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Supplementary Information for

Global impact of atmospheric arsenic on health risk: 2005-2015

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Supplementary Information Text

1. The development and improvement of arsenic emission inventory in 2005 and 2015

We first discuss the arsenic emission inventory in 2005. For Type I emission source, the same methods as Wai, *et al.* (1) are applied for U.S. (https://www.epa.gov/air-emissions-inventories/pollutant-emissions-summary-files-earlier-neis), Europe (https://prtr.eea.europa.eu/#/home), Australia (http://www.npi.gov.au/resource/arsenic-and-compounds-0) and Canada (https://www.canada.ca/en/environment-climate-change/services/national-pollutant-release-inventory/about-national-pollutant-release-inventory.html). Differently, the arsenic emission in Japan is replaced by Wai, *et al.* (1), the emission inventory from Japan's Pollution and Transfer Register (J-PRTR) (http://www2.env.go.jp/chemi/prtr/prtrinfo/contents/e-table.jsp), relative to Wai, *et al.* (1) in which arsenic emission was estimated based on SO_2 from EDGAR. For Type II emission source, the atmospheric arsenic emissions are mostly from copper smelters considering the world's largest copper mine production over there. The original inventory (1) only considers atmospheric arsenic emissions from central and northern Chile (2), whereas the southern copper smelter emissions were added based on copper production data from the Chilean government mining department (https://www.cochilco.cl) and emission factor from Pacyna and Pacyna (3) in this study. For anthropogenic arsenic emissions

in China, the original inventory only takes into account of three non-ferrous metals (Cu, Pb and Zn) smelting emissions and the coal-fired power plant emissions (1). However, there are large arsenic emissions from three other non-ferrous metals (Sn, Sb and W) smelting (4, 5), as well as household and industrial coal combustion (6), Iron and steel production (7, 8), cement production (9, 10) and liquid fuels combustion (11, 12), which are included in this study. For Type III emission source, the method in this study is the same as that used in Wai, *et al.* (1).

For the arsenic emission inventory in 2015, the same method as that of 2005 emission inventory is applied in general, with further improvement over China. Please note that for regions with arsenic emission estimation in 2015 based on As/S (Type III), the sulfur in the year of 2010 was used due to data availability of EDGAR (13), which may lead to certain overestimation, however, the change rate of sulfur after 2010 is in general small (Fig. 5a of Hoesly, *et al.* (14)). Over China, the emission inventory for 2015 has recently been developed by Liu, *et al.* (15). We initially adopted this inventory, but the model simulation showed obvious underestimation (pink bars vs. blue triangles, Fig. S1), and further examination showed the emission in Liu, *et al.* (15), underestimated the arsenic during coal combustion, i.e., power plants from (16), and households (17), as well as missed the smelting of non-ferrous metals (i.e., Cu, Pb, Zn, Sn, Sb and W). Therefore, this study

includes non-ferrous metals smelting (8, 18) and coal combustion (19, 20) emissions, the model results with the improved emission inventory showed much more reasonable performance (red bars vs. blue triangles, SI Appendix, Fig. S1), with MFB reduced from -95% to -22%. More details of the model evaluation can be seen in section 2.1, and the comparison of arsenic emission from coal combustion between this study and Liu, *et al.* (15) was shown in Fig. S2 in the SI Appendix.

The spatial resolution of GEOS-Chem in this study is 4° (latitude) by 5° (longitude). For the emission sources, there are in general three major data types. Firstly, for regions with arsenic emission directly available such as U.S., Europe, Japan, Australia and Canada, most of the emission sources are considered as point sources with specific latitude and longitude information provided, which can then be allocated directly to the model grid. Please note that for U.S., the arsenic emissions also come from nonpoint sources by county with Federal Information Processing Standards (FIPS) code provided, which are then allocated to the corresponding model grid considering the size of county is normally smaller than the spatial resolution of this study. Secondly, over China, the arsenic emission was estimated based on the coal combustion, nonferrous metal smelting, etc. at the provincial level, with size in general close to the spatial resolution of this study, therefore, it is reasonable to interpolate the provincial level emission to the model grid. Lastly, for all

the other regions with arsenic emissions estimated based on SO_2 from EDGAR, they are allocated to corresponding model grid due to much finer grid of 0.1° by 0.1° in EDGAR emission.

2. Discussion of sectoral, regional and global arsenic emission inventory in 2005 and 2015

For the global arsenic emission inventory, the contribution composition by different source categories and continents, as well as the decadal changes of source contributions in the top 10 emission countries have been shown in Fig. S4 and Fig. S5, respectively.

Based on the global arsenic emission in 2005 and 2015 (Fig. S3), more than half of the atmospheric arsenic emissions come from Power generation (PG), indicating that the combustion of fuel for energy plays a leading role in the global atmospheric arsenic emissions. In addition, industrial (IN) accounts for more than 40% of total emissions. From the perspective of decadal changes globally from 2005 to 2015, except for the slight increase in agricultural (AG) emissions from 117 tons to 177 tons, the emissions from all the other sectors decrease, resulting in an overall global arsenic emission reduction of 2% (435 tons). In view of different continents (Fig. S4), the emissions of Asia, Europe, North America and Oceania decreased by 6%, 6%, 22% and 29% respectively from 2005 to 2015, while the emissions over South America and Africa increased to some extent, with increase rate of 3% and 24%, respectively.

The top 10 emission countries in 2005 and 2015 were used to further explore the changes in arsenic emissions (Fig. S5), and the same

countries rank the top six in both of these two years, including China, Chile, India, Russia, Swaziland and Iran. Among all the countries, the total arsenic emissions are highest in China in both 2005 and 2015, accounting for 22% and 17% of global arsenic emissions in these two years, respectively, followed by Chile (14% and 16%) and India (7% and 12%). It is also clear that among these top 3 emission countries, the emissions in China decrease from 2005 to 2015, whereas increases of emission are obtained in the other two countries (Chile and India). The arsenic emission decrease in China is mainly due to sectors of PG and MI, reflecting an effect of energy conservation and emission reduction in China, i.e., the reduction of primary particulates matter emissions from coal-fired power plants and boilers (16). The arsenic emission increase in India mainly comes from the increase in the sector of PG resulting from the increase in fuel consumptions (21), whereas the slight increase in Chile was mainly attributed to the increase of copper production (Fig. S5).

The global arsenic emissions in 2005 and 2015 are estimated to be 18947 tons and 18512 tons (Fig. S3), respectively, which is somewhat higher than a recent study by Zhu, *et al.* (22) showing global arsenic emission of 8459.5 tons in 2012. If the uncertainty of 95th confidence interval is considered (Table S15 of Zhu, *et al.* (22)), the arsenic emission of 2015 in this study is relatively comparable to their upper bound (12588

tons), albeit of the existence of overestimation at 47% which may deserve more investigation in future studies. While uncertainties in estimating the arsenic emissions exist in both studies, the magnitude discrepancy may be attributed to the differences of method used to derive the arsenic emission (i.e. the section of Materials and Methods in this study vs. section 2.3 of Zhu, *et al.* (22)). In contrast, these two studies show consistency in the major sources, i.e., both studies indicate the same top 3 arsenic emission countries are China, Chile and India (Fig. 6 in Zhu, *et al.* (22) and Fig. S5 in this study), and the dominant arsenic emission source in China and India is coal combustion, whereas in Chile, almost all arsenic emission is from the nonferrous metal smelting.

3. Equation of Mean Fractional Bias (MFB) and Mean Fractional Error (MFE)

$$\mathrm{MFB} = \frac{2\sum_{i=1}^{n}\left(\frac{M_i - O_i}{M_i + O_i}\right)}{n} \times 100\%$$

$$\mathrm{MFE} = \frac{2\sum_{i=1}^{n}\left|\frac{M_i - O_i}{M_i + O_i}\right|}{n} \times 100\%$$

Where:

Oi is the observation data of each site;

M_i is the simulation value from model in each grid corresponding to the observation;

n is the number of observation sites.

4. The distribution of atmospheric arsenic deposition and its variation in 2005 and 2015

Atmospheric arsenic deposition plays a significant role on the ecosystem (23). The distribution of global arsenic deposition was shown in Fig. S10, with global total value of 21.70 Gg yr^{-1} and 21.02 Gg yr^{-1} in 2005 and 2015, respectively. The regions with high deposition mainly occur in Asia and South America, consistent with the arsenic concentration distribution discussed in section 2.2 in the main manuscript. The total deposition over Asia and South America accounts for 53% (50%) and 13% (15%) for the year of 2005 (2015), respectively. In terms of deposition per unit area, the maximal deposition flux in 2005 (1.40 mg m^{-2} yr^{-1}) and 2015 (1.17 mg m^{-2} yr^{-1}) was located in China and the junction of India, Bangladesh and Nepal, respectively.

5. Non-carcinogenic effect via global atmospheric inhalation

For the non-carcinogenic effect, we mainly use the regional hazard coefficient (HQ) to determine whether the non-carcinogenic effects of atmospheric arsenic in the region exceed the criteria. When the value of HQ is greater than 1, the cumulative non-carcinogenic effect caused by atmospheric arsenic inhalation cannot be ignored. The global HQ value distribution is shown in Fig. S13. The region with a global HQ of more than 1 appeared in southeastern China and northern Chile in 2005, with a maximum HQ of 1.28 in Chile. In 2015, there were no non-carcinogenic adverse effects in China, while there were still areas in Chile with HQ value higher than the acceptable level and the maximum value of HQ is 1.29, so the non-carcinogenic effects of arsenic still need to be addressed.

6. Uncertainty analysis

A Monte Carlo simulation(16, 24) was used to access uncertainty based on the uncertainties of atmospheric arsenic emission inventory and input parameters of health risk assessment model. For the uncertainty study of atmospheric arsenic emissions, normal distribution is assumed for the uncertainties by the coefficient of variation of daily atmospheric arsenic concentration by the GEOS-Chem simulation result. For the uncertainty study of the input parameters of the health risk assessment model, the input parameter was treated as a random variable, which is based on USEPA (25). The Monte Carlo simulation was used in 2005 and 2015 and each scenario was run repeatedly 50000 times. The probability distribution of carcinogenic effects of atmospheric arsenic inhalation was obtained by Monte Carlo simulation results, and the 95% confidence interval was obtained.

7. Differences of exposure with atmospheric and groundwater arsenic pollution between urban and rural areas

Based on the studies about groundwater arsenic carcinogenic risks (Table S3), we selected the studies specifically pointed out the characteristics of the observation site, i.e., urban or rural area, and compared the arsenic risks based on observation, model results as well as their ratio (Please see the Table below). The arsenic carcinogenic risks in rural areas are not necessarily lower, instead of even higher, than that over urban areas. For instance, the rural areas in Chapai-Nawabganj, Bangladesh (26) and Cambodia (27) may reach as high as $300*10^{-5}$, $183*10^{-5}$, more than 1000 times of the standard ($1*10^{-6}$(28)). For urban areas (26, 29-33)，the carcinogenic risks range from $1*10^{-5}$ to $62*10^{-5}$, even if 90% or more arsenic removal is achieved in groundwater considering of the water treatment (34, 35), most of them still exceed $1*10^{-6}$. For Punjab, Pakistan (36), the average carcinogenic risk over the urban and rural areas is very high ($168*10^{-5}$). Among the seven urban sites, the carcinogenic risks between atmospheric arsenic and groundwater arsenic become comparable if 90% arsenic is removed during groundwater treatment, with three sites showing even higher risk from atmospheric arsenic (Chandigarh, India; Mashhad, Iran; Subarnarekha River Basin, India), implicative of the importance of taking into account of the atmospheric arsenic pollution.

Figures and Tables

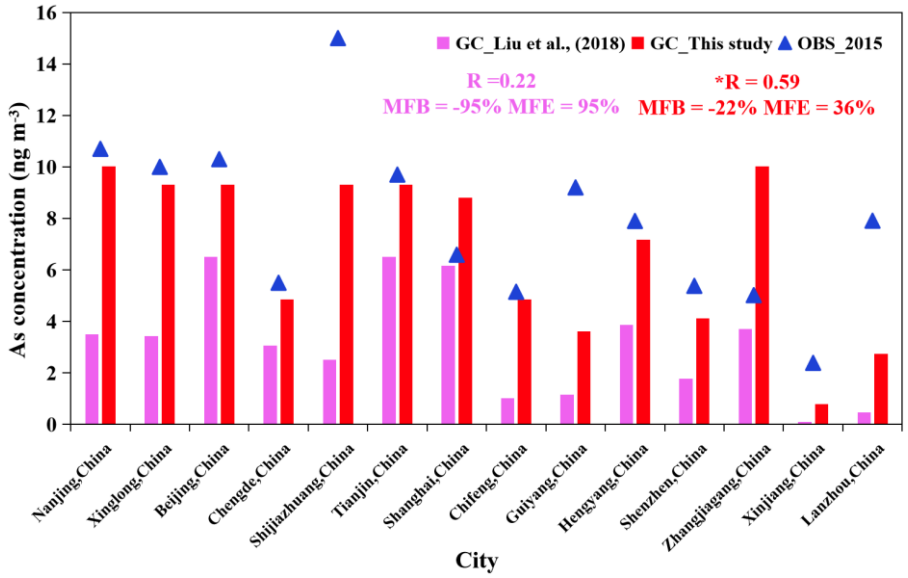

Fig. S1. The comparison of arsenic concentration between observation (blue triangles) and model results (bars), with pink and red bars indicating model simulations based on the arsenic emission from Liu, *et al.* (15) and this study, respectively.

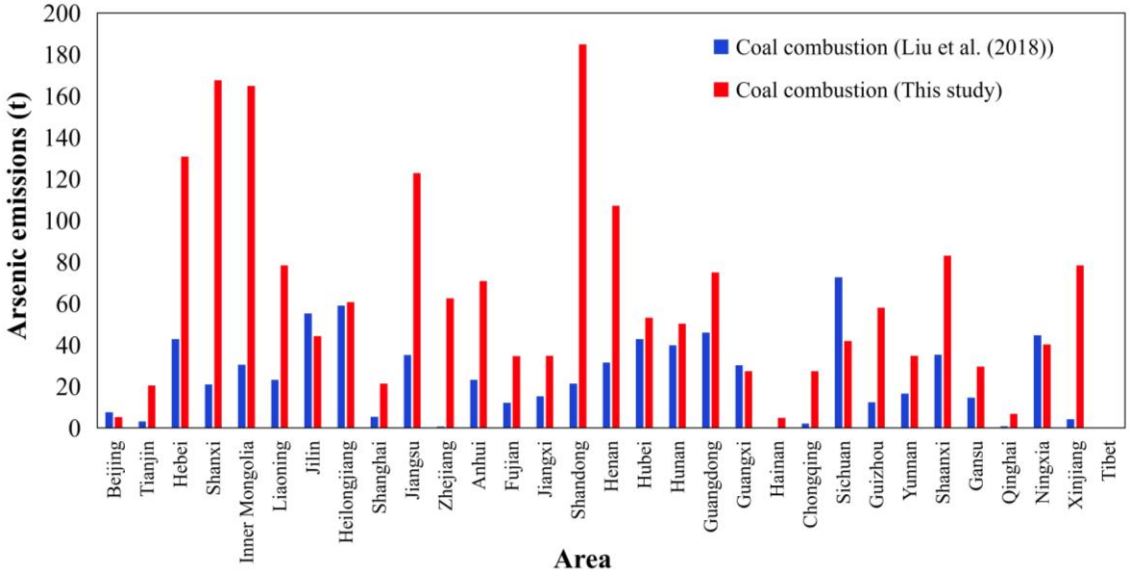

Fig. S2. Annual arsenic emissions in 2015 based on the coal combustion in Liu, *et al.* (15) (blue bars) and this study (red bars).

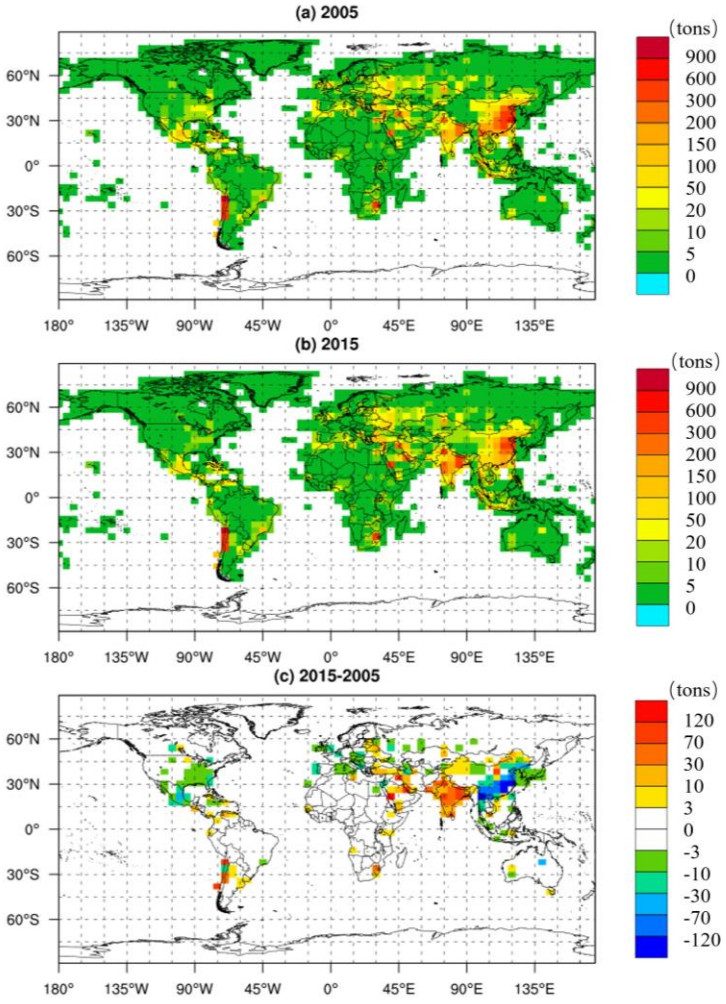

Fig. S3 The spatial distribution of arsenic emission in 2005, 2015 as well as their differences

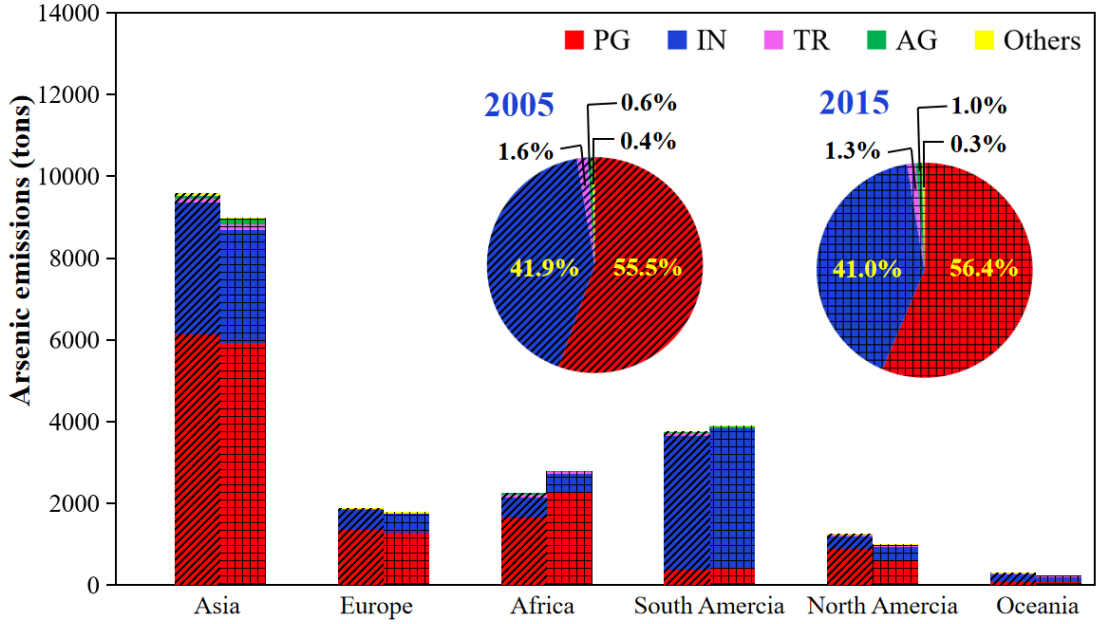

Fig. S4 The contribution composition of arsenic emission by different source categories and continents in 2005 and 2015. For each continent, the left and right bar indicates the emission in the year of 2005 and 2015, respectively. The pie charts show the fractional contribution to global arsenic emission from different sectors in 2005 and 2015. The sector definitions are based upon the information of Emission Database for Global Atmospheric Research (EDGAR: https://edgar.jrc.ec.europa.eu/overview.php?v=431), i.e., Power generation (PG): Power industry and residential combustion, Industry (IN): energy industry, manufacturing industry and process emission during production and application, Transport (TR): road and off-road transportation, Agricultural (AG): agriculture and waste burning, Others: solid waste, fossil fuel fires, soil emission.

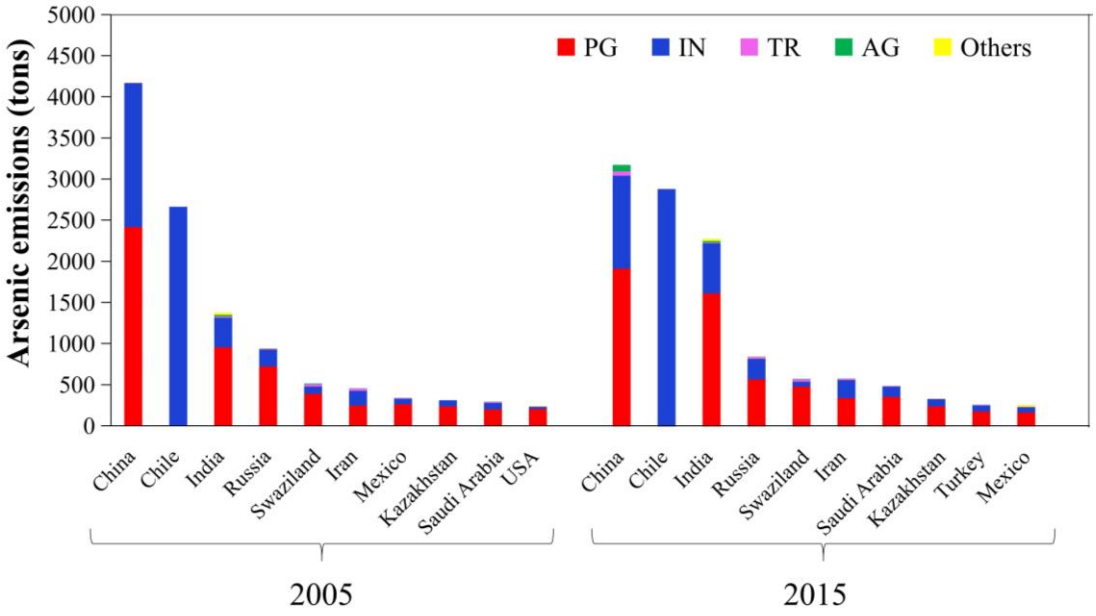

Fig. S5 The emission contribution by different sectors in the countries with top 10 amount of arsenic emission in 2005 and 2015. The sector definitions are the same as Fig. S4

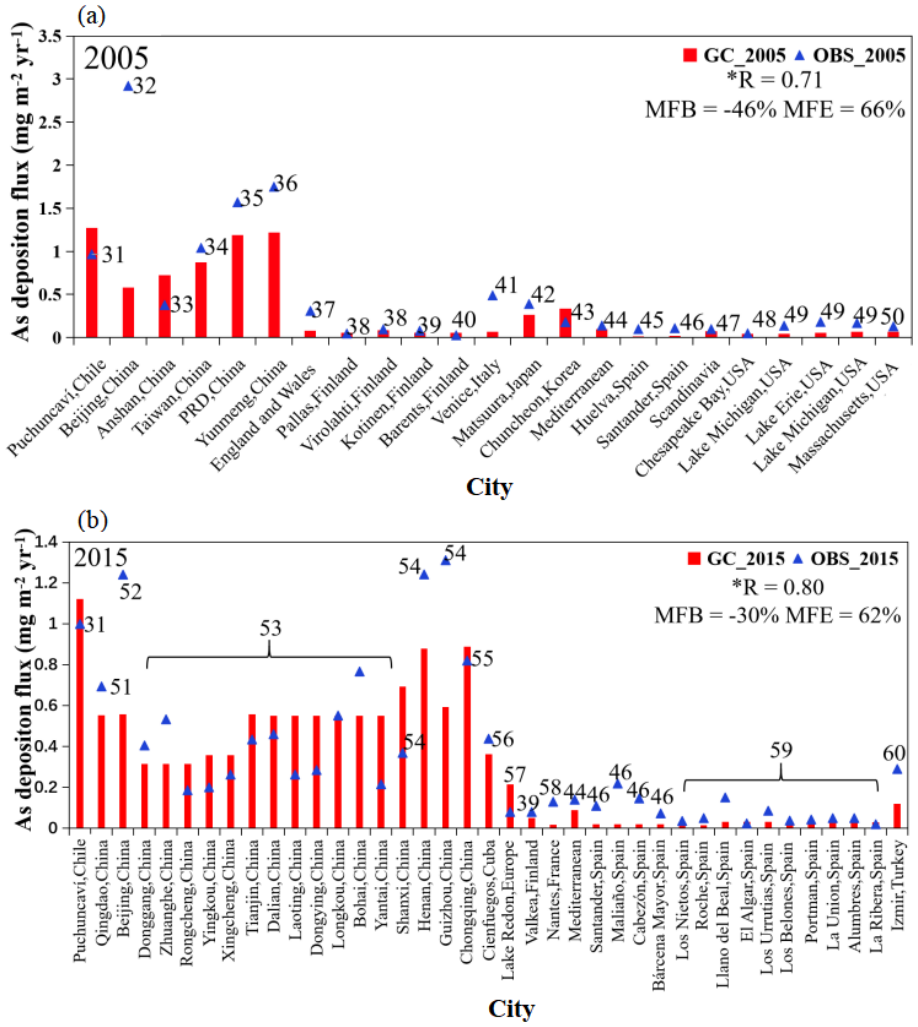

Fig. S6. Evaluation of atmospheric arsenic deposition in 2005 (a) and 2015 (b).

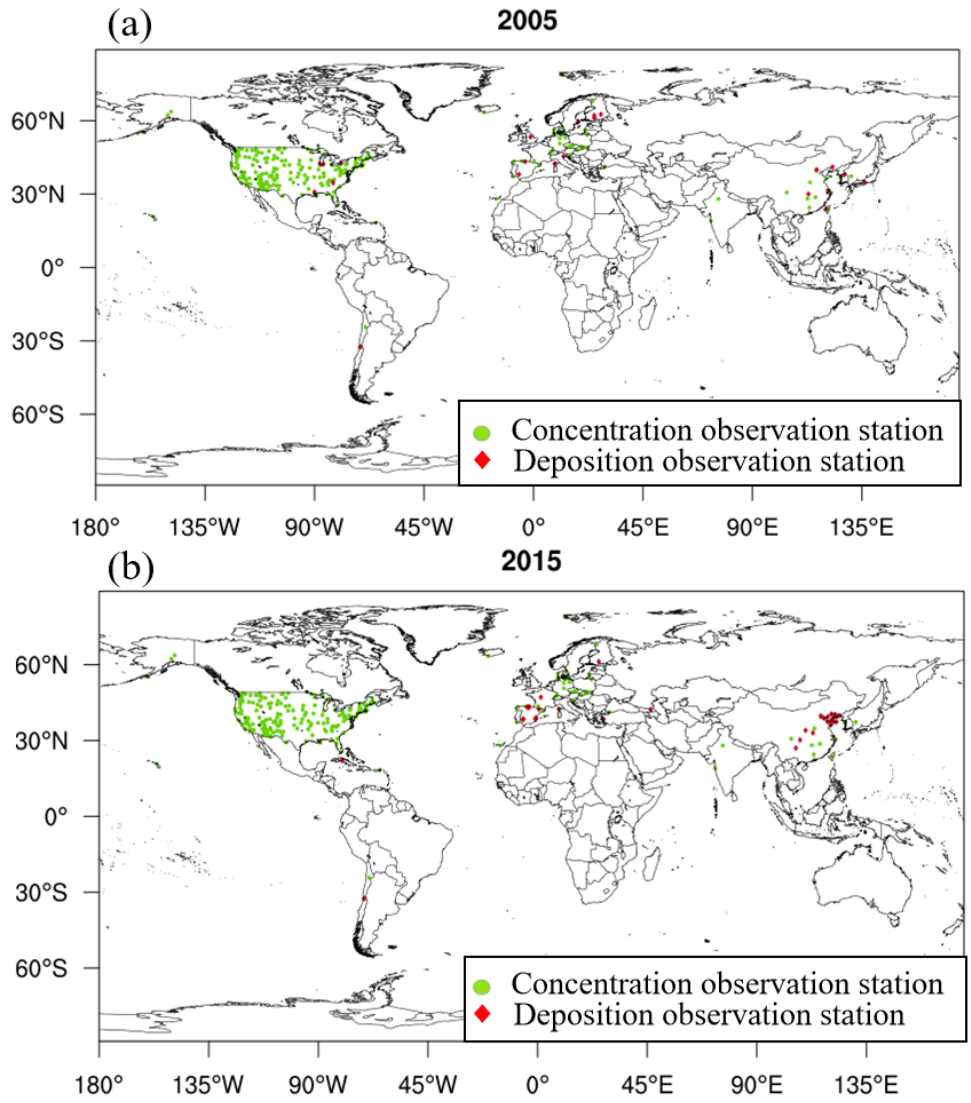

Fig. S7. The distributions of observed atmospheric arsenic concentration (green circles) and deposition (red diamond) from EMEP (http://ebas.nilu.no/default.aspx) for Europe, IMPROVE (http://views.cira.colostate.edu) for US, as well as from previous literature (Table S1-2) in 2005 (a) and 2015 (b).

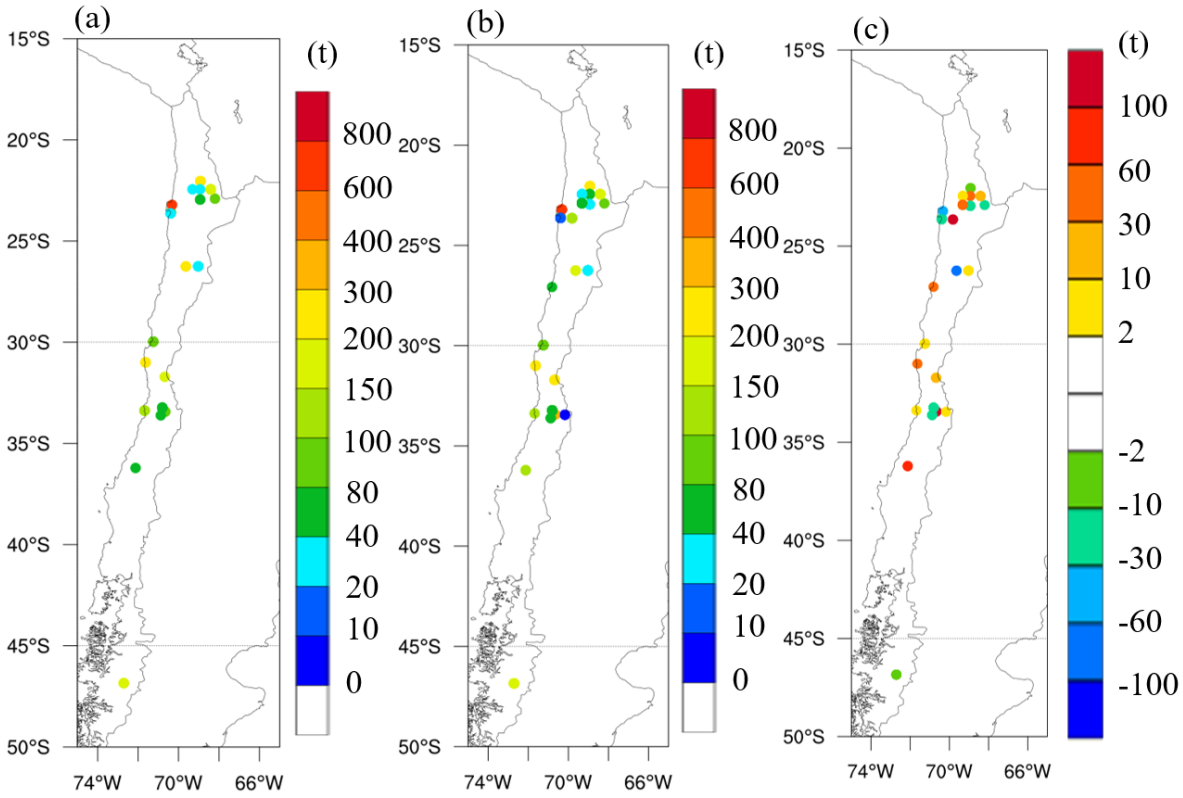

Fig. S8. Spatial distribution of annual total arsenic emission from copper smelters over Chile in 2005 (a), 2015 (b) as well as their changes (2015 minus 2005; c).

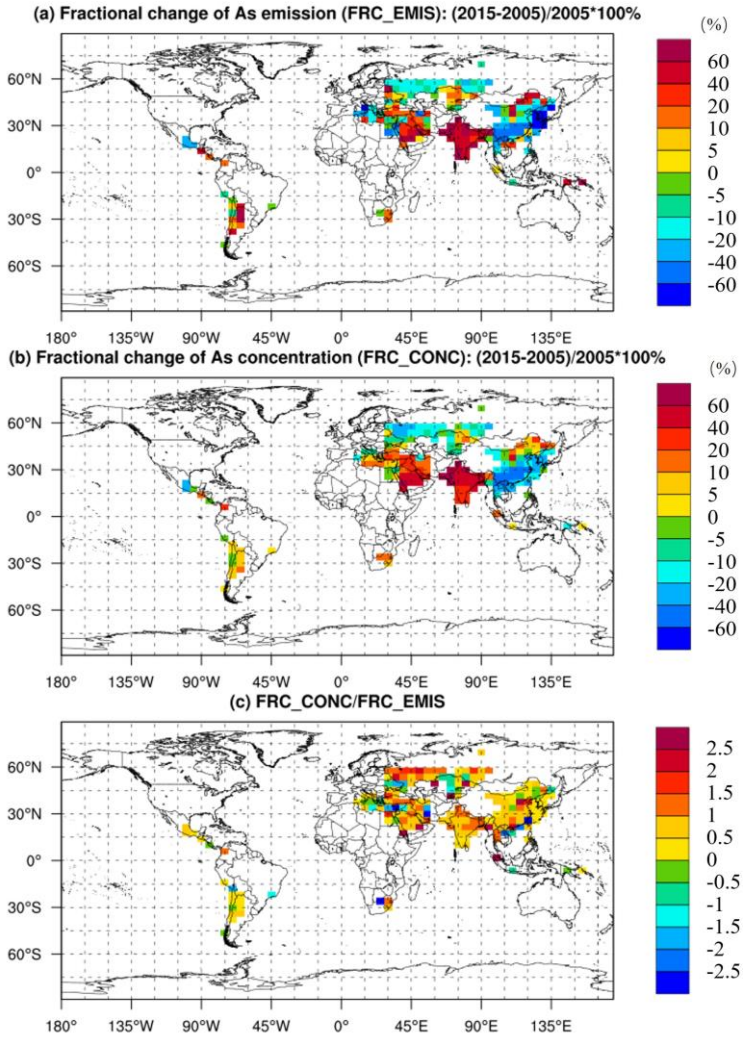

Fig. S9. Spatial distribution of fractional changes in concentration and emission, as well as their ratio. Only grids with arsenic concentration in 2005 greater than 1 ng/m^3 were shown.

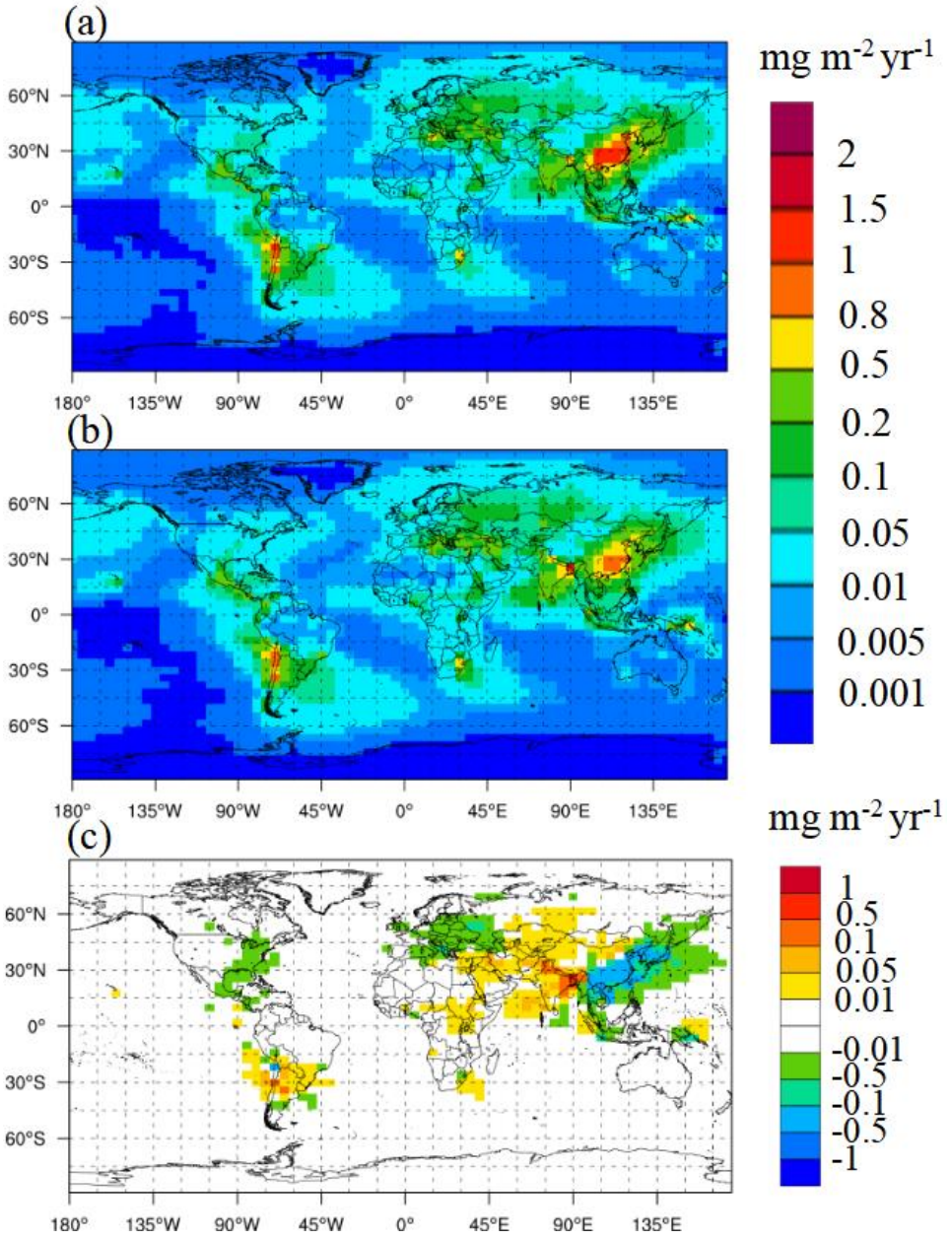

Fig. S10. Spatial distribution of atmospheric arsenic deposition flux in 2005 (a), 2015 (b) and their changes (2015 minus 2005; c).

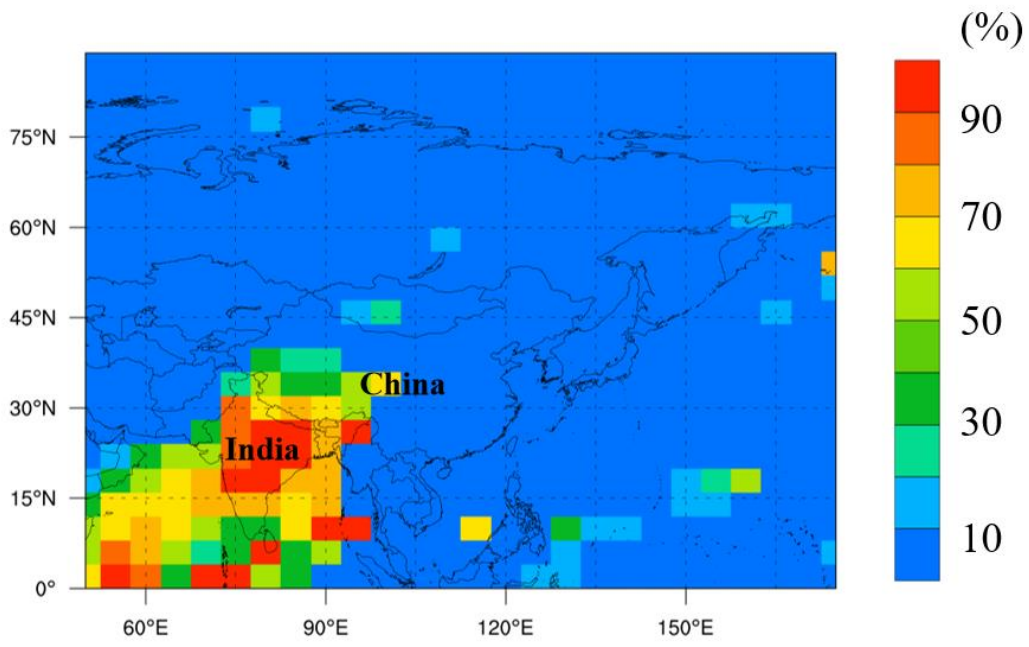

Fig. S11. The percentage contribution of arsenic concentration from the arsenic emission in India, based upon the differences in two scenarios with and without arsenic emission in India

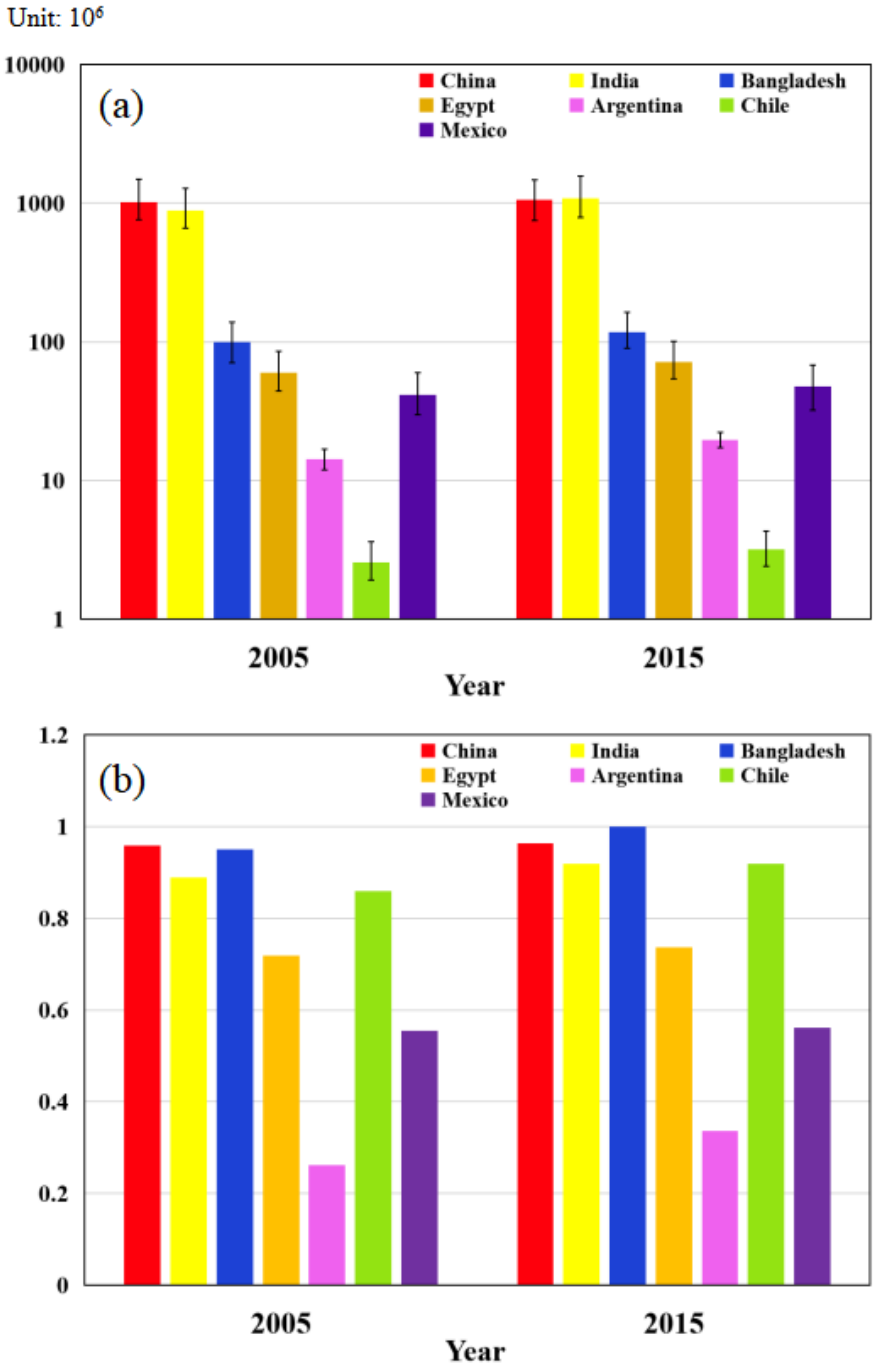

Fig. S12 The number of people affected by arsenic (a) and normalization by the total population in the respective country (b)

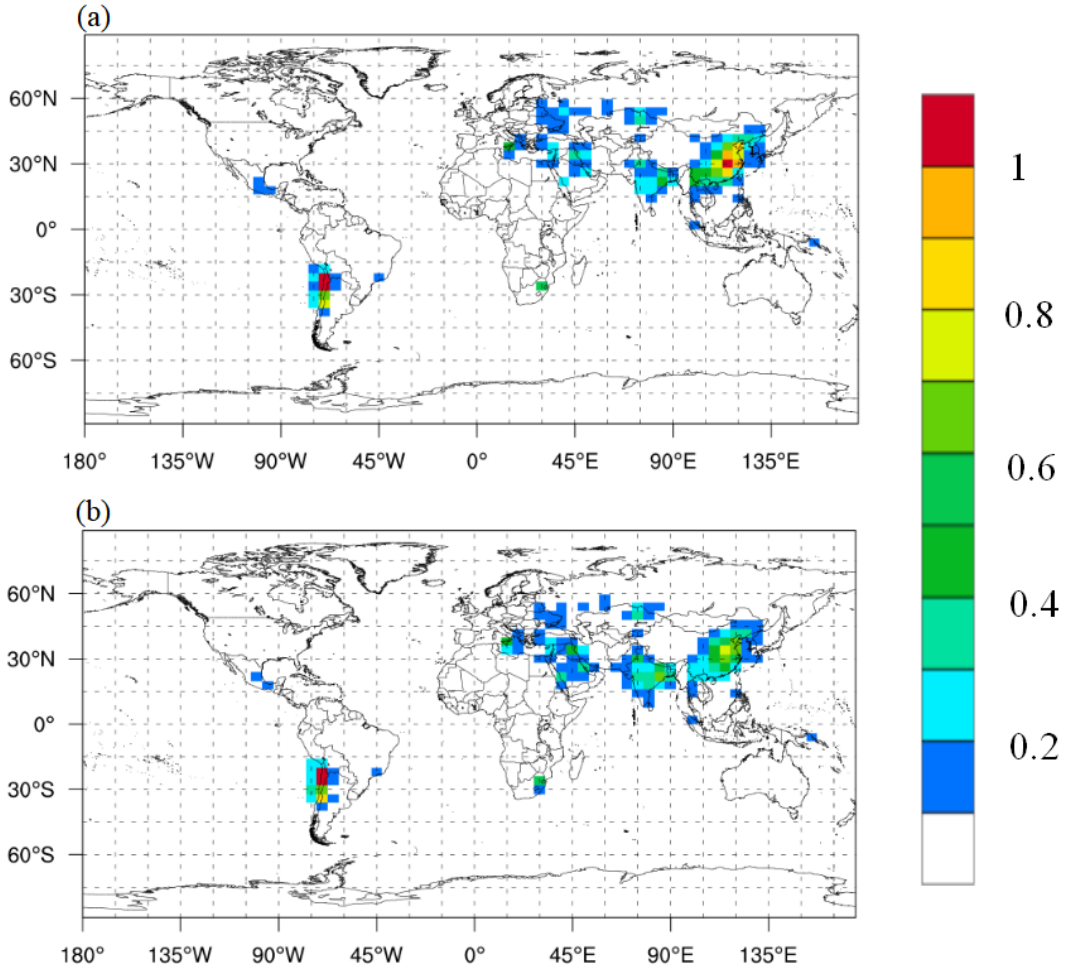

Fig. S13 The spatial distribution of non-carcinogenic hazard coefficient (HQ) value in 2005 (a) and 2015 (b).

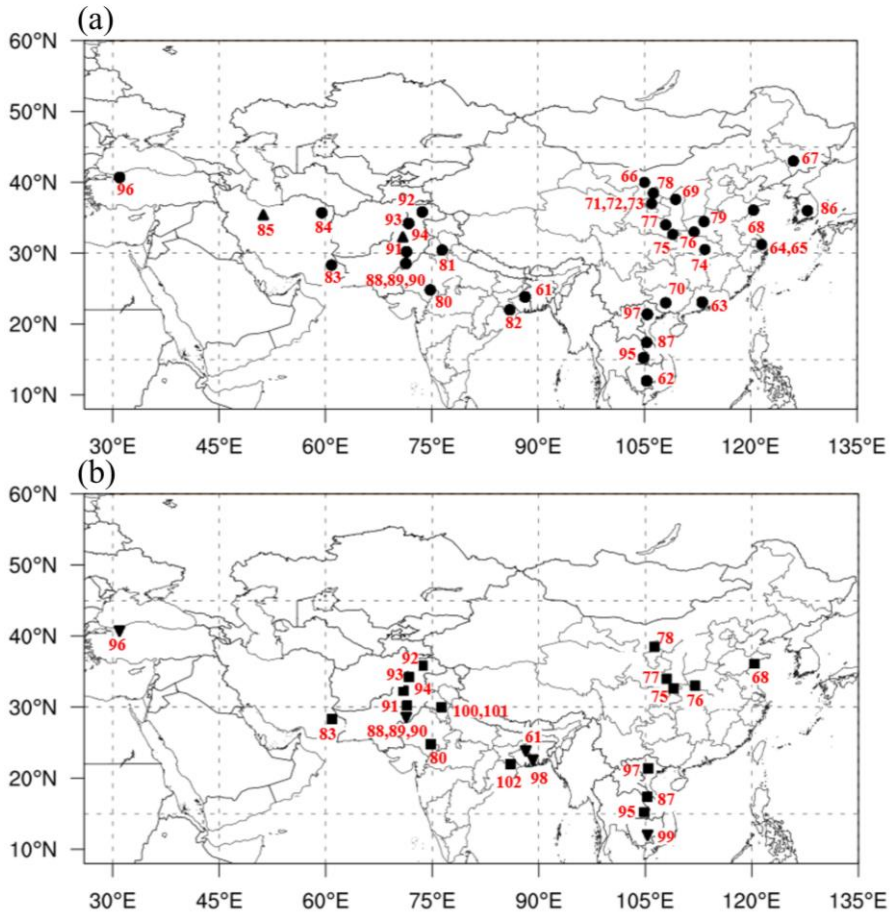

Fig. S14. References number for the observations over the sites with carcinogenic risk (a) and non-carcinogenic health risk (b). All references (61-102) indicated in the figure are available in Table S3.

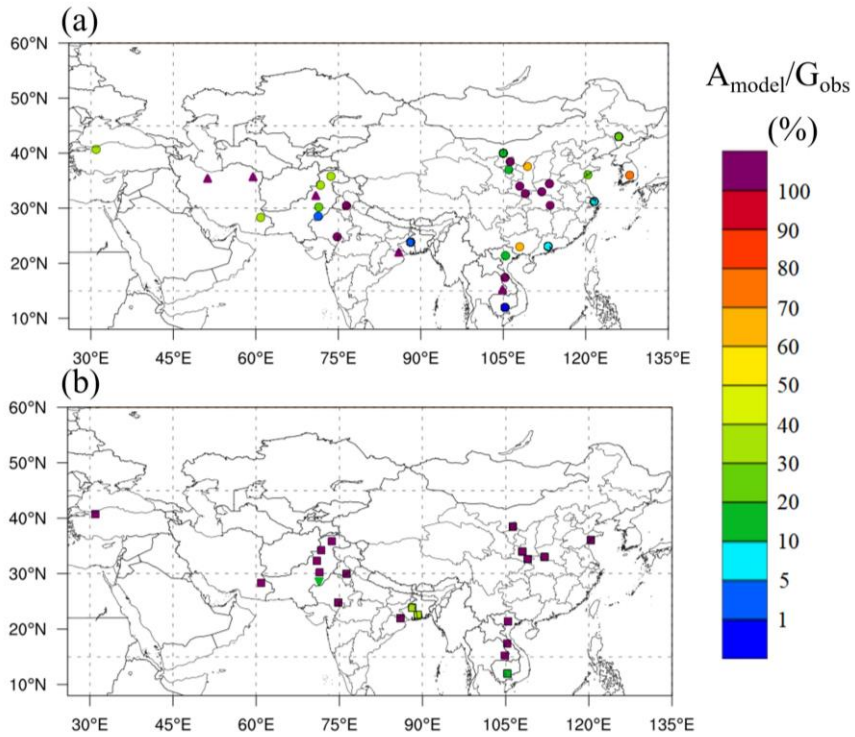

Figure S15. The ratio of carcinogenic (a) and non-carcinogenic (b) effect of arsenic in the atmosphere to that in groundwater (90% removal rate of arsenic). In Fig. 5a, the upper triangles (△) and circles (○) represent the CR value exceeding the threshold in atmosphere only and both atmosphere and water, respectively; Fig. 5b, the lower triangles (▽) represent the HQ value exceeding the criteria in groundwater whereas for the locations without HQ exceedance marked with hollow squares (□). All information about observational sites was acquired from literature described in SI Appendix, Table S1 and Figure. S8.

Table S1. References of observed atmospheric arsenic concentration and deposition, carcinogenic and non-carcinogenic risk due to groundwater arsenic.

NO.	Study period	Urban(U)/Rural(R)	Aerosol sample type	Study area	Ref
1	10.1999-10.2000	R	PM_{10}	Quillota, Chile	(37)
2	05.2000-10.2000	R	PM_{10}	Toconao, Chile	(2)
3	01.2004-12.2006	U	$PM_{2.5}$	China	(38)
4	06.2009-09.2009	U	$PM_{2.5}$	Changsha, China	(39)
5	04.2009-05.2019	U	$PM_{2.5}$	Chengdu, China	(40)
6	07.2007-08.2007	U	$PM_{2.5}$	Nanchang, China	(41)
7	01.2010-01.2011	U	$PM_{2.5}$	Zhengzhou, China	(42)
8	09.2009-08.2010	R,	$PM_{2.5}$	Xinglong, China	(43)
9	03.2004-04.2005	/	TSP	Shaoguan, China	(44)
10	2008	U and R	PM_{10}	Tiwan, China	(45)
11	06.1994-06.1995	R	$PM_{2.5}$	Palmelal, Potugal	(46)
12	01.2001-03.2002	R and U	$PM_{2.5}$	Spain	(47)
13	07.200808.2010	U	TSP	Rasathane, Turkey	(48)
14	10.2003-10.2008	/	$PM_{2.5}$	Ulleung Island, Korea	(49)
15	05.2004-01.2006	U	PM_{10}	Seoul, Korea	(50)
16	01.2010-12.2011	R,	$PM_{2.5}$	Trombay, India	(51)
17	01.2008-02.2008	U	TSP	India	(52)
18	01.1998-03.1999	R	$PM_{2.5}$	Chaumont, Switzerland	(53)
19	04.2013-05.2013	U	$PM_{2.5}$	Nanjing, China	(54)
20	06.2014-04.2015	U and R	$PM_{2.5}$	China	(55)
21	12.2015-06.2016	U	$PM_{2.5}$	Shanghai, China	(56)
22	01.2016-01.2017	U and R	$PM_{2.5}$	Chifeng, China	(57)
23	07.2015-08.2016	U and R	$PM_{2.5}$	Guiyang, China	(58)

24	12.2015-09.2016	U and R	$PM_{2.5}$	Hengyang, China	(59)
25	01.2014-12.2015	U	$PM_{2.5}$	Shenzhen, China	(60)
26	01.2017-12.2017	U	$PM_{2.5}$	Zhangjiagang, China	(61)
27	09.2015-08.2016	U	$PM_{2.5}$	Xinjiang, China	(62)
28	01.2015-12.2015	U	$PM_{2.5}$	Lanzhou, China	(63)
29	10.2014-11.2014	U	$PM_{2.5}$	Argentina and Japan	(64)
30	03.2010-04.2010	R	$PM_{2.5}$	Fukue Island, Japan	(65)

Note: The “/” represents the information is not available.

Table S2. References of observed atmospheric arsenic deposition flux.

No.	Study period	Urban(U)/Rural(R)	Study area	Ref
31	05.2010-08.2010	U	Puchuncaví, Chile	(66)
32	11.2005-11.2006	R	Beijing, China	(67)
33	06.2008-07.2008	U and R	Anshan, China	(68)
34	01.2009-12.2010	U and R	Tiwan, China	(69)
35	11.2006-11.2007	U	PRD, China	(70)
36	03.2009-03.2010	R	Yunmeng, China	(71)
37	1995-1998	/	England and Wales	(72)
38	01.2005	R	Finland	(73)
39	1988-2011	R	Kotinen, Finland	(74)
40	03.1994-04.1995	/	Barents, Finland	(75)
41	06.1998-06.1999	U and R	Venice, Italy	(76)
42	04.2004-03.2006	R	Matsuura, Japan	(77)
43	08.2006-06.2009	R	Chuncheon, Korea	(32)
44	03.2008-10.2011	R	Cap Cuittone, Mediterranean	(78)
45	06.2008-05.2011	/	Huelva, Spain	(79)
46	01.2009-02.2013	/	Spain	(80)
47	01.2002-12.2005	R	Scandinavia	(81)
48	06.1990-07.1991	/	Chesapeake Bay, USA	(82)
49	1993-1994	/	USA	(83)
50	09.1992-09.1993	U	Massachusetts Bay, USA	(84)
51	07.2016-07.2016	R	Qingdao, China	(85)
52	05.2014-04.2015	U	Beijing, China	(86)
53	summer 2014 - winter 2015	/	China	(87)
54	2015	R	China	(88)
55	01.2014-06.2014	R	Chongqing, China	(89)

56	03.2014-11.2016	U	Cienfuegos, Cube	(90)
57	04.2004-11.2013	/	Lake Redon, England	(91)
58	02.2014-12.2014	/	Nantes, France	(92)
59	06.2011-03.2013	R	Spain	(93)
60	11.2014-10.2015	R and U	Izmir, Turkey	(94)

Note: The “/” represents the information is not available.

Table S3. References of observed carcinogenic and non-carcinogenic risk due to groundwater arsenic.

NO.	Study period	Urban(U)/Rural(R)	Study area	Ref
61	11.2014-12.2014	R	Chapai-Nawabganj, Bangladesh	(26)
62	02.2009-08.2009	R	Cambodia	(27)
63	04.2011-12.2011	U	Guangdong, China	(29)
64	07.2006	/	Shanghai, China	(95)
65	01.2012-12.2012	U	Shanghai, China	(96)
66	/	/	Inner Mongolia, China	(97)
67	01.2006, 07.2007 and 09.2010	/	Jilin, China	(98)
68	01.2011 and 06.2011	/	Qingdao, China	(99)
69	09.2011	/	Yulin, China	(100)
70	2010	R	Guangxi, China	(101)
71	01.2013-12.2013	/	Ningxia, China	(102)
72	2015	/	Ningxia, China	(103)
73	07.2012	/	Ningxia, China	(104)
74	09.2009-12.2009	R	Wuhan, China	(105)
75	2005-2006	/	Han River, China	(106)
76	/	/	Huai River, China	(107)
77	2005	/	Xian, China	(108)
78	07.2013-09.2013	/	Weining Plain, China	(109)
79	12.2014	/	Zhengzhou, China	(110)
80	/	/	Rajasthan, India	(111)
81	12.2014	U	Chandigarh, India	(112)
82	05.2012	R and U	Subarnarekha River, India	(113)
83	/	U	Sistan and Baluchistan, Iran	(30)

84	03.2017-04.2017 and 08.2017-09.2017	U	Mashhad, Iran	(31)
85	2017	R	Dehgolan, Iran	(114)
86	1992-1995	U	Korea	(115)
87	05.2008-07.2018 and 02.2010-03.2010	/	Laos	(116)
88	/	U and R	Punjab, Pakistan	(36)
89	/	U	Punjab, Pakistan	(117)
90	/	R	Punjab, Pakistan	(118)
91	2017	U	Multan, Pakistan	(33)
92	/	/	Kohistan, Pakistan	(119)
93	/	/	Peshawar, Pakistan	(120)
94	2012	/	khyber Pakhtunkhwa, Pakistan	(121)
95	06.20102011.01	R	Ubon Ratchathani, Thailand	(122)
96	09.2010-08.2011	/	Melen watershed, Turkey	(123)
97	/	/	Thai Nguyen, Vietnam	(124)
98	/	/	Khulna division, Bangladesh	(125)
99	02.2009 and 08.2009	/	Kandal, Cambodia	(126)
100	2013	R	Ropar wetland, India	(127)
101	2013	/	Ropar wetland, India	(128)
102	11.2011, 05.2012 and 08.2012	/	Subarnarekha River, India	(129)

Note: The “/” represents the information is not available.

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