Supplement of

Global anthropogenic emissions of particulate matter including black carbon

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S1 Comparison of temporal distribution patterns

Fig S1.1 shows a comparison of the temporal patterns (it is an aggregate as the actual patterns are grid specific) for residential combustion sector, applied in the ECLIPSE project, with other data for selected countries.



Figure S1.1. Comparison of monthly distribution of emissions used in ECLIPSE with profiles from EDGAR (EC-JRC/PBL, 2010), EMEP (http://emep.int/mscw/), national Finish model FRES (Karvosenoja, 2008), and US EPA.

S2 Particulate matter emission factors for residential combustion

The GAINS model distinguishes three principal solid fuel stove categories: *traditional, improved* and *new stoves*. *Traditional heating stoves* using wood or coal as fuel have simple grate based firebox designs with usually only primary air supply and no heat storing components. Consequently there is restricted availability of air for combustion and poor mixing

- 5 of air and pyrolysis gases. *Traditional stoves* in general have very high PM emission factors compared with more advanced technologies, but within this category the variability in the emission factors is also large. For example highest emission factors for traditional wood stoves have been measured in situations with restricted combustion air supply that leads to lower burn rate (Jordan and Seen, 2005). Such conditions might prevail when the user wants a lower heat supply to the room. *Improved stoves* have secondary air supply and heat storing components in the firebox construction that improve the
- 10 combustion performance and reduce emissions of PM compared with the *traditional stoves*. *New stoves* represent the most advanced stove models on the market that have firebox, construction and airflow characteristics that optimize combustion efficiency. Additionally, an electrostatic precipitator (ESP) can be fitted into the latest stoves, which further improve the PM emission performance. GAINS distinguishes also *wood pellet stoves*. Pellets are a very homogenous fuel and combustion is more optimized than batch fired wood log stoves and thus also the PM emissions are lower than with wood log stoves.
- 15 A stove heats the surrounding room, but a boiler heats water to be circulated through a piping system to heat an entire house (Johansson et al., 2004). In *old-type wood log boilers* up-draught combustion is commonly used, which resembles the combustion in a stove; *modern wood boilers*, however, use downdraught combustion and often have an isolated burn-out zone (Johansson et al., 2004). In contrast to stoves, wood boilers can be connected to a water tank to store heat, which allows the boiler to be run at a regular heat output and to certain extent optimizing the combustion conditions. Storage tanks are
- 20 common in modern wood boilers and also old boilers may be equipped with them, leading to lower emissions and higher efficiencies (Johansson et al., 2004). The single family house boilers are typically smaller than 50 kW_{th}, the larger residential boilers are allocated to a category *medium size boilers* where manual and automatic boilers are distinguished (Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007). Such boilers might be an important emission source, especially when many of them are fired with coal, but there are not a of lot measurements available. The GAINS model relies on studies discussed
- 25 previously (EEA, 2013; Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007) but for a number of countries in Europe updates were made drawing on national information provided within EU consultations (Amann et al., 2015) and recent measurements in China where 100,000s of such installations are used in both residential as well as industrial sector (Wang et al., 2009).

GAINS distinguishes also open fireplaces as a separate category which is of relevance mostly in North America and some

30 European countries, even though in Europe less than 5% of fuelwood would be used in such installations (Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007).

Here we summarize the published measurements of emission factors for cooking and heating stoves boilers and compare them to the current ranges of region- and technology-specific GAINS values. The focus is on studies that appeared after the original development of the GAINS particulate matter module (Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007).

Emission factors (mg/MJ) S		Share	es (%)		References		
PM	TC ^a	BC	OC	TC^{a}	BC	OC	
wood log							
1300 (350-2200) ^b 120	715			55			(Boman et al., 2008) old, no accumulator, large fuel charge (Boman et al., 2008)
(73-260) ^b 95	60			50			old, no accumulator, adjusted fuel charge (Boman et al., 2008)
(87-100) ^b 44	48			50			old, with accumulator (Boman et al., 2008)
$(11-450)^{b}$	18			42			modern, with accumulator
37	27	12	16	75	32	43	(Gaegauf et al., 2005), 35 kW apartment house
70-700		20	30-335				GAINS ^c , >50 kW, uncontrolled boiler
230-1300		75-200	75-600				GAINS ^c , <50 kW, old uncontrolled boiler
80-520		32-50	22-230				GAINS ^c , <50 kW, improved
40-260		13-37	12-100				GAINS ^c , <50 kW, new/modern
wood chip							
				44	23	21	(Schmidl et al., 2011) 40 kW moving grate, start-up
				5	1	4	(Schmidl et al., 2011) 40 kW moving grate, full load
				35	33	2	(Schmidl et al., 2011) 40 kW moving grate, part load
85	8	2	6	9	2	7	(Gaegauf et al., 2005) 70 kW, institute building
wood pellet							
20		0.1	0.9		0.5	5	(Lamberg et al., 2011a) efficient combustion
12 (3-29) ^b		0.8 (0-14) ^b	0.3 (0-3) ^b		6 (0-51) ^b	2 (2-11) ^b	(Lamberg et al., 2011b), 25 kW, nominal load
16		1	0.1				(Tissari et al., 2008), 20 kW, nominal load
24		3	0.2				(Tissari et al., 2008), 20 kW, partial load
49	35	24	11	72	49	23	(Gaegauf et al., 2005) 10-32 kW, apartment house
8-25		0.8-1	0.4-1				GAINS ^c , >50 kW
20-68		5	2.5-10				GAINS ^c , <50 kW

5 Table S2.1: Summary of PM emission factors for residential wood boilers.

^a Total Carbon (TC) ^b (min-max)

^c PM value refers to PM2.5

Emission factors (mg	g/MJ)		Shares (%)	Reference
PM	BC	OC	BC	OC	
traditional					
673-1373	24-72	263-623	2-7	39-53	(Alves et al., 2011)
300-1400	-	-	2-9	35-50	(Gonçalves et al., 2011) incl. cold start
90-900	-	-	2-9	35-48	(Gonçalves et al., 2011) incl. hot start
750-1060	-	-	-	-	(Jordan and Seen, 2005), full airflow
1560-1700	-	-	-	-	(Jordan and Seen, 2005), half airflow
1870-3000	-	-	-	-	(Jordan and Seen, 2005), closed airflow
128-400	20	157	8	64	(McDonald et al., 2000)
-	39-43	70-390	5-14	47-67	Studies in Kupiainen& Klimont (2007)
150 ^a - 930 (2400) ^b	32 ^a - 100	$60^{a} - 435 (1200)^{b}$	4-22	41-50	GAINS; the PM value represents PM2.5
improved					
22-180	-	-	-	-	(Boman et al., 2008)
86-105	9-11	52-58	-	-	(Fine et al., 2004)
130	88	39	68	30	(Gaegauf et al., 2005)
60-160	-	-	11-37	20-43	(Gonçalves et al., 2010)
75-97	15-28	17-35	24-32	27-39	(Schmidl et al., 2011)
38-350	-	-	-	-	(Pettersson et al., 2011)
-	56-79	11-16	-	-	Studies in Kupiainen& Klimont (2007)
55 ^a - 372	30 ^a - 95	11 ^a -133	25-55	19-35	GAINS; the PM value represents PM2.5
new					
67-122	13-15	43-67	-	-	(Fine et al., 2004), catalytic
72-89	21-33	16-32	30-37	22-36	(Schmidl et al., 2011)
30 ^a - 186	9 ^a - 30	8 ^a - 67	18-30	28-35	GAINS; the PM value represents PM2.5
pellet					
10-66	-	-	-	-	(Boman et al., 2008)
15-47	-	-	-	-	(Boman et al., 2011)
17	0.7	-	4	-	(Frey et al., 2014)
20	0.1	0.9	0.5	5	(Lamberg et al., 2011b)
3-29	0-14	0.1-3	0-51	2-11	(Lamberg et al., 2011a)
-	-	-	14	11	(Schmidl et al., 2011)
47-129	0.5-1.3	0.3-5.2	1-2	1-9	(Sippula et al., 2007)
10 ^a - 47	1.3 ^a - 4	2 ^a - 7	10-17	12-17	GAINS; the PM value represents PM2.5

Table S2.2: Summary of PM emission factors for residential heating wood stoves.

^a The lowest values represent Swiss data

^b Norwegian wood stove

Emission factors (mg/MJ)			References
PM	BC	OC	
traditional			
530	44	250	(Just et al., 2013)
106	50	44	(Roden et al., 2009), 3-stone, lab measurements
515 (300-1000) ^a	83 (10-210) ^a	254 (90-660) ^a	(Roden et al., 2009), Honduras, field measurements
510 (280-510) ^b	65-75 (40-75) ^b	229 (125-229) ^b	GAINS ^c
improved			
150	80	20	(Just et al., 2013), rocket stove
270 (100-500) ^a			(Li et al., 2009), improved stoves, PM2.5
394 (120-700) ^a	102 (6-325) ^a	208 (60-460) ^a	(Roden et al., 2009), improved no chimney, field measurements
205 (105-270) ^b	50-75 (27-75) ^b	63 (31-68) ^b	GAINS ^c
new			
255 (40-720) ^a	116 (6-660) ^a	93 (33-370) ^a	(Roden et al., 2009), improved with chimney, field measurements
56-102	11-21	19-34	GAINS ^c
fan assisted			
86 (25-125) ^a	33 (6-100) ^a	38 (4-71) ^a	(Roden et al., 2009), fan assisted, lab measurements
54	33	14	(Just et al., 2013), gasifier with fan
17	4	9	GAINS ^c

 Table S2.3: Summary of PM emission factors for cookstoves using biofuels.

^a (min-max)

^b central value for fuelwood and in brackets the whole range including also dung and agricultural residues

Emission factors (mg/MJ	I)		References
PM	BC	OC	
traditional			
805 (214-1360) ^a	250 (11-540) ^a	400 (116-710) ^a	(Zhi et al., 2009), portable stove, bituminous coals
	332 (10-610) ^a	472 (129-822) ^a	(Chen et al., 2009), simple low-efficiency stove without chimney, bituminous coals
351	135	108	GAINS ^b (cooking)
315-495	90-220	160-200	GAINS ^b (heating)
improved			
	466 (6-1377) ^a	248 (35-551) ^a	(Chen et al., 2009), high-efficiency stove with chimney
492	183	200	(Zhang et al., 2008), steel stove, brown coal
36	1	16	(Zhang et al., 2008), steel stove, bituminous coal
408 (155-685) ^a	40 (2-140) ^a	230 (78-470) ^a	(Zhi et al., 2009), bituminous coals
246	132	60	GAINS ^b (cooking)
315-350	82-200	88-112	GAINS ^b (heating)
new			
270	23	96	(Li et al., 2016), average for bituminous coals
176	108	32	GAINS ^b (cooking)
158-248	73-176	48-60	GAINS ^b (heating)
briquettes			
	16 (2-33) ^a	329 (71-668) ^a	(Chen et al., 2009), simple low-efficiency, no chimney
	4 (0.5-9) ^a	219 (27-423) ^a	(Chen et al., 2009), high-efficiency, with chimney
184	3	80	(Zhang et al., 2008), steel stove
440 (98-930) ^a	12 (2-23) ^a	233 (67-460) ^a	(Zhi et al., 2009), traditional portable stove
202 (90-346) ^a	2 (0.5-6) ^a	124 (36-217) ^a	(Zhi et al., 2009), improved stove with chimney
17	0.4	6.5	(Li et al., 2016), semi-coke briquettes
23-135	0.3-1	9-55	GAINS ^b

Table S2.4: Summary of PM emission factors for coal cooking and heating stoves

^a (min-max)

S3 Summary of particulate matter emissions factors for diesel generators

Emission factors (mg/MJ)		Shares (%)		Reference	
PM	BC	OC	BC	OC	-
69-189					Uma et al. (2004), 10 kW (higher value), 40 kW (lower value)
139			66%		Bond et al. (2004)
13/22					Gilmore et al. (2006), ICE 10 kW, with/without DPF
		116-585			Watson et al. (2006) ^a
59-190	12-54	30-120	31%	51%	Shah et al. (2007) ^b 300 kW 1985 Detroit Diesel V92, 2-str
45-219	30-145	8-56	67%	21%	Shah et al. (2007) ^b 350 kW 2000 Cat 3406C, 4-str
22-143	10-80	6-37	53%	25%	Shah et al. (2007) ^b 300 kW 1985 Detroit Diesel V92, 2-str, DOC
59-203	28-145	4-16	67%	8%	Shah et al. (2007) ^b 350 kW 2000 Cat 3406C, 4-str, DOC
23-190	9-96	10-81	49%	36%	Shah et al. (2007) ^b 300 kW 1985 Detroit Diesel V92, 2-str, DOC+FBC
4-26	2.5-19	1-3	76%	15%	Shah et al. (2007) ^b 350 kW 2000 Cat 3406C, 4-str, passive-DPF
1-3	0.8-2	1-6	67%	49%	Shah et al. (2007) ^b 350 kW 2000 Cat 3406C, 4-str, active-DPF
				20-70%	Watson et al. (2008)
14-42					Zhu et al. (2009) ^c
174-433					Tsai et al. (2010) ^d
55					Anayochukwu et al. (2013)
GAINS emiss	sion factor	s; the PM	value rep	resents PM	12.5
96	40	28	41%	29%	No control
48-64	20-26	14-19	41%	29%	Controlled, no DPF
<1-3	0.5-2	0.3-0.8			Controlled, with DPF
<1-3	0.5-2	0.3-0.8			Controlled, with DPF

Table S3.1: Summary of PM emission factors for diesel generator sets

^a Higher value with 10% load and lower value with 100% load for a 100 kW DG set

^b Lower value with 100% load and higher value with 10% load, share of BC/OC is average of all loads

^c Average of 14 military diesel generators with rated capacities of 10, 30, 60, and 100 kW under different load conditions. The fleet average EFs are 1.2+/-0.6 g/kg for PM.

^d Higher value with no load and lower value with 10 kW

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S4 Transport sector

	Table S4.1: Comparison of selected measured emissions factors and ranges used in the GAINS model for diesel and gasoline cars and
5	light duty vehicles.

	Emission factors (mg/MJ)			Reference
	PM	BC	OC	
Diesel				
Pre-/early regulation	44-67	9-17	13-34	(Subramanian et al., 2009)
Euro 1	67	17	13	(Subramanian et al., 2009)
Euro 2	30-33	7-16	8-12	(Cheung et al., 2009; Subramanian et al., 2009)
Euro 3	10-29			(Graham, 2005)
Euro 4	6-11	3-8	1-2	(Cheung et al., 2009; Geller et al., 2006)
Euro 4 with DPF	0.2-0.3	0.02-0.1	0.02-0.06	(Dwyer et al., 2010; Louis et al., 2016)
Pre-/early regulation	56-133	38-76	21-51	GAINS ^a
Euro 1	22-50	16-35	5-11	GAINS ^a ; for developing countries the values only marginally lower than pre/early regulation GAINS ^a ; for developing countries, the values only
Euro 2	15-40	12-32	3-6	marginally lower than pre/early regulation
Euro 3	11-29	10-22	1-2	GAINS ^a
Euro 4	5-20	4-17	0.5-1.6	GAINS ^a
Euro 4 with DPF	0.5-1	0.1-0.3	0.1-0.6	GAINS ^a
Gasoline				
Pre-/early regulation	4-10	0.5-2	2-10	see studies in Kupiainen and Klimont (2004, 2007)
Euro 1, 2	1-4	0.6-1.5	0.3-1.6	see studies in Kupiainen and Klimont (2004, 2007)
Euro 3	0.2-2	0.01-0.2	0.2-0.6	(Cheung et al., 2009; Geller et al., 2006; Graham, 2005)
Euro 4		0.001-0.4		(Louis et al., 2016)
Pre-/early regulation	6	1	3-4	GAINS ^a
Euro 1, 2	1-4	0.2-1	0.3-1.7	GAINS ^a
Euro 3, 4	0.3-1.1	0.05-0.5	0.1-0.4	GAINS ^a

	Emission factors (mg/MJ)		()	Reference
	PM	BC	OC	
Diesel heavy duty trucks				
Pre-/early regulation	28-33			(Herner et al., 2009; Yanowitz et al., 2000)
	44-244	4-50	15-122	(Subramanian et al., 2009), Bangkok, Thailand
	30-50			(Liu et al., 2009), on-road measurements in China
Euro I	11			(Yanowitz et al., 2000)
	22	4	9	(Subramanian et al., 2009), Bangkok, Thailand
	10-20			(Liu et al., 2009), on-road measurements in China
Euro II	22-44	2-9	7-22	(Subramanian et al., 2009), Bangkok, Thailand (Liu et al., 2009; Wang et al., 2011), on-road
	7-17	16		measurements in China
Euro III	3-7	9		(Liu et al., 2009; Wang et al., 2011), on-road measurements in China
Euro IV		4		(Wang et al., 2011), on-road measurements in China
Pre-/early regulation	34-107	17-53	10-37	GAINS ^a
Euro I	21-71	17-53	6-19	GAINS ^a
Euro II	11-44	7-30	2-10	GAINS ^a
Euro III	10-27	8-25	2-7	GAINS ^a
Euro IV, V	2-7	2-5	0.3-1	GAINS ^a
Euro VI	0.1-0.4	0.01-0.06	0.06-0.15	GAINS ^a

Table S4.2: Comparison of selected measured emissions factors and ranges used in the GAINS model for diesel heavy duty vehicles

	Emission fa	Emission factors (mg/MJ)		Reference
	PM	BC	OC	
Diesel locomotives				
Pre-/early regulation	on 49-67			(Dincer and Elbir, 2007; Johnson et al., 2013; Tang et al., 2015)
Regulated	20-40	20		(Dincer and Elbir, 2007; Galvis et al., 2013; Johnson et al., 2013; Tang et al., 2015)
	30	14		(Galvis et al., 2013)
	20	15		(Jaffe et al., 2014)
	37	21		(Krasowsky et al., 2015)
pre-regulated	49-98	24-45	12-25	GAINS ^a
regulated (stage I)	26-49	11-22	6-12	GAINS ^a
Agriculture				
Pre-regulation	141	58	41	(Kupiainen and Klimont, 2007)
	89	49		(EEA, 2013)
Stage I	20-39	16-21		(EEA, 2013)
Stage II	15	11.5		(EEA, 2013)
Pre-regulation	100-170	41-70	29-50	GAINS ^a
Stage I	57-96	23-40	16-27	GAINS ^a
Stage II, III	27-43	10-19	8-12	GAINS ^a
Stage IV,V	6-10	0.7-1.2	0.5-0.8	GAINS ^a
Construction				
Pre-regulation	140	65	30	(Kupiainen and Klimont, 2007)
	103	56		(EEA, 2013)
Stage I	85	47		(EEA, 2013)
Pre-regulation	95-140	46-68	21-31	GAINS ^a
Stage I	57-76	26-39	12-18	GAINS ^a
Stage II, III	24-36	12-17	5-8	GAINS ^a
Stage IV,V	6-8	0.8-1.2	0.4-0.6	GAINS ^a

Table S4.3: Comparison of selected measured emissions factors and ranges used in the GAIN	S model for non-road machinery.
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	Emission factors	s (mg/MJ)		References
	PM	BC	OC	
2-stroke				
Euro 0 mopeds	250 (198-295)			(Spezzano et al., 2008), hot start
	160 (121-878)			(Spezzano et al., 2008), cold start
Euro 1 mopeds	169 (102-235)			(Spezzano et al., 2008), hot start
	42 (26-71)			(Spezzano et al., 2008), cold start
Euro 2 mopeds	147-217			(Spezzano et al., 2008), hot start
	13-215			(Spezzano et al., 2008), cold start
CNG rickshaw, Delhi, India	124-160			(Grieshop et al., 2012)
Euro 0 mopeds	132-1400	10-75	90-1015	GAINS ^a
Euro 1 mopeds	12-450	7-49	40-300	GAINS ^a
Euro 2 mopeds	37-280	6-45	23-172	GAINS ^a
Euro 3 mopeds	14-112	3-30	8-61	GAINS ^a
4-stroke				
Motorcycles	2.6-3.7			(Yang et al., 2005), cold start
Euro 0 motorcycles	4			(Spezzano et al., 2007)
Euro 1 motorcycles	2			(Spezzano et al., 2007)
Rickshaw, Delhi, India	30-45			(Grieshop et al., 2012)
CNG rickshaw, Delhi, India	12-13			(Grieshop et al., 2012)
Euro 0 motorcycles	6-14	1-2	3-9	GAINS ^a
Euro 1 motorcycles	5-12	1-2	2-7	GAINS ^a
Euro 2 motorcycles	3-5	0.5-0.8	0.4-1.7	GAINS ^a
Euro 3 motorcycles	2-3	0.5-0.75	0.3-1.4	GAINS ^a

Table S4.4: Comparison of selected measured emissions factors and ranges used in the GAINS model for 2-wheelers.

	Emission factors (mg	/km)		
	PM_{10}	PM _{2.5}	BC	OC
Brake wear				
Cars	3.5 - 12	2.5 - 5	0.05 - 0.12	0.8 - 2.2
Light duty vehicles	3.5 – 19	2.5 - 8	0.05 - 0.2	0.8 - 3.5
Heavy duty vehicles	21 - 53	13 – 21	0.25 - 0.5	5 – 17
Tyre wear				
Cars	1.5 - 9	0.15 - 0.7	0.2 - 1	0.5 - 2.4
Light duty vehicles	2.5 - 7	0.2 - 0.7	0.35 - 1	0.85 - 2.4
Heavy duty vehicles	40 - 47	4.2 - 4.7	6 – 7	15 – 17
Road abrasion				
Cars & Light duty vehicles	7 - 10	3 – 5	0.15 - 0.6	0.7 - 1
	$30 - 140^{a}$	$20-80^{a}$	$0.2 - 1.5^{a}$	4-14 ^a
Heavy duty vehicles	38 - 50	18 - 27	0.7 - 1	3-5

Table S4.5: Summary of PM emission factor ranges used in the GAINS model for non-exhaust transport sources

^a vehicles with studded tires; variation between estimates for Scandinavian and alpine countries

S5 Industry

GAINS model PM emission factors (as used for the ECLIPSE *V5a*) for brick making compared with values used in GAINS previously (UNEP/WMO, 2011) and recent set of measurements on typical kilns used in South Asia (Weyant et al., 2014).

5 Table S5.1: Comparison of emissions factors used in the GAINS model for brick kilns with selected other stu	dies.
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Emission factors (g kg	⁻¹ brick)		References
PM2.5	BC	OC	
Clamp kiln			
1.6	0.35	0.3	(UNEP/WMO, 2011) ^a
1	0.3	0.1	GAINS (Asia)
1	0.35	0.15	GAINS (Latin America and Africa)
Downdraft kiln			
0.49	0.19	0.07	(Weyant et al., 2014)
0.97	0.29	0.09	GAINS (all regions)
Bull's trench kiln (BTK)			
1.31	0.27	0.24	(UNEP/WMO, 2011) ^a
0.19 (0.08-0.33)	0.15 (0.09-0.27)	0.007	(Weyant et al., 2014) ^b
0.18/0.8	0.13/0.25	0.01/0.07	GAINS (Asia); fixed /moving chimney
Vertical shaft brick kiln (VSBK	()		
0.77	0.175	0.15	(UNEP/WMO, 2011) ^a
0.07 (0.005-0.009)	0.0015 (0.001-0.002)	0.014	(Weyant et al., 2014) ^b
0.093	0.001-0.004	0.002-0.059	GAINS (Asia)
0.093	0.002	0.059	GAINS (Latin America and Africa)
Zig-zag kiln			
0.06 (0.03-0.06)	0.01 (0.014-0.03)	0.005	(Weyant et al., 2014) ^b
0.13	0.04	0.02	GAINS (Asia)
Tunnel kiln (coal)			
0.28	0.0035	0.003	(UNEP/WMO, 2011) ^a
0.24	0.01	< 0.00	(Weyant et al., 2014)
0.18	0.002	0.0035	GAINS (all regions)
Hoffman kiln			
0.08	0.003	0.005	GAINS (all regions)
Marquez kiln (MK)			
0.15	0.06	0.02	GAINS (Latin America)

^a Previous version of the GAINS model was used

^b Central value and ranges of average values; all measurement data provided in the original study

Brick sector production structure in Asia has been analysed in a number of studies addressing either the whole region where selected countries, typically key producers including China, India, Pakistan, Bangladesh, Vietnam, are discussed (AIT, 2003; BASIN, 1999; FAO, 1993; Heierli and Maithel, 2008; Maithel, 2014) or focusing on particular countries like China (Zhang, 1997), India (BASIN, 1998; Maithel et al., 2012; Verma and Uppal, 2013), Bangladesh (Croitoru and Sarraf, 2012;

- 5 Guttikunda et al., 2013; World Bank, 2011), Cambodia (Rozemuller, 1999), Afghanistan (Samuel Hall Consulting, 2011), Nepal (Heierli et al., 2007). More recently, a number of development programs and local air pollution studies focused on this sector in the Latin America and Caribbean regions, including some where information about kiln structure was collected (Bellprat, 2009; EELA, 2011; Erbe, 2011; PRAL, 2012; Stratus Consulting, 2014; SwissContact, 2014a). Fewer assessments exist for Africa (Scott, 2013; SwissContact, 2014c). The updated and country specific data for Latin America and Caribbean
- 10 (LAC) is included only in version *V5a* of ECLIPSE since the previous versions included just five regions for the whole LAC; Argentina, Brazil, Chile, Mexico, other LAC.
 - GAINS activity data has been built on the basis of several regional studies where production, energy efficiency, and sector structure were discussed, i.e., Asia (AIT, 2003; Co et al., 2009; Croitoru and Sarraf, 2012; FAO, 1993; Guttikunda et al., 2013; Heierli et al., 2007; Heierli and Maithel, 2008; Maithel, 2014; Maithel et al., 2012; Samuel Hall Consulting, 2011;
- 15 Subrahmanya, 2006; Verma and Uppal, 2013; World Bank, 2011; Zhang, 1997), Africa (Alam, 2006; Scott, 2013; SwissContact, 2014c), Latin America and Caribbean (Bellprat, 2009; EELA, 2011; PRAL, 2012; Stratus Consulting, 2014; SwissContact, 2014b). For several countries where we found no regional analysis, the United Nations data on 'building bricks, made of clay' was used (<u>http://unstats.un.org/unsd/industry/commoditylist2.asp</u>). There are some differences between different versions of the ECLIPSE datasets; specifically during the development of the *V5a* version, the data for all countries
- 20 in Latin America and Caribbean was revisited and updated, and a new version of the UN statistics was downloaded.

	1990	1995	2000	2005	2010
Global	1542	2357	2688	3022	3574
Asia	1314	2130	2530	2819	3320
of which:					
China	1050	1800	2106	2204	2508
India	131	178	254	406	553
Vietnam	20	20	27	46	65
Bangladesh	9	15	18	17	25
Pakistan	32	41	50	59	74
Other Asia	71	75	76	87	95
Africa	18	18	15	17	22
Europe	158	156	72	82	79
Latin America and Caribbean	29	30	43	75	127
Other	23	23	27	29	25

Table S5.2: Brick production in key regions; GAINS model assumptions - ECLIPSE V5a, Tg bricks year⁻¹

S6 Emissions of PM species over time in ECLIPSE datasets

The Fig S6.1 shows emissions of PM10, PM2.5, BC, and OC calculated with the GAINS model within different versions of the ECLIPSE dataset. These have been created between 2013 and 2015 and include a number of updates to activity data and emission factors; the methodology remained the same. The changes for PM10 and PM2.5 are similar, driven by updates of

- 5 activity data, i.e., the energy statistics from IEA were reimported for the whole time series for the version *V5* and *V5a* and for China the regional coal statistics were used. Control strategies have been updated continuously considering more up to date information available over time. Additionally, in version *V5a* Latin America and Caribbean were revised since higher spatial resolution was introduced in the GAINS model. Several of the above mentioned updates affected also emissions of BC and OC but the largest impact on the BC emissions was due to introduction of emissions from kerosene lamps which
- 10 were not specifically distinguished in *V4a*; this represents the key component of the higher emissions in *V5*, *V5a*. For OC the change is in the opposite direction and *V5a* has significantly lower emissions than previous versions which is due to update of the OC emission factor for residential cooking in Asia and Africa.



15 **Figure S6.1.** Global emissions of PM (excluding international shipping and open biomass burning) in the period 1990-2010 in different ECLIPSE scenarios; unit [Tg year⁻¹]

Table S6.2: Global anthropogenic (excluding international shipping & aviation) emissions of PM10 in ECLIPSE V5a; [Gg year-1]

Region	1990	1995	2000	2005	2010
1 Canada	333	315	345	337	334
2 USA	2416	2158	1954	1920	1630
3 Mexico	643	621	653	574	572
4 Rest Central America	454	455	479	498	516
5 Brazil	1228	1295	1250	1385	1456
6 Rest South America	1018	1155	1138	1131	1192
7 Northern Africa	1022	1152	1355	1144	1194
8 Other Africa	4393	4993	5831	6425	7150
10 South Africa	682	738	747	848	818
11 Western Europe	3294	2458	2031	1747	1577
12 Central Europe	2944	1608	1236	1046	1038
13 Turkey	1007	756	525	477	571
14 Ukraine+	1854	856	679	707	680
15 Asia-Stan	836	325	303	314	392
16 Russia+	5833	2434	2314	2316	2161
17 Middle East	836	954	1055	962	996
18 India	7828	8785	8654	7952	8061
19 Korea	1227	913	844	816	768
20 China+	14057	17612	18205	21230	21976
21 Southeastern Asia	2291	2855	2783	2451	2526
22 Indonesia+	1383	1576	1673	1768	1902
23 Japan	545	435	354	319	267
24 Oceania	295	303	354	354	342
25 Rest South Asia	1695	1894	2211	2349	2533
Global	58112	56646	56974	59071	60651

Table S6.3: Global anthropogenic (excluding international shipping & aviation) emissions of PM2.5 in ECLIPSE V5a; [Gg year-1]

Region	1990	1995	2000	2005	2010
1 Canada	252	244	250	242	241
2 USA	1629	1482	1296	1275	1027
3 Mexico	495	498	526	459	454
4 Rest Central America	395	394	416	428	446
5 Brazil	938	974	933	1054	1098
6 Rest South America	825	933	909	901	949
7 Northern Africa	762	852	982	847	909
8 Other Africa	4056	4606	5308	5887	6575
10 South Africa	408	444	431	501	490
11 Western Europe	2125	1700	1360	1157	1037
12 Central Europe	1610	1020	843	752	775
13 Turkey	585	480	388	356	425
14 Ukraine+	1072	531	464	483	455
15 Asia-Stan	562	222	211	222	283
16 Russia+	3702	1614	1530	1495	1413
17 Middle East	686	778	845	784	794
18 India	5768	6453	6472	5957	6032
19 Korea	784	600	547	565	529
20 China+	10863	13072	13633	15673	16096
21 Southeastern Asia	1878	2257	2198	1974	2012
22 Indonesia+	1230	1371	1447	1510	1604
23 Japan	337	295	236	203	160
24 Oceania	188	193	210	201	188
25 Rest South Asia	1455	1629	1859	1962	2065
Global	42606	42640	43294	44888	46055

Table S6.4: Global anthropogenic (excluding international shipping & aviation) emissions of PM1 in ECLIPSE V5a; [Gg year-1]

Region	1990	1995	2000	2005	2010
1 Canada	184	195	196	187	190
2 USA	1163	1095	949	930	727
3 Mexico	375	378	395	361	357
4 Rest Central America	329	331	353	366	390
5 Brazil	706	720	718	819	846
6 Rest South America	657	732	708	712	764
7 Northern Africa	447	476	514	485	542
8 Other Africa	3724	4213	4838	5416	6064
10 South Africa	285	309	307	354	354
11 Western Europe	1397	1171	966	834	751
12 Central Europe	894	667	619	579	607
13 Turkey	386	341	286	263	311
14 Ukraine+	565	325	279	278	261
15 Asia-Stan	292	154	146	154	198
16 Russia+	1988	1078	1011	936	852
17 Middle East	501	562	596	614	615
18 India	4500	4992	5016	4700	5031
19 Korea	635	510	450	464	429
20 China+	9153	11251	11731	12473	11606
21 Southeastern Asia	1800	2204	2093	1791	1803
22 Indonesia+	1135	1254	1315	1373	1453
23 Japan	258	229	157	126	87
24 Oceania	140	146	155	143	133
25 Rest South Asia	1303	1445	1625	1714	1811
Global	32816	34780	35422	36073	36180

	Table S6.5: Global anthropogenic	(excluding international shi	ipping & aviation)	emissions of BC in ECLIPSE	V5a; [Gg year ⁻¹]
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Region	1990	1995	2000	2005	2010
1 Canada	44	49	51	49	49
2 USA	311	291	281	279	201
3 Mexico	76	77	82	84	88
4 Rest Central America	52	54	61	65	71
5 Brazil	143	148	160	171	179
6 Rest South America	115	135	140	150	169
7 Northern Africa	127	120	117	121	140
8 Other Africa	752	836	942	1030	1135
10 South Africa	57	59	57	74	72
11 Western Europe	331	335	307	287	246
12 Central Europe	126	112	112	121	134
13 Turkey	60	59	53	51	67
14 Ukraine+	88	59	45	41	36
15 Asia-Stan	50	28	33	38	55
16 Russia+	439	251	238	226	177
17 Middle East	174	183	210	243	262
18 India	853	931	884	908	1022
19 Korea	135	84	71	84	74
20 China+	1348	1347	1655	1823	1924
21 Southeastern Asia	300	299	304	328	333
22 Indonesia+	243	260	275	279	290
23 Japan	67	74	66	50	29
24 Oceania	30	32	35	35	33
25 Rest South Asia	288	304	325	337	348
Global	6210	6129	6505	6872	7134

	Table S6.6: Global anthropogenic	(excluding international	shipping & aviation)	emissions of OC in ECLIPSE	V5a; [Gg year ⁻¹]
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Region	1990	1995	2000	2005	2010
1 Canada	72	77	77	72	74
2 USA	448	434	388	379	308
3 Mexico	162	162	164	158	155
4 Rest Central America	144	149	159	169	181
5 Brazil	251	258	275	311	314
6 Rest South America	297	329	315	324	344
7 Northern Africa	145	150	155	166	192
8 Other Africa	1627	1842	2124	2408	2701
10 South Africa	101	108	110	129	130
11 Western Europe	495	422	343	284	253
12 Central Europe	224	201	217	220	234
13 Turkey	114	108	95	88	107
14 Ukraine+	149	102	82	77	72
15 Asia-Stan	90	66	62	64	86
16 Russia+	509	332	304	256	231
17 Middle East	190	217	220	237	229
18 India	1530	1623	1596	1630	1755
19 Korea	200	157	147	157	148
20 China+	3147	3264	3500	3564	3599
21 Southeastern Asia	526	548	567	598	632
22 Indonesia+	431	473	514	551	595
23 Japan	51	54	49	40	29
24 Oceania	52	55	57	51	46
25 Rest South Asia	502	562	628	680	726
Global	11456	11695	12150	12610	13140

S7 Regional resolution

5

The spatial resolution of the GAINS model is discussed section 2.4 of the paper and the list of all 170 regions can be obtained from the online model. In principle, GAINS distinguishes single countries in Europe (exception in Russia for which European and Asian part is included separately) North America, Australia and New Zealand, for Asia several larger countries are divided into provinces or states (larger administrative units in, e.g., China, India, Indonesia, Japan, etc.) while

- Middle East represented as one region or (most recent versions) distinguishes Iran, Saudi Arabia, Israel, and the rest of Middle East. Africa is divided into four regions: South Africa, Egypt, North Africa, and other Africa. Latin America and Caribbean includes now 13 regions with all larger countries treated separately while Central America as well as Caribbean states are grouped in two regions. While such resolution of 170 regions is used for the calculation of emissions, the
 presentation of data and results differs between the on-line models available for specific world regions, e.g., for Europe and
- Asia the full resolution is available, while in the global model application (<u>http://magcat.iiasa.ac.at/gains/IAM/index.login</u>) the data and results are presented for 25 regions (Fig. S7.1). This follows closely the IMAGE model¹ resolution; often used or compatible with several global integrated assessment models.



15 Figure S7.1. Regions distinguished in the global GAINS online application.

http://themasites.pbl.nl/models/image/index.php/Region_classification_map

S8 Sectoral resolution

Table S8.1: Source sector resolution	in the GAINS model for calcu	lation of PM emissions
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Key source category	Source sectors	Fuel category or activity type
Energy sector		
	Power plants (distinguishing small, large, old, new plants);	Coal, oil, gas, biomass, waste
	Diesel generators;	
	Extraction and distribution of solid and liquid fuels (fugitive as	Coal, oil
	well as combustion from gas flaring)	Draduction
	Briquette production	Production
Residential combustion	,	
	Cooking stoves; Heating (distinguishing fireplaces, stoves, house	Coal, fuelwood, dung, oil, gas,
	boilers, mid-size residential boilers)	agricultural residues, charcoal
	Weste (trash) hurming	Weste
	waste (trash) burning	w aste
Industrial combustion		
	Iron and Steel; Pulp and Paper; Chemical; Non-ferrous metals;	Coal, oil, gas, biomass, waste
	Non-metallic minerals (excl. Bricks); Other	
Industrial processes		
	Iron and steel industry divided into: Pig iron; Coke ovens;	Production
	Agglomeration plants – pellets; Agglomeration plants – sinter;	
	Open hearth; Electric Arc; Basic oxygen; Rolling mills; Cast Iron	
	Non-ferrous metals (copper and nickel smelters); Primary	Production
	aluminium; Secondary aluminium; Cement; Lime; Carbon black	
	manufacturing: Pulp and paper	
	Refineries	Crude oil throughput
	Handling and storage of bulk industrial and agricultural products	Million tons of products
	(fugitive)	in the set produces
Road transport		
Roua transport	Passenger cars and yans: Light duty vehicles: Heavy duty	Gasoline diesel CNG LPG
	vehicles: Busses: Motorcycles (4-stroke): Mopeds (2-stroke)	km driven (for calculation of
		non-exhaust emissions)
Non road transport		,
won-roaa iransport	Agricultural and forestry: Construction and mining: Railways:	Diesel gasoline CNG jet fuel
	Inland navigation: Coastal shipping: Aviation (landing and take-	and kerosene heavy fuel oil
	off): 2-stroke engines (e.g., in household, forestry, etc.): Other	coal
	land based machinery	cour
Acriculture		
Agriculture	Arable land operations	Arable land area
	Livestock housing	Cattle nigs poultry
	Open burning of agricultural waste	Waste burned
Othon		
Oiner	Fireworks: Cigarette smoking: Barbaques: Cremation	Population
	Construction (fugitive)	Constructed area
		Construction area

S8 Comparison of regional estimates with selected studies

The table S8.1 provides ECLIPSE *V5a* PM estimates for selected regions and years (from the period 2000-2010) and compares them with selected regional peer-reviewed studies.

Region – (Source) – Year	PM10	PM2.5	PM1	BC	OC
Global					
This study – 1995	57830	43762	35902	6206	11949
(Bond et al., 2004) - 1996				4997	10481
This study - 2000	58366	44613	36741	6595	12449
(Bond et al., 2013) - 2000				4870	
This study - 2010	62537	47843	37819	7264	13548
HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010	50292	32761		5525	13581
China					
This study - 2000	18061	13554	11685	1646	3487
(Cao et al., 2006) - 2000				1496	4211
(Streets et al., 2003) - 2000				1049	3385
(Klimont et al., 2009) - 2000				1345	3205
(Lu et al., 2011) - 2000				1244	2823
(Ohara et al., 2007) - 2000				1093	2563
(Bond et al., 2013) - 2000				1200 ^b	2800^{b}
(Zhang et al., 2006) - 2001	17120	12100			
This study - 2005	21087	15593	12428	1813	3552
(Zhang et al., 2009) - 2006	18223	13266		1811	3217
(Klimont et al., 2009) - 2005				1366	2812
This study - 2010	21827	16019	11564	1915	3589
(Lu et al., 2011) - 2010				1838	3907
(Kurokawa et al., 2013) - 2008	21606	14514		1589	3081
(Guan et al., 2014) - 2010		12100			
HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010	16615	12199		1764	3384
(Kondo et al., 2011) - 2008				1940	
India					
This study, 2000	8654	6472	5016	884	1596
(Streets et al., 2003) - 2000				600	2837
(Ohara et al., 2007) - 2000				795	3268
(Klimont et al., 2009) - 2000				842	1887
(Lu et al., 2011) - 2000				736	1990
(Bond et al., 2013) - /2000				500 ^b	1600 ^b

Table S8.1: Comparison of regional estimates for anthropogenic ^a emissions of PM species, Gg year⁻¹

Region – (Source) – Year	PM10	PM2.5	PM1	BC	OC
(Reddy and Venkataraman, 2002a, 2002b) - 1998-99		4300		380	1250
This study, 2005	7952	5957	4700	908	1630
(Zhang et al., 2009) - 2006	4002	3111		344	888
(Klimont et al., 2009) - 2005				1029	2132
This study, 2010	8061	6032	5091	1022	1755
(Lu et al., 2011) - 2010				996	2582
HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010	8280	6230		1019	2530
(Kurokawa et al., 2013) - 2008	6651	4884		713	2286
Europe ^c					
This study - 1995	6905	4584	3071	675	1021
(Kupiainen and Klimont, 2007) - 1995				717	1053
(Schaap et al., 2004) - 1995				760	
(Bond et al., 2004) - 1996				678	947
This study - 2000	5579	3843	2668	618	910
(Kupiainen and Klimont, 2007) - 2000				680	996
(Kupiainen and Klimont, 2004) - 2000			2772	672	988
This study - 2010	5008	3471	2393	562	806
HTAP_v2 (Janssens-Maenhout et al., 2015) ^d - 2010	2951	2133		382	638
LRTAP reporting (<u>www.ceip.at</u>) - 2010	4784	3250			
Russian Federation					
This study - 2010	2108	1368	815	170	213
HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010	562	313		60	42
(Huang et al., 2015) - 2010				224	
Russian Federation – European part only					
This study - 2010	1090	734	427	71	122
LRTAP reporting (<u>www.ceip.at</u>) - 2010	569	367			
US					
This study - 2000	1954	1296	949	289	388
(Battye et al., 2002) - 1999				430	
(Reff et al., 2009) - 2000				440	960
(Bond et al., 2013) - 2000				350 ^b	500 ^b
This study - 2010	1630	1027	727	201	308
(US EPA, 2011) ^e - 2011	17597	4513		513 (313 ^f)	
HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010	1973	1640		295	471

^a Based on the information available in the quoted studies, all presented estimates exclude forest fires but include agricultural burning, unless stated otherwise; ^b Excluding agricultural burning; ^c Includes European part of Russian Federation (except HTAP_v2); ^d Excluding any territories of Russian Federation; ^e Including wildfires and prescribed burning; ^f Excluding wildfires and prescribed burning

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