Scavenging of $PM_{2.5}$ by precipitation and the effects of precipitation pattern changes on health risks related to $PM_{2.5}$ in Tokyo, Japan

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ABSTRACT

Fine particulate matter (aerodynamic diameter <2.5 μm; $PM_{2.5}$) poses risks to human health. While precipitation is the main process for decreasing ambient pollutant concentrations, scavenging of $PM_{2.5}$ by precipitation remains to be investigated. Here we formulated the processes of $PM_{2.5}$ scavenging by precipitation from observed $PM_{2.5}$ concentrations ([$PM_{2.5}$]) and precipitation intensities. Then we analyzed how changes in precipitation patterns would affect health risks related to $PM_{2.5}$ on the basis of a Monte Carlo simulation. Tokyo, the capital of Japan, was selected as the target for this study because of its social significance. We found that [$PM_{2.5}$] decreased significantly through scavenging of $PM_{2.5}$ from the atmosphere by precipitation. In contrast, we found no significant correlation between reduction of [$PM_{2.5}$] and precipitation intensity. Our model for estimating the reduction of $PM_{2.5}$ and the Monte Carlo simulation showed good agreement with observations. Among various changes in potential precipitation patterns, changes in the arithmetic mean of the number of events and/or in precipitation duration were more influential on reduction of [$PM_{2.5}$] than changes in their standard deviations. Health risks due to $PM_{2.5}$ will increase with decreases in precipitation duration and occurrence.

Key words | cancer risk, climate change, particulate matter 2.5, precipitation

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INTRODUCTION

Among various risk factors, exposure to ambient particulate matter (PM) is of global concern for human health. Lim *et al.* (2012) estimated that 3.2 million people died in 2010 due to exposure to particles with an aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$). Because of the serious health risks, sources and environmental processes of $PM_{2.5}$ and the effectiveness of countermeasures are of interest (e.g., Harrison *et al.* 1997; Sanderson *et al.* 2013).

Precipitation decreases ambient pollutant concentrations through scavenging. Schumann *et al.* (1988) found that the scavenging coefficient of the ambient pollutant concentration depended on rainfall rate. Poissant & Béron (1994) confirmed an inverse relationship between ambient pollutant concentration and rainfall intensity, and found that concentration decreased notably during the initial stage of a rainfall event. Yoo *et al.* (2014) found that particles with an aerodynamic diameter smaller than 10 μm (PM_{10}) were scavenged more effectively than other ambient pollutants (SO_2, NO_2, CO, O_3). However, to the best of our knowledge, the scavenging of $PM_{2.5}$ by precipitation has not been investigated, with the exception of a few studies (Feng & Wang 2012; Ouyang *et al.* 2015) and therefore the scavenging process has not been well formulated. Although model-based studies for the projection of $PM_{2.5}$ transport and health risks have been carried out (e.g., Goto *et al.* 2014), the wet deposition of $PM_{2.5}$ is considered as sub-cloud scavenging, which lacks the optimization of parameters in the relationship between precipitation patterns and decreases in $PM_{2.5}$ concentration ([$PM_{2.5}$]). Therefore, the processes of $PM_{2.5}$ scavenging by precipitation should be investigated to quantify the effects of precipitation on health risks through the removal of $PM_{2.5}$ from the atmosphere. Furthermore, since climate change may cause changes in precipitation patterns (IPCC 2013), the effects of such changes on health risks related to $PM_{2.5}$ need to be understood.

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The aim of this paper is twofold: to analyze the relationship between [$PM_{2.5}$] and precipitation patterns and formulate the process of $PM_{2.5}$ scavenging by precipitation; and to calculate the effect of changes in precipitation patterns on health risks due to $PM_{2.5}$.

METHODS

Data and models

Tokyo, the capital of Japan, was selected as the target, because it has a large population (~13 million people) and the impact of $PM_{2.5}$ on human health is a social concern (Yorifuji *et al.* 2005). We obtained and used hourly-monitored $PM_{2.5}$ data (Bureau of Environment, Tokyo Metropolitan Government 2014; collected at 35.69°N, 139.77°E) and precipitation data (Japan Meteorological Agency 2014; collected at 35.69°N, 139.76°E) collected in Tokyo in 2012. Hourly data were used because of data availability. Characteristics of precipitation were described in Supplementary Material S1 (available online at http://www.iwaponline.com/wst/072/346.pdf). The physical distance between these two stations is about 1 km. Note that the $PM_{2.5}$ observations have missing values (184 of 8784, i.e., 2.1% of total), and these were excluded in the proceeding calculation. While this procedure may result in underestimation in calculating reduction of $PM_{2.5}$ (stated below), its effect is negligible.

To evaluate whether precipitation effectively removes $PM_{2.5}$ from the atmosphere, we estimate decreases in $PM_{2.5}$ per hour as $-\Delta C/\Delta t$, where ΔC is the change in [$PM_{2.5}$] and $\Delta t = 1$ hour. We then performed a one-sample two-tailed *t*-test to evaluate whether the rates of decrease were significantly higher than 0 during both dry and wet weather. Dry and wet hours are 8135 and 465 hours, respectively (the period when $PM_{2.5}$ data were missing was excluded).

While the precipitation scavenging process has been formulated by a theoretical approach (e.g., Beverland & Crowther (1992) modeled scavenging with the integrated cross sectional area of rain droplets), models with easily available data are preferable in terms of practical use. Schumann *et al.* (1988) proposed that the rate of decrease in ambient pollutant concentrations depends on rainfall intensity, particularly for large particles. On the other hand, Dickhut & Gustafson (1995) found that this relationship was weaker for particulate organic pollutants than for gaseous organic pollutants, and that precipitation intensity might be less important for removal of particles. Hence, we compared two models:

$$\text{Model 1:} \quad \frac{dC(t)}{dt} = -k \cdot C(t-1) \cdot P(t)$$

$$\text{Model 2:} \quad \frac{dC(t)}{dt} = -k \cdot C(t-1)$$

where $dC(t)/dt$ = decrease in [$PM_{2.5}$] between time $t-1$ and t, $C(t-1)$ = [$PM_{2.5}$] (μg/m^3) at time $t-1$, $P(t)$ = precipitation intensity (mm/h) at time t, and k is a parameter with values determined as explained below. Note that $C(t-1)$ and $P(t)$ are antecedent hourly monitored data at time $t-1$ and t, respectively. The reason for the difference in the time steps of [$PM_{2.5}$] and precipitation intensity is that decrease in [$PM_{2.5}$] at time t is influenced by [$PM_{2.5}$] at the previous time step and precipitation during the period of $t-1$ and t.

Model 1 assumes that the decrease in [$PM_{2.5}$] depends on both the concentration and the precipitation intensity at each time step. Model 2 assumes that it depends only on the concentration, which indicates that precipitation duration, rather than precipitation intensity, is influential in decreasing [$PM_{2.5}$]. Parameter k is optimized per precipitation event by fitting the above models and applying the least squares method (we also optimized k for all precipitation events. This result is available in the Supplementary Material S2, online at http://www.iwaponline.com/wst/072/346.pdf). To remove weak precipitation events, we focused on events lasting >2 hours. This allowed us to use plots with at least four points to obtain the parameter k to improve reliability of optimization. Note that the number of removed events with 1 and 2 hour precipitation is 118 (the number of all precipitation events is 191), their total amount of precipitation is 195 mm (12.5% of all precipitation events, 1561 mm), and their total time is 159 hours (22.5% of all precipitation events, 707 hours). Hence, 1 and 2 hour precipitation events are minor in terms of total precipitation and precipitation duration. Correlation coefficients between observed and estimated [$PM_{2.5}$] were calculated to evaluate the model performance, and we chose the better of the two models for further analyses.

Exposure to $PM_{2.5}$ was defined as the integration of concentration over time:

$$\text{Exposure to } PM_{2.5} = \int_0^{t_{end}} C(t)\, dt$$

where t_{end} is the end of a target year.

Reduction of $PM_{2.5}$ due to precipitation was defined as the total decrease only during the event (this conservative

Water Science & Technology | 72.8 | 2015

approach avoids overestimating the effects of scavenging):

$$\text{Reduction of } PM_{2.5} = \sum_{m=1}^{n} \int_0^{t_m} \{C_m - C(t)\}\, dt$$

where m = a particular precipitation event, n = the number of events in a target year, t_m = the duration of event m, and C_m = the initial [$PM_{2.5}$] just before precipitation event m starts.

Reduction of $PM_{2.5}$ was divided by the total number of hours in the year to calculate the yearly-average reduction. This reduction was considered as a positive effect of scavenging by precipitation on human health.

Estimation of [$PM_{2.5}$] under changes in various precipitation patterns

Two precipitation parameters were used to estimate the amount of $PM_{2.5}$ scavenging: the duration of each event and the number of events in each year. We formulated three types of simulations: observed values in 2012 ('2012sim'), observed values in 1976–2007 ('historical'), and scenarios of future precipitation pattern change. Hourly precipitation data in Tokyo in 1976–2007 were obtained from CD-ROM (the Japan Meteorological Agency).

To estimate exposure to $PM_{2.5}$ in scenarios of precipitation pattern change, we assumed changes of ±10% in both the arithmetic mean and the standard deviation (SD) of the distributions of the duration of each event and the number of events in a year. Climate change is likely to cause changes in the long-term mean (monthly or daily) precipitation and intensification of extreme events (IPCC 2013). However, how they will change is still unknown. Furthermore, in an urban setting such as Tokyo, other factors such as urbanization contribute to changes in precipitation pattern, so it is difficult to set reasonable assumptions. We therefore assumed various scenarios for the duration of each event and the number of events in a year (Table 1) and compared the results of all scenarios.

We conducted a Monte Carlo simulation by randomly choosing values of parameters in order to simulate the reduction of $PM_{2.5}$ and overall exposure to $PM_{2.5}$. Since we selected model 2 in the assessment (see 'Modeling the change in [$PM_{2.5}$] due to precipitation' in the Results and discussion section), three values were needed as inputs to the model: initial [$PM_{2.5}$], k, and precipitation duration. The initial [$PM_{2.5}$] was randomly selected from observation collected during dry weather in 2012. The duration was also randomly selected from the data assumed in each scenario. Since k and initial [$PM_{2.5}$] showed a weak but significant correlation (Spearman's rank test, $\rho = -0.24$, $P < 0.05$), we divided the distribution of k into four groups based on the median of initial [$PM_{2.5}$] and precipitation duration as thresholds, to enhance the independence of the values of the three parameters. Next, k was randomly selected according to the corresponding four groups, which was determined from the initial [$PM_{2.5}$] and duration. We then simulated $PM_{2.5}$ reduction due to one precipitation event 10,000 times and obtained the distribution of the simulated $PM_{2.5}$ reduction due to one precipitation event.

Next we estimated reductions of [$PM_{2.5}$] under the different scenarios. For each scenario we used the number of precipitation events in 1 year (N) and the 10,000-member dataset of $PM_{2.5}$ reduction for one event. The $PM_{2.5}$ reduction for one event was extracted N times from the 10,000-member dataset, and the reductions in a year were summed. This computation was also performed 10,000 times. The reduction of $PM_{2.5}$ in a year in each scenario was averaged over the year (divided by the total number of hours in the year). The annual mean [$PM_{2.5}$] in each scenario was then calculated by subtracting the median of the

Table 1 | Scenarios for precipitation change. Scenario 0 is the historical simulation (1976–2007) and the others are simulations. SD = standard deviation

	Number of events		Precipitation duration	
Scenarios	**Arithmetic mean (%)**	**SD (%)**	**Arithmetic mean (%)**	**SD (%)**
0	± 0	± 0	± 0	± 0
1-1	+10	+10	+10	+10
1-2	−10	−10	−10	−10
2-1	+10	+10	± 0	± 0
2-2	−10	−10	± 0	± 0
2-3	± 0	± 0	+10	+10
2-4	± 0	± 0	−10	−10
3-1	+10	± 0	+10	± 0
3-2	−10	± 0	−10	± 0
3-3	± 0	+10	± 0	+10
3-4	± 0	−10	± 0	−10
4-1	+10	± 0	± 0	± 0
4-2	−10	± 0	± 0	± 0
4-3	± 0	+10	± 0	± 0
4-4	± 0	−10	± 0	± 0
4-5	± 0	± 0	+10	± 0
4-6	± 0	± 0	−10	± 0
4-7	± 0	± 0	± 0	+10
4-8	± 0	± 0	± 0	−10

reduction of [$PM_{2.5}$] in a year in each scenario from the sum of the observed annual mean of [$PM_{2.5}$] and the observed reduction of [$PM_{2.5}$] in 2012.

Calculation of health risks

We estimated the health risks due to $PM_{2.5}$ using the method of Yorifuji *et al.* (2005). We did not consider the effect of location because [$PM_{2.5}$] was similar among fixed sites and indoor or outdoor residences (Michikawa *et al.* 2014). First, P_e was calculated as:

$$P_e = P_o/\{1 + [(RR - 1)(O - B)/10]\}$$

where P_e = the expected health outcome frequency at the reference exposure level B (6, 8, or 10 μg/m^3), P_o = the observed health outcome frequency (8.3 per 1000 people in Tokyo in 2012; Ministry of Health, Labour and Welfare 2012), RR = the relative risk of all-cause mortality associated with a 10 μg/m^3 increase in $PM_{2.5}$ (1.14; Lepeule *et al.* 2012: 1974–2009), and O = the observed [$PM_{2.5}$] (μg/m^3). Although RR depends on location, we adopted 1.14 because similar values were found in several cohort surveys (Lepeule *et al.* 2012). One in Japan reported $RR = 1.24$ for lung cancer mortality (Katanoda *et al.* 2011), but we did not use this value because the survey period was only 10 years. Since Lepeule *et al.* (2012) showed that the concentration–response relationship was linear down to [$PM_{2.5}$] = 8 μg/m^3, we used values of B similar to those reported previously (Lim *et al.* 2012: 5.8–8.8 μg/m^3).

The annual mortality risk due to $PM_{2.5}$ under each scenario was calculated as:

$$P_s = \frac{(E_s - B)(RR - 1)}{10} \cdot \frac{P_e}{1000}$$

where P_s = the annual mortality risk, and E_s = the estimated [$PM_{2.5}$] (μg/m^3).

RESULTS AND DISCUSSION

Modeling the change in [$PM_{2.5}$] due to precipitation

[$PM_{2.5}$] decreased after a precipitation event (Figure 1). Here we exemplify the result in July, when Japan has its rainy season, so that a decrease in [$PM_{2.5}$] can be clearly detected. It also fluctuated in the absence of precipitation because of variations in the outputs of $PM_{2.5}$ from biomass burning, industry, and motor vehicles (e.g., Song *et al.* 2006; Zhang *et al.* 2013).

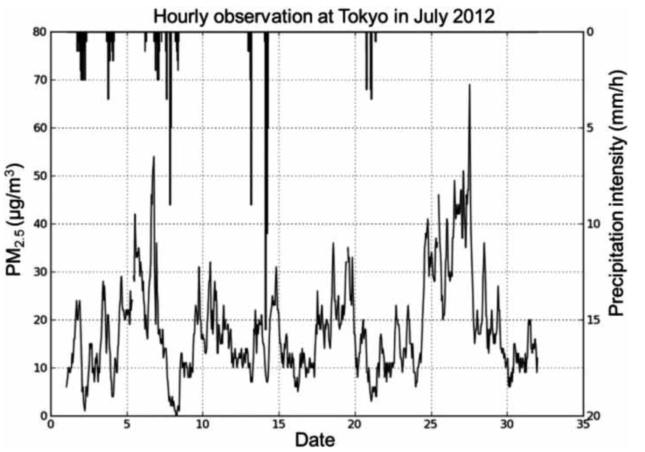

Figure 1 | Precipitation and [$PM_{2.5}$] in Tokyo in July 2012. Gaps represent missing data.

During dry weather (0 mm/h), the rate of decrease of [$PM_{2.5}$] did not deviate from zero (one-sample, two-tailed t-test, $P > 0.05$; Figure 2). However, during wet weather, [$PM_{2.5}$] decreased significantly ($P < 0.001$), similar to other studies (Feng & Wang 2012; Ouyang *et al.* 2015). This shows that precipitation scavenges $PM_{2.5}$ from the atmosphere, with potentially positive effects on human health.

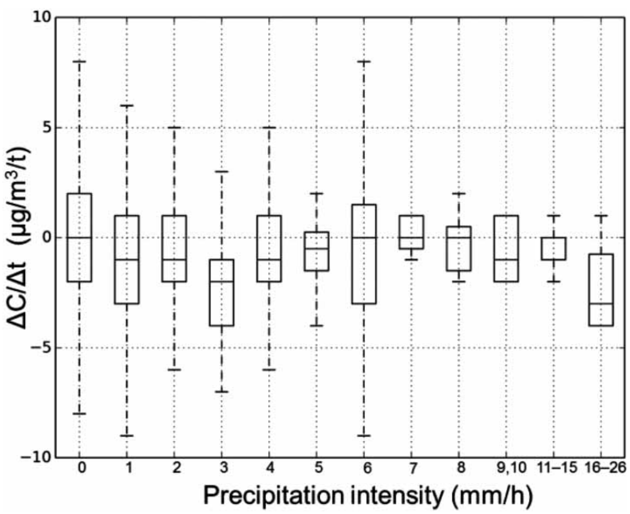

Figure 2 | Change in [$PM_{2.5}$] per hour by different precipitation intensities per hour. The black lines located in the middle of boxes indicate median values. Bottom of box, 25th percentile (Q1); horizontal line in middle of box, 50th percentile; top of box, 75th percentile (Q3). Minimum and maximum values represent Q1 − 1.5 (Q3 − Q1) and Q3 + 1.5 (Q3 − Q1), respectively. The numbers of precipitation for each intensity are: 8008 (0 mm/h), 141 (1 mm), 167 (2 mm/h), 33 (3 mm/h), 53 (4 mm/h), 12 (5 mm/h), 19 (6 mm/h), 7 (7 mm/h), 7 (8 mm/h), 10 (9, 10 mm/h), 10 (11–15 mm/h), 9 (16–26 mm/h). Note that the sum of these numbers (8476) is not equal to the number of available [$PM_{2.5}$] data (8784–184 = 8600). This is because one missing [$PM_{2.5}$] datum at time t makes it impossible to calculate decrease in [$PM_{2.5}$] due to precipitation at time t and $t + 1$, which cause the above discrepancy in the precipitation numbers.

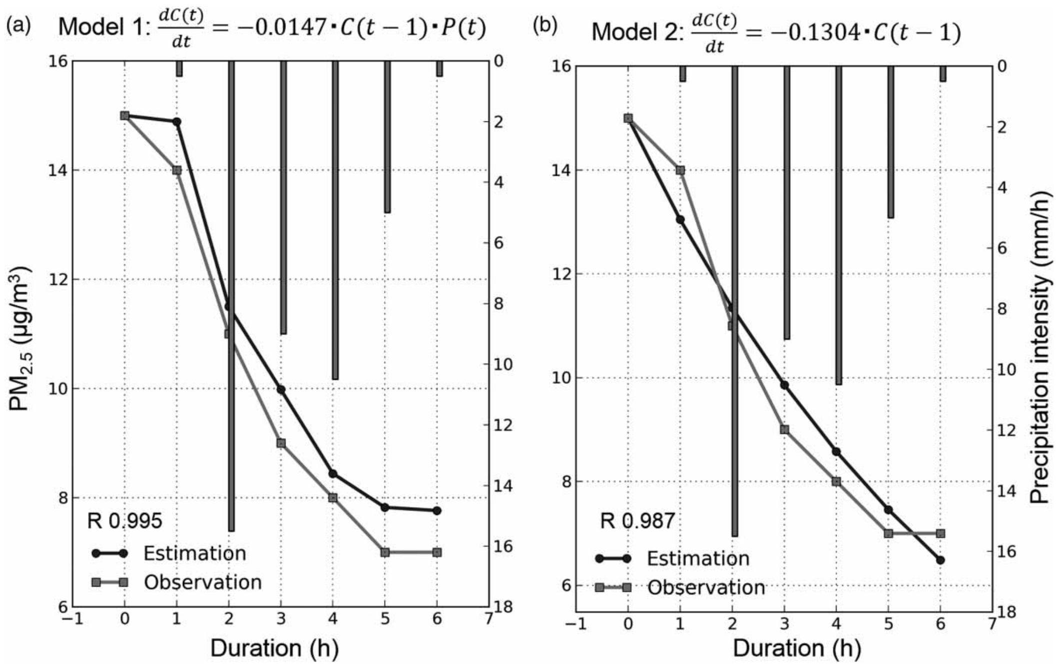

Figure 3 | Examples of the model calibration for changes in [$PM_{2.5}$] in the case of the precipitation event on 14 July 2012: (a) model 1, (b) model 2. Bars show precipitation.

Interestingly, there were no significant correlations between reduction of [$PM_{2.5}$] and precipitation intensity during wet weather (Spearman's rank test, $\rho = 0.003$, $P > 0.05$). The extent of decrease did not differ greatly among precipitation intensities (Figure 2). Note that the monitoring time step of 1 hour may have an unidentifiable effect on the analysis.

Figure 3 illustrates the results of model calibration using a precipitation event on 14 July 2012 as an example (this event

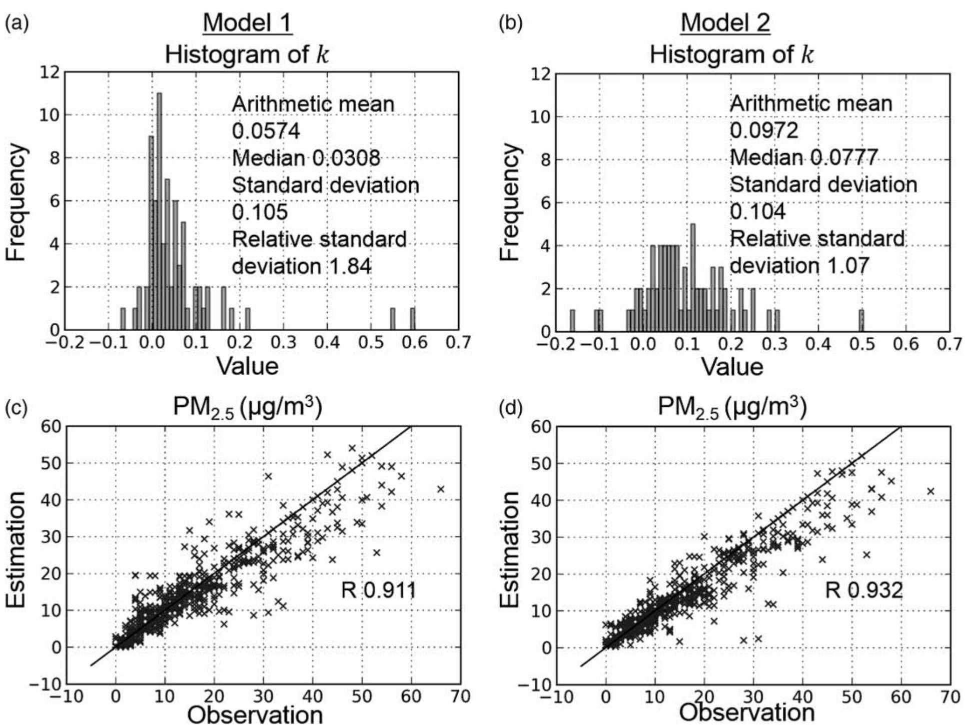

Figure 4 | Comparison between models of reproducibility of all precipitation events with >2 h precipitation in 2012. (a), (b) Distribution of *k* and (c), (d) correlation between observed and estimated [$PM_{2.5}$] by (a), (c) model 1 and (b), (d) model 2. Each plot indicates observed and estimated [$PM_{2.5}$] data per hour. *R* in each figure indicates correlation coefficient.

was taken from 73 precipitation events. The reason for the choice of this event is that its precipitation duration (6 hours) is the median of all precipitation events with >2 hours precipitation). Both models successfully ($R \geq 0.98$) represented the decrease of [$PM_{2.5}$] by precipitation. Figure 4 compares the reproducibility of estimated [$PM_{2.5}$] per hour between models. Statistical measures of parameter k, which was optimized for each event, gave a relative SD of 1.84 for model 1 and 1.07 for model 2 (Figure 4(a), (b)). The relative SD in model 2 was smaller than that in model 1. R was 0.911 for model 1 and 0.932 for model 2 (Figure 4(c), (d)), indicating that the inclusion of precipitation intensity did not improve the representation of $PM_{2.5}$ scavenging by precipitation. This result indicates that the duration of precipitation contributes more than intensity to decreasing [$PM_{2.5}$]. This is supported by the fact that the longer the precipitation duration, the larger the decrease in [$PM_{2.5}$] (see Supplementary Material S1 and Figure S3, available online at http://www.iwaponline.com/wst/072/346.pdf). Furthermore, model 2 is simpler than model 1 with regard to their parameters. Thus, we selected model 2 for the calculation of health risk. Even if we optimize k for all precipitation events, the correlation coefficient of model 2 was slightly larger than model 1 in accordance with results shown above (see Figure S4 in Supplementary Material S2, online at http://www.iwaponline.com/wst/072/346.pdf).

Estimation of [$PM_{2.5}$] under precipitation pattern changes and calculation of health risks

Observations in 2012 gave an annual mean [$PM_{2.5}$] of 14.70 μg/m^3 and a yearly-average reduction of 0.39 μg/m^3.

Table 2 | Estimated reduction of [$PM_{2.5}$], change relative to the baseline scenario, annual mean [$PM_{2.5}$], annual mortality risk due to $PM_{2.5}$, and change in risk relative to baseline scenario. See Table 1 for details in each scenario

	Yearly-average reduction of $PM_{2.5}$ concentration (μg/m^3)										
Scenarios	**5%**	**25%**	**50%**	**75%**	**95%**	**Arithmetic mean**	**Reduction of total $PM_{2.5}$ (%)[a]**	**Change in reduction relative to baseline (%)[b]**	**Annual mean concentration of $PM_{2.5}$ (μg/m^3)[c]**	**Annual mortality risks due to $PM_{2.5}$ (10^{-4})[d]**	**Increase of annual mortality risk to baseline (10^{-6})[b]**
2012sim	0.17	0.29	0.36	0.42	0.52	0.35	2.3	–	14.73	7.15 (5.16–9.05)	–
0	0.14	0.22	0.27	0.33	0.42	0.24	1.6	–	14.82	7.24 (5.25–9.13)	–
1-1	0.18	0.30	0.37	0.44	0.56	0.33	2.2	34.7	14.72	7.14 (5.15–9.03)	−10
1-2	0.12	0.17	0.21	0.26	0.33	0.21	1.4	−22.0	14.88	7.31 (5.32–9.19)	6.4
2-1	0.14	0.24	0.30	0.36	0.47	0.28	1.9	10.3	14.79	7.21 (5.22–9.10)	−3.0
2-2	0.13	0.20	0.25	0.30	0.39	0.24	1.6	−9.0	14.84	7.27 (5.28–9.16)	2.6
2-3	0.18	0.27	0.34	0.40	0.52	0.33	2.2	22.5	14.76	7.18 (5.18–9.07)	−6.5
2-4	0.12	0.19	0.24	0.29	0.37	0.23	1.5	−13.7	14.85	7.28 (5.29–9.17)	4.0
3-1	0.17	0.29	0.36	0.42	0.53	0.31	2.1	29.9	14.73	7.15 (5.16–9.05)	−8.7
3-2	0.10	0.17	0.21	0.26	0.35	0.19	1.3	−22.3	14.88	7.31 (5.32–9.20)	6.5
3-3	0.14	0.24	0.30	0.36	0.47	0.29	1.9	8.7	14.79	7.22 (5.23–9.11)	−2.5
3-4	0.14	0.22	0.27	0.32	0.41	0.26	1.7	−1.7	14.82	7.25 (5.26–9.14)	0.5
4-1	0.16	0.25	0.30	0.36	0.46	0.29	1.9	10.8	14.79	7.21 (5.22–9.10)	−3.1
4-2	0.12	0.20	0.25	0.30	0.39	0.21	1.4	−10.2	14.84	7.27 (5.28–9.16)	3.0
4-3	0.13	0.22	0.28	0.34	0.44	0.27	1.8	1.1	14.81	7.24 (5.25–9.13)	−0.3
4-4	0.13	0.23	0.28	0.34	0.43	0.27	1.8	2.7	14.81	7.23 (5.24–9.13)	−0.8
4-5	0.17	0.27	0.33	0.39	0.50	0.32	2.1	19.9	14.76	7.18 (5.19–9.08)	−5.8
4-6	0.11	0.19	0.24	0.29	0.38	0.22	1.5	−12.5	14.85	7.28 (5.29–9.17)	3.6
4-7	0.13	0.24	0.30	0.36	0.47	0.28	1.8	8.4	14.79	7.22 (5.23–9.11)	−2.4
4-8	0.14	0.22	0.27	0.32	0.41	0.26	1.8	−2.2	14.82	7.25 (5.26–9.14)	0.6

[a]Reduction of total possible exposure in the absence of precipitation. Arithmetic mean of $PM_{2.5}$ reduction for each scenario (14.70 + 0.39).
[b]Baseline is scenario 0 (historical).
[c]Median values were used to estimate annual mean concentrations.
[d]Values represent estimates from reference exposure level = 8 μg/m^3 (10–6 μg/m^3).

The reduction due to precipitation was 2.6% (=0.39/(14.70 + 0.39)) of total possible exposure in the absence of rain.

Table 2 summarizes the distribution of the yearly-average reduction of [$PM_{2.5}$], the change in reduction relative to the baseline scenario (Scenario 0–historical), and annual mean [$PM_{2.5}$] calculated from the Monte Carlo simulation. Scenario 2012sim gave a yearly average reduction of [$PM_{2.5}$] with a median of 0.36 μg/m^3 and an arithmetic mean of 0.35 μg/m^3, close to the observed value of 0.39 μg/m^3, indicating that model 2 and the Monte Carlo simulation successfully estimated the reduction of [$PM_{2.5}$].

Using precipitation data recorded in 1976–2007, we simulated historical reductions and prospective scenarios of precipitation patterns. While annual means of [$PM_{2.5}$] are not much different among scenarios (ranging between 14.7–14.9), changes in annual means of [$PM_{2.5}$] were not negligible among scenarios. Scenario 1-1 (mean and SD of the number of events and duration increase by 10%) increased scavenging by 34.7%. In contrast, Scenario 3-2 (mean of the number of events and duration decrease by 10%) reduced scavenging by 22.0%. Changes in the means of the number of events and/or duration influenced the reduction of [$PM_{2.5}$] more than changes in their SDs (Scenarios 3-1 vs 3-3, 3-2 vs 3-4, 4-1 vs 4-3, 4-2 vs 4-4, 4-5 vs 4-7, and 4-6 vs 4-8). Decreases in the number of events and in duration made similar contributions to the reduction of [$PM_{2.5}$] (Scenarios 3-2 vs 4-2 vs 4-6), whereas the increase in duration reduced [$PM_{2.5}$] more than the increase in the number of events (Scenarios 3-1 vs 4-1 vs 4-5). An increase in the SD of the number of events did not greatly affect the reduction of [$PM_{2.5}$] (Scenario 4-3), but increases in the SD of duration reduced [$PM_{2.5}$] substantially (Scenarios 3-3 and 4-7).

Table 2 also shows the estimated annual mortality risks due to $PM_{2.5}$ and changes in risk relative to the baseline scenario. The annual mortality risk in 2012sim was estimated as 7.15×10^{-4}, comparable to preventable risks due to $PM_{2.5}$ in another study (Yorifuji *et al.* 2005). As pointed out before (e.g., Yorifuji *et al.* 2005; Katanoda *et al.* 2011; Lepeule *et al.* 2012; Lim *et al.* 2012; Sanderson *et al.* 2013), the health risks due to exposure to $PM_{2.5}$ are serious. Scenario 3-2 (10% decrease in means) and 1-2 (10% decrease in means and SDs) gave the greatest increase in annual mortality risk (6.5×10^{-6} and 6.4×10^{-6}) in comparison with the baseline scenario. Scenario 4-2 and 4-6 (10% decrease in means of either the number of events or precipitation duration) gave relatively large increase (3.0×10^{-6} and 3.6×10^{-6}). Thus, decreases in the arithmetic means of the number of events and/or duration may increase the health risks due to $PM_{2.5}$.

CONCLUSIONS

We confirmed statistically that precipitation reduces [$PM_{2.5}$] in the atmosphere. Our formulation of [$PM_{2.5}$] decrease gave results consistent with observations. The decrease depended more on the precipitation duration than on intensity. Among various scenarios of precipitation pattern changes, 10% decreases in the arithmetic mean of the number of events and in duration increased the annual mortality risk by 6.5×10^{-6}.

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