

Emission inventories of primary particles and pollutant gases for China

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Detailed high-resolution emission inventories of primary particles (PM_{2.5}, BC and OC) and pollutant gases (SO₂, NO_x, NH₃, CO and VOCs) for China in 2007 were constructed on the basis of the latest fuel consumption data, mostly at the county level, and from socio-economic statistics and data on fossil and biomass fuels obtained from government agencies. New emission factors reflecting local features were also used. The calculated emissions were 13.212 Mt PM_{2.5}, 1.4 Mt BC, 2.946 Mt OC, 31.584 Mt SO₂, 23.248 Mt NO_x, 16.017 Mt NH₃, 164.856 Mt CO and 35.464 Mt VOCs. The national and regional emissions were gridded with 0.5° × 0.5° resolution for use in air quality models. Larger emissions were found in eastern and central China than in western China. The emissions estimated here are roughly equal to those obtained in previous studies, but with different contributions from because of seasonal changes in residential heating and biomass combustion. Finally, uncertainties in inventories were analyzed.

China, primary particle, pollutant gas, emission inventory, seasonality

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An accurate description of China's regional emissions in terms of the temporal and spatial distributions of particulate matter (PM) and pollutant gases is important for the modeling of regional environmental quality, pollution prevention, environmental policy development and climate effect research [1–3]. Because of their impacts on human health, air quality and climate, the importance of PM and pollutant gases has been recognized by scientists worldwide [4–6]. SO₂, NO_x and NH₃ are the reactive gases most critical in acid deposition [7–9]. Combining with CO and VOCs, they are also important precursor gases to form secondary aerosols in the atmosphere and are largely responsible for the reduced visibility and formation of haze. VOCs, CO and NO_x are the most important precursors in the formation of O₃ and photochemical pollution [10].

PM, black carbon (BC), organic carbon (OC), SO₂, NO_x

and CO are mainly emitted in combustion processes, in particular, the burning of coal and biomass. About 97% of all PM_{2.5}, 90% of SO₂, 70% of NO_x and 32% of CO are produced in the combustion of coal [11–17]. NH₃ is mainly released by agricultural processes (about 20%–33% of the total NH₃) and animal decomposition of organic matter (about 50%–60%) [18,19]. VOC emission sources are more complex and mainly relate to transport, solvent evaporation and waste disposal [12,20]. Located in East Asia, China has a large area with a relatively non-modern economy and lifestyle and extensive use of coal and biomass as fuels, and a high level of poultry and livestock feeding with less advanced waste disposal methods. All these factors contribute to large emissions of particles and pollution gases to the global environment [3,17,21–24]. Because of China's unique geographical location and rapid economic development, changes in China's emissions (annual, seasonal, and monthly changes) are large and yet difficult to accurately

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estimate.

In the 1990s, foreign scholars systematically studied East Asian (mainly Chinese) emissions [3,17,23–28], but these studies involved great uncertainty owing to the lack of detailed information on China, especially important data on emission factors. Simulation and experimental studies have shown that these estimates, including the Streets' emission inventories, may be underestimated by 50% [29] for some species. In particular, the absence of data on China's rural and township industries introduces large uncertainty into the emission inventories. In addition, some emission inventories did not take into account seasonal variations, especially those in biomass combustion in rural China. Because of the heavy pollution of PM and acid rain in China, some national scholars have begun to construct emission inventories of PM and pollutant gases [11,13,15,16,18,19,30–34]. However, many of these studies did not give a spatial and temporal distribution, and thus, it is difficult to support a numerical simulation for comprehensive analysis of the situation of China's emissions at the required spatial and temporal resolution.

On the technical front of the emission inventories, many scholars have carried out much pioneering work [3,20,35]. Some domestic emission inventories have focused on information relating to technology and equipment [12]. As more domestic emission experiment data have become available, the uncertainty in inventories has tended to reduce.

In this work, to meet the needs of the National Key Basic Research Development Program (Aerosols over China and their Climate Effect) for numerical simulations, the emissions of PM_{2.5}, BC, OC and pollution gases SO₂, NO_x, NH₃, CO and VOCs from mainland China (excluding Hong Kong, Macao and Taiwan) were estimated with fine spatial and temporal resolutions.

1 Methodology

Emission inventories are obtained using the level of emissions in different regions and for various types of sources. The general approach to estimate emissions of PM_{2.5}, BC, OC, SO₂, NO_x, CO and VOCs employs an equation described by Klimont et al. [20]:

$$E = \sum_j \sum_k \sum_l A_{j,k,l} EF_{j,k,l} (1 - \eta_{j,k,l}),$$

where E is the emission, A is the activity rate, EF is the emission factor and η is the removal efficiency; the subscripts j , k and l indicate the region, sector and fuel/activity type, respectively.

Emissions from forest and grassland fire are calculated as

$$E = \sum_j A_j B_j C_j EF_j,$$

where E is the emission, A is the burning area, B is the fuel load, C is the burning efficiency and EF is the emission factor; j indicates the region.

NH₃ emission is calculated using

$$E_i = \sum_j L_j Ef_{i,j} + \sum_k (Ef_{i,k} FC_{i,k} + Ef_{i,k} FP_{i,k}) + \sum_l FL_l Ef_{i,l},$$

where L is the animal population, FC is the fertilizer consumption, FP is the fertilizer production, Ef is the emission factor and FL is the fuel consumption. i , j and l indicate the region, livestock and poultry type, and fertilizer type and fuel type.

1.1 Emission sources

Emission source types in this work include biomass combustion (open burning of agricultural waste and forest and grassland fires), residences, power generation, industry, transportation, livestock and poultry breeding, fertilizer use, waste disposal, solvent use, and light industrial product manufacture. Emission sources are divided into two major categories: point sources and area sources.

A total of 383 large point sources were accounted for in this inventory, covering mainly power plants with generating capacity greater than 6.0 MW [36], while small power plants and power plants that are about to be decommissioned or modified were not included in the statistics. These large point sources are located mainly in eastern China and their main fuel is coal. A total of 98 large fertilizer manufacturing firms (<http://www.chinafufuhei.com/>) and 23 petrochemical manufacturers with processing capacity over 5 million tons (<http://www.cpcic.com.cn/html/>), also located mainly in eastern China, were considered. Each point source is entered with the latitude and longitude of the address identification. Area sources include all stationary sources that are not included in the point-source category. The base year for the estimation in this work is 2007.

1.2 Emission factors

In most cases, the emission factors are simply obtained from averaged data under an acceptable condition, which is not recommended for all standards or limits of sources of emission. There are many direct experimental measurements of emission factors of PM and pollutant gases emitted in fuel combustion around the globe, but experimental work in China is still very limited. The emission factors in this work mainly relate to domestic and foreign research results [3,11,12,15–18,20,22,31,32,35,37–41], and we also cited relatively new experimental results [13,42–49], especially in the calculation of emissions from biomass combustion and motor vehicle emissions and VOC emissions. The sulfur content of domestic coal and oil and the proportion of SO₂ produced in the burning of fuel were taken from the research results of Luo [50] and Arndt [51]. Since a large

number of emission factors were considered in this work (e.g. more than 100 factors for VOCs), they are not all listed in this paper (this data is available on request).

1.3 Basic data

The basic data used to estimate emissions, such as population, area, gross domestic product, industrial output, fuel usage, vegetative cover, meteorological data and soil pH, were taken from the National Bureau of Statistics and other government agencies, mostly at county level. Fossil fuels including coal, oil, natural gas, liquefied gas and coal are China's main energy fuels [52]. Biomass fuels include straw and firewood, and are used mainly for cooking and heating in rural areas [53]. Emission factors for the open burning of biomass were obtained from related research results [54]. Emissions from forest fires [55] and grass fires [56] were estimated from relevant data of the fuel load [57] and combustion efficiency [57,58]. The number of poultry livestock [56], chemical fertilizer application [56], various daily pro-

duction capacities, number of motor vehicles, waste disposal and other public data were obtained from national statistical offices [52].

2 Results and discussion

2.1 National and regional emissions

The national and regional emissions of primary particles and pollutant gases in China for 2007 are listed in Table 1. Large provincial contributions are observed for Shandong, Hebei, Shanxi, Henan and Sichuan because these regions have high rural population densities and coal consumption. Higher coal consumption, especially coal burned in uncontrolled or poorly controlled coal-fired boilers, kilns and furnaces in rural industry result in higher emissions, while high rural population densities result in higher emissions from biomass and coal combustion in the residential sector. Owing to low coal consumption and lower rural population densities, emissions from western China (Qinghai, Gansu

Table 1 Provincial emission inventories for 2007 (10^4 t)

Area	PM _{2.5}	BC	OC	SO ₂	NO _x	NH ₃	CO	VOCs
Beijing	16.2	2.3	4.2	17.2	43.7	11.7	199.8	74.4
Tianjin	15.1	1.5	3.0	26.3	33.7	10.2	162.5	46.7
Hebei	117.2	11.2	23.5	231.8	163.4	99.4	1266.9	232.7
Shanxi	116.0	9.3	17.9	219.0	116.9	40.0	1319.3	161.7
Inner Mongolia	46.1	5.6	12.2	104.7	115.9	50.8	520.4	95.5
Liaoning	58.3	6.2	11.6	123.6	108.7	62.4	675.5	201.0
Jilin	32.1	4.3	9.2	33.2	56.2	60.1	445.5	89.2
Heilongjiang	38.6	4.8	11.9	26.4	69.1	40.0	570.3	157.5
Shanghai	20.6	1.9	2.9	54.9	50.9	8.3	193.0	99.0
Jiangsu	69.0	6.0	15.3	137.9	162.3	88.8	801.0	240.9
Zhejiang	34.2	3.3	5.8	87.1	107.6	30.3	421.8	153.6
Anhui	51.4	5.9	14.6	90.6	75.7	70.0	732.7	102.6
Fujian	26.2	2.4	4.2	51.6	55.4	27.7	305.6	96.0
Jiangxi	26.2	2.6	5.0	87.5	40.5	39.2	309.3	112.2
Shandong	118.5	11.3	26.2	347.3	210.7	161.8	1398.9	279.1
Henan	93.2	9.6	22.4	234.2	147.1	138.5	1149.3	183.4
Hubei	62.5	6.4	13.7	98.4	85.1	64.9	748.7	101.4
Hunan	52.6	5.7	10.8	165.1	72.1	64.7	653.6	94.4
Guangdong	47.3	5.5	8.4	153.6	153.6	77.7	665.5	279.0
Guangxi	36.1	4.5	8.9	182.6	51.5	66.0	609.4	117.9
Hainan	4.1	0.5	0.7	9.6	11.3	9.9	44.9	10.9
Chongqing	16.0	2.1	4.3	109.8	30.4	26.6	263.5	45.3
Sichuan	55.7	7.3	18.5	91.5	80.7	106.8	865.4	168.9
Guizhou	38.1	5.6	9.9	178.8	63.9	44.5	539.5	84.2
Yunnan	34.1	3.7	6.9	56.5	55.4	58.0	466.4	94.0
Tibet	6.8	0.4	0.7	18.5	9.9	19.8	13.5	2.9
Shaanxi	33.9	3.7	7.9	104.4	54.6	34.3	463.5	82.5
Gansu	22.3	2.6	6.2	28.8	31.6	30.7	252.9	60.8
Qinghai	4.5	0.5	1.2	3.4	7.9	16.2	53.2	6.5
Ningxia	9.0	0.8	1.8	52.7	24.0	10.1	108.7	18.6
Xinjiang	19.5	2.3	4.9	31.5	35.1	32.6	265.5	53.5
Total	1321.2	139.9	294.6	3158.4	2324.8	1601.7	16485.6	3546.4

and Tibet) and Inner Mongolia are lower.

Figure 1 shows the gridded primary particle and pollutant gases emissions for the year 2007 at $0.5^\circ \times 0.5^\circ$ resolution for BC and NH_3 . As most of the basic data are at county level, we can better localize the emissions on higher resolution maps in many cases. When county-level data are not available in some areas, we distribute the provincial data for each category using appropriate socio-economic statistics such as population, area, industrial output or gross domestic product, crop acreage, forest lawn area, food crop production, per capita income, the amount of chemical fertilizer and livestock. The distribution method employed has been used by many scholars [3,12,33].

As shown in Figure 1, the emission densities are higher in eastern and central China, such as in the provinces of Hebei, Shandong, Henan, Shanxi and Jiangsu, and are con-

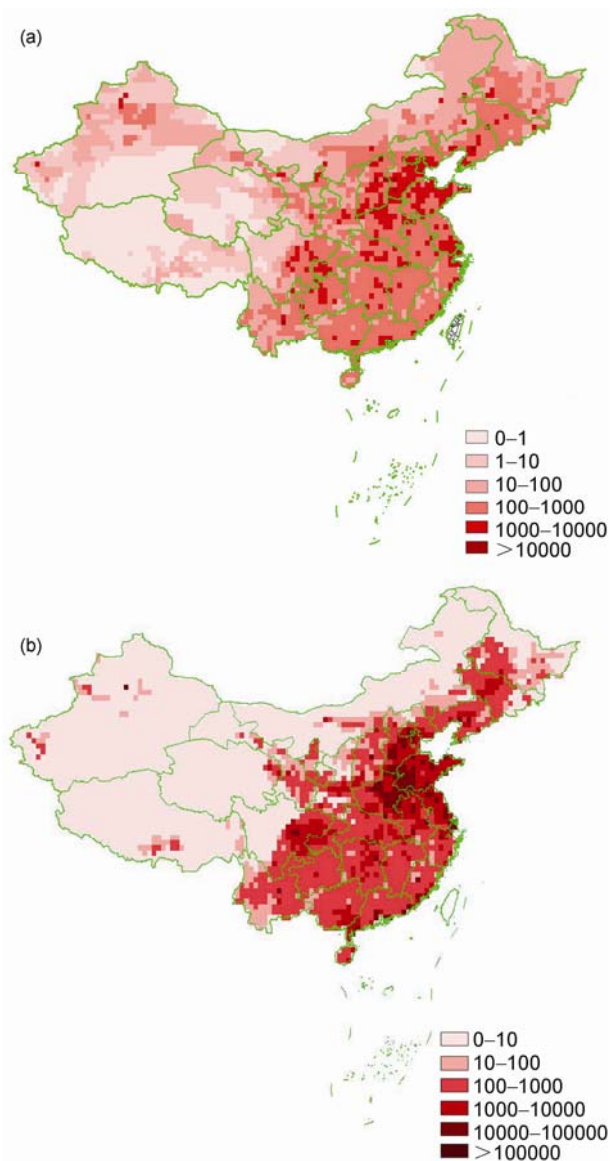


Figure 1 Gridded emissions for the year 2007 in China ($0.5^\circ \times 0.5^\circ$, unit: t/grid). (a) BC; (d) NH_3 .

centrated in a curve around the agricultural and industrial heartland of China, from northeastern China to eastern China, while areas of lowest emission are in western China and Inner Mongolia, which have lower rural population densities and economic levels.

2.2 Emission estimates by sectors

Emissions estimated by sector are listed in Table 2. Large contributions to $\text{PM}_{2.5}$, BC, OC and CO emissions come from the industrial and residential sectors, while coal-fired power generation is the largest contributor to SO_2 and NO_x emissions. NH_3 primarily comes from poultry (livestock), human waste disposal and agricultural production, such as the use of chemical fertilizers. VOCs are mainly derived from biomass combustion, industrial solid-waste incineration, other waste-disposal processes and evaporating solvents used in printing.

2.3 Comparison with other inventories

There are significant improvements in our emission inventories compared with previous inventories. First, our inventories are based on the most recent data available (statistical data of fuel consumption, population and technology for 2007 provided by the Chinese government). Second, emission factors of biofuel combustion, residential coal combustion and vehicles were derived from local measurements.

Some scholars have compared earlier emission inventories for China [12,28] as summarized in Table 3. Except in the cases of NH_3 and VOCs, our estimated emissions are close to those in previous inventories [12,28,59], but the emissions of different species and sectors are not the same. For example, the emission ratios by sector between this work for the year 2007 and Zhang's inventory [12] for the year 2006 are 1.02 (power generation), 0.50 (transport), 1.65 (industry) and 2.08 (residential). These differences can be largely attributed to the classification methodology for the emission sources. It should also be noted that emissions from coal combustion in rural industry, rural residents and open biomass combustion were significant but often underestimated in previous studies. The same underestimation has been noted by Wang et al. [33] for emissions in Shandong Province. Another important reason for these differences is that the latest government statistics and local emission factors in China are used in the present work. The emission ratios of the main sectors and the emission distribution processed using a geographical information system (Figure 1) in this work are consistent with relatively new inventories [3,12,28,35].

2.4 Seasonality of emissions

One can expect seasonal variations [3,34] in emissions due to (1) people in northern China burning coal or biofuel for

Table 2 Summary of emission estimates by sector

Species	Emission by sector (10 ⁴ t)						Total
	Biomass burning	Power generation	Transport	Industry	Residential		
PM _{2.5}	66.7	18.6	59.9	905.9	270.1		1321.2
BC	10.4	1.3	10.3	52.9	65.1		139.9
OC	43.3	1.7	14.5	80.5	154.6		294.6
SO ₂	1.4	961.6	41.1	1995.2	159.1		3158.4
NO _x	50.5	939.3	256.7	886.0	192.5		2324.8
CO	1227.0	2905.4	1405.1	5532.8	5415.3		16485.6
	Waste disposal	Agricultural	Industry	Fertilizer production			
NH ₃	957.6	362.3	248.0	33.8			1601.7
	Transport	Solvent use	Industry	Residential	Petrochemical production	Waste disposal	
VOCs	409.9	865.3	266.8	575.3	443.7	985.4	3546.4

Table 3 Comparison of estimates of Chinese emissions

Study	Base year	Emissions (10 ⁴ t)							
		PM _{2.5}	BC	OC	SO ₂	NO _x	CO	NH ₃	VOCs
This work	2007	1321.2	139.9	294.6	3158.4	2324.8	16485.6	1601.7	3546.4
Zhang et al. [12]	2006	1326.6	181.1	321.7	3102.0	2083.0	16688.9		2324.7
Ohara [28]	2003		114	262	3660	1450	15800		
Wei et al. [59]	2005								2010
Klimont et al. [60]	2005		136.6	281.2	3436.4	1692.6			
Lu et al. [61]	2007				3232.3				

heat but people in southern China not doing so, (2) emissions from poultry and livestock waste disposal in rural China being dependent on outdoor temperature, (3) open burning of agricultural waste in the field mainly occurring after the harvest period and (4) the fertilizer application time being closely related to phenological period and the farming season. Considering the above factors, monthly emissions at the national level are listed in Table 4 by species and strong seasonality is observed for PM_{2.5}, BC, OC, CO, NH₃ and VOC emissions. As the emission of NH₃ is more sensitive to temperature change, it has the strongest seasonal variation, with an emission ratio of 4.5 between its maximum (July) and minimum (December). Observation data for the seasonal variation in air pollutants have been discussed in detail [62,63].

3 Uncertainty in emission estimations

Generally, the estimated uncertainty of emissions is a com-

bination of the uncertainties in the emission factors for typical sources and the corresponding activity data. Uncertainty in the quantification methodology employed for emission inventories has been discussed in the literature [64,65], but less progress has been made in the case of China.

We take guidance from the IPCC in analyzing the estimation uncertainty, and use an error propagation equation to estimate the overall uncertainty. There are two convenient rules for combining uncorrelated uncertainties under addition and multiplication. When uncertain quantities are combined by addition, the standard deviation of the sum is the square root of the sum of the squares of the standard deviations of the quantities that are added with the standard deviations:

$$U_{\text{total}} = \frac{\sqrt{(u_1 \cdot x_1)^2 + (u_2 \cdot x_2)^2 + \dots + (u_n \cdot x_n)^2}}{x_1 + x_2 + \dots + x_n},$$

where U_{total} is the percentage uncertainty in the sum of the quantities and x_i and u_i are the uncertain quantities and the percentage uncertainties associated with them, respectively.

Table 4 Monthly emissions in China in 2007 (10⁴ t)

Species	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
PM _{2.5}	110.7	109.7	108.9	102.9	118.3	111.0	100.5	102.9	117.5	119.2	108.7	110.8
BC	12.3	12.1	11.8	10.2	12.6	11.3	9.8	10.1	12.5	13.2	11.7	12.2
OC	24.5	23.9	23.5	20.1	30.1	25.4	18.7	20.2	29.6	30.7	23.4	24.6
SO ₂	267.7	267.1	266.2	260.9	260.8	260.4	260.1	260.2	260.7	261.1	265.7	267.5
NO _x	191.6	191.1	190.7	189.2	202.4	195.1	188.2	189.8	201.8	202.9	190.6	191.4
CO	1348.0	1336.7	1317.3	1256.2	1559.9	1397.6	1218.9	1261.2	1545.9	1585.7	1315.6	1342.6
NH ₃	67.3	64.4	89.5	109.3	208.5	201.5	221.7	179.1	150.0	200.3	61.3	48.7
VOCs	284.4	284.4	284.4	284.0	322.2	302.7	284.0	288.6	321.1	321.9	284.3	284.4

When uncertain quantities are to be combined by multiplication, the same rule applies except that the standard deviations must all be expressed as fractions of the appropriate mean values:

$$U_{\text{total}} = \sqrt{U_1^2 + U_2^2 + \dots + U_n^2},$$

where U_{total} is the percentage uncertainty in the product of the quantities and U_i is the percentage uncertainties associated with the different quantities.

We have made the following assumptions when estimating the uncertainty of the emissions. (1) Statistics and activity data taken from the National Bureau of Statistics and other government agencies [52,55,56] are believed to be highly reliable and are assigned the lowest uncertainty value of 5%. (2) If emission factors are taken from quite limited sources, or a certain type of emission factor is unique, the uniform designed uncertainty is 200%. Streets assumed that the uncertainty in the emission factor was 5%–500% in his inventories [3], which was much higher than that in this work. Since his inventories were developed for the whole of Asia, the applicability of his emission factors seems to be less certain. As the level of activity data has been improved and more local experimental data for emission factors been taken into account, the uncertainties in emissions from power plants and industrial and transport sectors are relatively low, but the uncertainties in emissions from biomass combustion and residential sectors remain high.

The estimated overall uncertainties of emissions in this work are listed in Table 5. For comparison, the estimated uncertainties of Streets et al. [3] and Zhang et al. [12] are also listed. It is seen that less uncertainty was achieved in this work, especially for BC, OC and CO emissions. This is due to the difference in the assumed activity level and emission factors, particularly the lower uncertainty in emission factors as more local experimental data for emission factors were taken into account.

4 Conclusions

This paper presents detailed emission inventories for China in 2007 based on the latest fuel consumption data (including socioeconomic statistics and data on fossil and biomass fuels that were obtained from government agencies) and new emission factors obtained from local measurements. National and regional summaries of emissions were presented with $0.5^\circ \times 0.5^\circ$ resolution. The calculated total emissions in China were 13.212 Mt $\text{PM}_{2.5}$, 1.4 Mt BC, 2.946 Mt OC,

31.584 Mt SO_2 , 23.248 Mt NO_x , 16.017 Mt NH_3 , 164.856 Mt CO and 35.464 Mt VOCs. The dominant contributions to $\text{PM}_{2.5}$, BC, OC and CO emissions are from industrial and residential sectors, while coal-fired power generation is the largest contributor to SO_2 and NO_x emissions. NH_3 is primarily from poultry (livestock) and human waste disposal and agricultural production processes, such as chemical fertilizer use. VOCs are mainly derived from biomass combustion, industrial solid-waste incineration and other waste disposal processes, and solvents used in printing. The emission densities are higher in eastern and central China, with emissions concentrated in a curve around the agricultural and industrial heartland of China, from northeastern to eastern China, while areas of lowest emission are in western China and Inner Mongolia, which have lower rural population densities and levels of economic development.

Except for NH_3 and VOCs, the total emissions estimated here are roughly equal to those obtained in previous investigations, but they differ in terms of species and sector contributions. The uncertainties in the current inventories are found to be smaller than those of previous estimates. A strong seasonal dependence was observed for some species, mainly owing to seasonal changes in residential heating, the open burning of agriculture waste and fertilizer application.

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Table 5 Uncertainty in emission estimates ($\pm\%$)

Inventory	$\text{PM}_{2.5}$	BC	OC	SO_2	NO_x	CO	NH_3	VOCs
This work	70.3	76.2	66.6	12.0	33.4	36.8	36.6	51.6
Streets et al. [3]		360	450	16	37	185	72	130
Zhang et al. [12]	130	208	258	12	31	70		68

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