331   weighted estimates of exposure to $PM_{2.5}$ and ozone for 188 different countries for the years 1990,

332   1995, 2000, 2005, 2010 and 2013. As such, these data represent one of the most extensive

333   collections of global air pollution concentration estimates produced to date. Given the advances

334   in our methodology used to develop these estimates, compared to those previously reported[2], we

335   consider these estimates to be more accurate.

336

337   Although these estimates incorporated recent advances in satellite-based estimation, newly

338   developed emissions inventories and substantially more ground measurements, they are not

339   without limitations. Given the need to produce similar estimates in future years in support of

340   regular updates to the Global Burden of Disease[30] and other assessments, we anticipate further

341   enhancements to the methodology to address these limitations. Specifically, we note that there is

342   still poor agreement between these estimates and ground-based measurements in some locations,

343   notably in parts of South America (e.g. Chile), southern Poland and Turkey, and in specific

344   urban areas with high levels of ambient $PM_{2.5}$ such as Ulaanbaatar, Mongolia (Figure S8;

345   Supporting Information) where ground measurements were underestimated by our approach.

346   The same underestimation of ground level measurements in southern Poland and Ulaanbaatar

347   was identified by van Donkelaar et al, who suggested that higher wintertime (and in the case of

348   Ulaanbaatar also nighttime) emissions when satellite retrievals are more limited due to more

349   frequent winter cloud cover (or unavailable at night) are likely to be responsible[14]. A similar

350   phenomenon may also be contributing to poor agreement in Chile where winter, nighttime wood

351   burning is a major contributor to elevated $PM_{2.5}$ concentrations[31,32]. We do note however, that

352   these same discrepancies and general underestimation of ground measurements in specific

353   locations are not restricted to the satellite-based estimates as they are also evident in TM5-

354   FASST simulations suggesting that both approaches may fail to accurately estimate ground-level

355    $PM_{2,5}$ in relatively small areas having very high levels.

356

357   In addition, we used available PM_{10} ground measurements to estimate $PM_{2.5}$ concentrations in

358   locations where $PM_{2.5}$ measurements were not available. Our use of $PM_{2.5}$:PM_{10} ratios for the

359   measurement calibration dataset represents a balance between measurement specificity and

360   spatial representativeness of the ground measurements. For example in North Africa and the

361   Middle East, South Asia, and high-income Asia Pacific countries, less than 20% of available

362   ground measurements were of $PM_{2.5}$. Despite increases in the number of available $PM_{2.5}$

363   measurements used in our calibration (4073 in this analysis compared to 679 in that reported

364   previously[2]), more than 50% of the ground measurements that were used were estimated from

365   PM_{10} measurements. Given the importance of these estimates in the calibration, we placed

366   additional importance on using location-specific $PM_{2.5}$:PM_{10} ratios to estimate $PM_{2.5}$

367   concentrations, for example using ratios from measurements within 50km in 30% of cases and

368   in-country ratios for an additional 23% of cases. Further, our regression calibration included a

369   term to account for the use of direct vs estimated $PM_{2.5}$ measurements. Van Donkelaar et al.,

370   using the same satellite-derived estimates included here, reported 58%, 53% and 65% of

371   explained variability in ground measurements of $PM_{2.5}$ in North America, Europe and elsewhere,

372   respectively[14]. In comparison, our candidate simple regression model, which treated directly

373   measured and estimated ground $PM_{2.5}$ measurements equally, explained 60% of variability in

374   ground measurements, while the (advanced) calibration model that was ultimately used

375   explained slightly more variability (64%). Overall, there is a need for additional $PM_{2.5}$

376   measurements and greater global coverage. The establishment of a new global network to

377   address these shortcomings and to improve the capability of satellite-based estimates of global

378   particulate matter levels offers the promise of enhanced accuracy and representativeness[33]. The

379   need for additional air pollution measurements was also specifically highlighted in the recent

380   World Health Assembly Resolution on Air Pollution[34].

381

382   High levels of uncertainty in our estimates exist in regions of elevated windblown mineral dust.

383   The latter is partially driven by TM5-FASST use of standard dust contributions that do not align

384   with a specific year and the temporally variable levels of resuspended mineral dust in affected

385   regions. As we observed variability between regions in the function used to calibrate the fused

386   estimates ground measurements, there is a need to improve the incorporation of measurement

387   information. For example, in future work we aim to make use of the increasing number of

388   surface measurements, especially those in China, and to implement advanced approaches to

389   incorporate ground measurements and other information more directly into the estimates. Future

390   availability of additional ground measurements may also increase the feasibility of allowing

391   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-

20

415   specific contributions to ambient concentrations and disease burden to inform air quality

416   management[4–7]. Improvements in the quality of these estimates and the application of a

417   consistent methodology to evaluate temporal trends in exposure over a 23 year period should

418   inform the choice of air quality management strategies and other approaches to mitigate the

419   health impacts of air pollution exposure. Accordingly, we are committed to making these

420   estimates available for others to use and have provided the associated files of country-level

421   population weighted (File S1) and gridded estimates (linked to population data and urbanicity

422   indicators, File S2) and a data dictionary (File S3) in the Supporting Information. Given the

423   evidence indicating the importance of ambient air pollution to global disease burden[1,8], we

424   anticipate a need for regular updating and improving of these estimates and their use in policy

425   assessments and comparative analyses.

426

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437

Country	1990 PM2.5	2013 PM2.5	% 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

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

442

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