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Development of human health damage factors for PM$_{2.5}$ based on a global chemical transport model

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Abstract

Purpose Health damage from ambient fine particulate matter (PM$_{2.5}$) shows large regional variations and can have an impact on a global scale due to its transboundary movement. However, existing damage factors (DFs) for human health in life cycle assessments (LCA) are calculated only for a few limited regions based on various regional chemical transport models (CTMs). The aim of this research is to estimate the human health DFs of PM$_{2.5}$ originating from ten different regions of the world by using one global CTM.

Methods The DFs express changes in worldwide disability-adjusted life years (DALYs) due to unit emission of black carbon and organic carbon (BCOC), nitrogen oxides (NO$_x$),

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and sulfur dioxide (SO$_2$). DFs for ten regions were calculated as follows. Firstly, we divided the whole world into ten regions. With a global CTM (MIROC-ESM-CHEM), we estimated the concentration change of PM$_{2.5}$ on the world caused by changes in the emission of a targeted precursor substance from a specific region. Secondly, we used population data and epidemiological concentration response functions (CRFs) of mortality and morbidity to estimate changes in the word's DALYs occurring due to changes in the concentration of PM$_{2.5}$. Finally, the above calculations were done for all ten regions.

Results and discussion DFs of BCOC, NO$_x$, and SO$_2$ for ten regions were estimated. The range of DFs could be up to one order of magnitude among the ten regions in each of the target substances. While population density was an important parameter, variation in transport of PM$_{2.5}$ on a continental level occurring due to different emission regions was found to have a significant influence on DFs. Especially for regions of Europe, Russia, and the Middle East, the amount of damage which occurred outside of the emitted region was estimated at a quarter, a quarter, and a third of their DFs, respectively. It was disclosed that the DFs will be underestimated if the transboundary of PM$_{2.5}$ is not taken into account in those regions.

Conclusions The human health damage factors of PM$_{2.5}$ produced by BCOC, NO$_x$, and SO$_2$ are estimated for ten regions by using one global chemical transport model. It became clear that the variation of transport for PM$_{2.5}$ on a continental level greatly influences the regionality in DFs. For further research to quantify regional differences, it is important to consider the regional values of concentration response function (CRF) and DALY loss per case of disease or death.

Keywords Disability-adjusted life years · Global chemical transport model · Human health damage · Life cycle impact assessment · Outdoor air pollution · PM$_{2.5}$

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1 Introduction

Transboundary air pollution first received significant attention through the issue of acid rain in Europe and North America during the 1970s. Since then, a region-wide monitoring network has been developed, which provides scientific data to diagnose the status of acid deposition. Additionally, integrated assessment models for acid deposition, such as the Regional Acidification Information and Simulation (RAINS) model (Alcamo et al. 1990), have been developed and used to support the policymaking processes. Based on these scientific efforts and their derived knowledge, the Convention on Long-Range Transboundary Air Pollution (CLRTAP) was concluded in 1979 by European and North American countries. Under the CLRTAP, the scope of the convention has been widened to address the ground-level ozone, persistent organic pollutants (POPs), heavy metals, and particulate matter (PM). The parties of the convention have been obliged to take preventive measures against transboundary air pollution, such as emission control. Because of these measures, many countries in Europe have achieved reductions in air pollutant emissions, and a decline in the concentrations or depositions of several air pollutants has also been observed and confirmed (Barrett et al. 2000; Schulz et al. 2013). However, even in these regions, the issue of transboundary air pollution has not been fully resolved. Recently, in addition to transboundary air pollution within continents, the transport of air pollutants on an inter-continental or hemispheric scale has gained much attention. In December 2004, the Task Force on Hemispheric Transport of Air Pollution (TF HTAP), organized by CLRTAP, was launched to attain a better understanding of hemispheric transport (Dentener et al. 2010). It issued a scientific assessment report that showed the significance of hemispheric transport of air pollutants between four regions in the world (Europe, South Asia, East Asia, and North America). This was estimated from the results of multiple global chemical transport model (CTM) calculations. For instance, the report showed that a decrease in inter-continental transport from emissions that decreases in the other three regions would lead to a 5 to 20 % reduction in the annual mean ground-level PM concentration averaged in one region. This would be because of a 20 % decrease in emissions in all four regions. However, efforts to estimate the impact of air pollution on human health have so far not sufficiently considered the issue of inter-continental scale transport of air pollutants.

In terms of a life cycle impact assessment (LCIA), recently, several new globally scaled and spatially explicit assessment methods such as IMPACT World + (2012) and LC-IMPACT (Huijbregts et al. 2013) have been put forward. The establishment of this type of method is expected to contribute to an assessment of products in the worldwide supply chain. Therefore, it is necessary to develop impact factors which

concerned different environmental conditions on regional or global scale. For the case of health damage due to PMs, the results of CTMs suggest that the health damage could occur not only in higher concentrate area near the source point but also in regions with a lower concentrate, which are far from the emission region due to long-range transport of PMs. Furthermore, because it is difficult to differentiate emission area on a small scale through a life cycle of a product, thus, development of damage factors considering transboundary on a larger regional scale is effective.

In life cycle assessment (LCA), the human health damage factors (DFs) of air pollution are expressed as the amount of disability-adjusted life years (DALYs) occurring due to emitted units of air pollutant. They can be estimated by a fate and exposure analysis using atmospheric dispersion and transport models, as well as an effect analysis based on epidemiologic studies (Hofstetter 1998; Van Zelm et al. 2008; Itsubo and Inaba 2010). While almost all existing researches tend to estimate health effects by adopting cohort studies of long-term exposure, huge variation was found with fate analysis step.

Van Zelm et al. (2008) provided DFs for Europe at a high resolution level (50 km × 50 km) by using an atmospheric fate model EUTREND (Van Jaarsveld et al. 1997). This combines a Gaussian plume model, which describes short-range local transport and dispersion, with a Lagrangian trajectory model, which describes long-range transport. However, health damages outside of Europe caused by pollutants emitted in Europe were ignored and only PM_{10} was considered. Krewitt et al. (2001) estimated DFs on a country level in Europe based on the EcoSense model which contains a Gaussian plume model (short range) and a windrose trajectory model (long-range) for fate analysis. They also calculated DFs for Asia and South America based on the same model and indicated that population density was the most important parameter that affected the difference between targeted continental regions. However, the inter-continental transport of air pollution was not considered because only the health damage in targeted emission areas was aggregated for the calculation of DFs.

Humbert et al. (2009) developed a spatially resolved multimedia, multi-pathway, fate, exposure, and effect model for North America called IMPACT North America. It can differentiate variations in intake fractions ($mass_{intake}/mass_{emitted}$) on the direct surroundings (indoor or outdoor), a local scale (urban or rural), and a regional scale (North America). However, it only focuses on primary air pollutants and does not consider the secondary air pollution like PM produced by SO_2 or NO_x. Greco et al. (2007) estimated the intake fractions from exhaust emissions in the USA based on a source to receptor matrix. This is a regression-based derivation of output from the Climatologic Regional Dispersion Model (CRDM). The intake fractions covered both primary and secondary $PM_{2.5}$. They indicated that the transport distance

of sulfate was longer than that of nitrate and primary particles, but did not consider transport on a continental level. Itsubo and Inaba (2010) provide DFs for six regions in Japan. The fate analysis was conduct by a simple plume and puff model for primary pollutants. A source-receptor matrix based on the results of the OPU model (Ikeda 2001) was adopted for secondary pollutants.

Based on the above, recent researches such as those by Van Zelm et al. (2008) developed damage factors based on a higher resolution measurement compared to earlier researches like Hofstetter (1998). As the result, influences of stack height and different emission area (city centers or rural areas) on DFs were disclosed. Furthermore, health damage that has a strong correlation between emission strength and population density were found as a key finding. On the other hand, only DFs for limited regions (Europe, America, and Japan) were estimated, and no DFs were estimated by considering inter-continental transport of air pollution. Furthermore, no research defines a quantitative relationship between the source regions and receptor regions. Thus, the aim of the research is to estimate the human health damage factors of fine particulate matter (PM$_{2.5}$) in ten regions of the world by using a global chemical transport model. This research also attempted to disclose the effect on DFs from transboundary air pollution.

2 Methods

2.1 Calculation procedure

The DFs for human health damage (DALY/kg) caused by emitted substance p from region r are defined as the yearly marginal change in worldwide DALYs occurring due to marginal changes in the emission of substance p from region r. Health damage occurring due to both primary and secondary PM$_{2.5}$ aerosols are considered. In this study, black carbon (BC) and organic carbon (OC) from primary sources are considered and treated as a group (hereinafter "BCOC"). Secondary formed OC from atmospheric hydrocarbon species are not considered because of the inability to treat it in the global CTM used. Secondary formed nitrate (NO$_3^-$) and sulfate (SO$_4^{2-}$) PM$_{2.5}$ from the oxidation of nitrogen oxides (NO$_x$) and sulfur dioxide (SO$_2$) in the atmosphere are considered.

To express the inter-continental transport of air pollutants, whole continental areas are separated into ten regions, except Polar Regions (Fig. 1). This is done by considering the resolution of adopted global CTM and the availability of data used. Japan is treated independently from the East Asian region (R8) because of the differences in the levels of economic development and emission rate, even though the resolution may not be fit for Japan as one region.

The procedures used to estimate DFs are composed of three steps, as follows:

- Fate analysis: calculates mass concentration changes of PM$_{2.5}$ in the whole world that occur due to additional unit emission of substance p from region r.
- Effect analysis: calculates the increment of mortality and morbidity in the world per unit of PM$_{2.5}$ concentration.
- Damage analysis: calculates the health damage (DALY) with the effect analysis results. This is done by using population density data and DALY loss per case of disease and death.

As an equation, the results are summed up by mortality and morbidity, as shown below:

$$DF_{r,p} = D_{r,p,\text{mortality}} + D_{r,p,\text{morbidity}} \tag{1}$$

$$D_{r,p,\text{mortality}} = \sum_i \left(\Delta C_{p,r,i} \times R \times M_c \times P_i \times S_w \right) \tag{2}$$

$$D_{r,p,morbidity} = \sum_i \sum_d \left(\Delta C_{p,r,i} \times R_d \times P_{d,i} \times S_{d,w} \right) \tag{3}$$

DF$_{r,p}$	Damage factor of substance p emitted from region r (DALY/kg).
D$_{r,p,\text{mortality}}$	Health damage of mortality occurring due to the unit of substance p emitted from region r (DALY/kg).
D$_{r,p,\text{morbidity}}$	Health damage of morbidity occurring due to the unit of substance p emitted from region r (DALY/kg).
$\Delta C_{p,r,i}$	Yearly average concentration of PM$_{2.5}$ produced by substance p and emitted from region r in grid cell i ((μg/m^3)/kg).
R	Increase rate of mortality occurring due to rises in the concentration of PM$_{2.5}$ (%/(μg/m^3)).
M$_c$	Baseline all-cause mortality rate of country c (case/cap).
P$_i$	Adult population in grid cell i (cap).
S$_w$	DALY per case of death in 14 WHO sub-regions (DALY/case).
R$_d$	Increase rate of morbidity occurring due to rises in the concentration of PM$_{2.5}$ for each of disease d ((case/cap)/(μg/m^3)).
P$_{d,i}$	Population in grid cell i for each disease d (cap).
S$_{d,w}$	DALY per case of disease in 14 WHO sub-regions for each disease d (DALY/case).

Fig. 1 Boundary of ten regions considering in this research
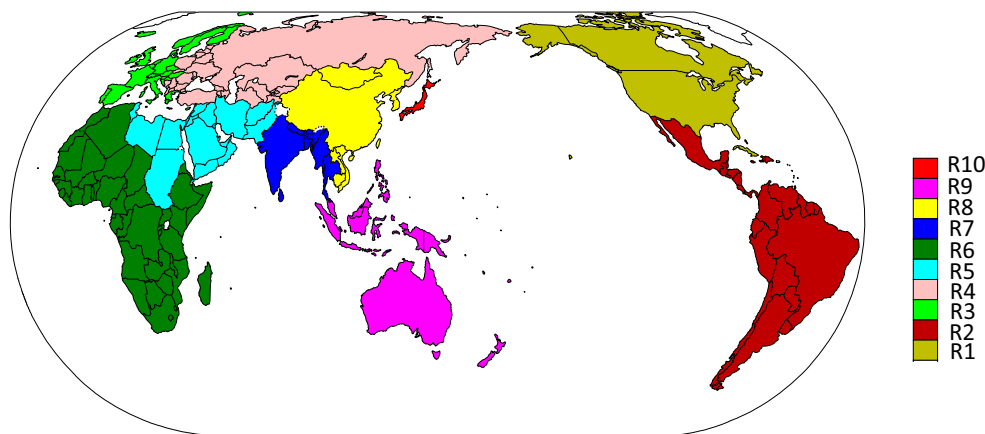
2.2 Fate analysis

For fate analysis of $PM_{2.5}$ on a global scale, we used a global CTM, MIROC-ESM-CHEM (Watanabe et al. 2011). MIROC-ESM-CHEM can calculate the global distributions of tropospheric aerosols and gas-phase species. It is based on the procedures adopted from a tropospheric aerosol transport model, SPRINTARS (Takemura et al. 2005), and a CTM for tropospheric gas-phase species, CHASER (Sudo et al. 2002). This represents key processes for the fate of air pollutants, including emission, transport, gas and liquid phase chemical reactions, wet and dry deposition, and gravitational settling. The model covers the $PM_{2.5}$ species of BC, OC, NO_3^-, SO_4^{2-}, soil dust, and sea salt. The first four of these species, which have large anthropogenic sources, were used in further analyses. The horizontal grid spacing used in the current study (ca. 2.8° by 2.8° in longitude and latitude) is somewhat coarse compared to what is used in the published air pollution studies addressing regional or sub-regional scale issues in particular. Several studies (e.g., Qian et al. 2010, Gustafson et al. 2011, and Stroud et al. 2011) have reported that the coarse horizontal resolution can underestimate the concentrations of PMs mainly due to insufficient representation of the fine structure in local emissions, topography, or flow patterns. They have shown good ability of the models with fine horizontal resolution of around 3 km to represent the observed horizontal and temporal fluctuations of air pollutants. However, these studies only provided the cases for limited small areas near the source of PMs, and for limited periods, the scale-induced differences in the concentrations of PMs far from the source areas have not yet been examined well. Moreover, since the purpose of the current study is to estimate the impact of hemispheric transport of PMs on human health, we absolutely required a bunch of calculations with global scale CTM which cannot have such a fine horizontal resolution as ca. 3 km because of its high computational costs. The emissions of primary $PM_{2.5}$ (BC and OC in the current study) and the precursors of

secondary $PM_{2.5}$ in the year 2000 from a historical (1850–2000) emission dataset compiled by Lamarque et al. (2010) were selected for calculation.

In order to estimate changes in the mass concentration of targeted $PM_{2.5}$ species that occurs due to additional unit emission (kg) of it (or its precursors) in one region in Fig. 1, we needed a set of modeled calculations, with and without an increase in emissions of that $PM_{2.5}$ (or its precursors) in that region. The difference between the calculated mass concentrations of the $PM_{2.5}$ from the two calculations was then divided by the emission increments. For this, we carried out one simulation without additional emission increment in any regions in Fig. 1 (standard simulation). We also individually carried out simulations with a 20 % increase in emissions for every targeted $PM_{2.5}$ species per region in Fig. 1 (sensitivity simulations). A 20 % increment of emissions in source regions was adapted from the procedure employed in the assessment report of TF HTAP (Dentener et al. 2010).

2.3 Effect analysis

The increments of worldwide mortality and morbidity occurring due to changes in the concentration of $PM_{2.5}$ were calculated by using the concentration response function (CRF) estimated by epidemiologic studies (Table 1). For chronic mortality caused by long-term exposure of $PM_{2.5}$, the CRF that is estimated by cohort research and based on data from the American Cancer Society (ACS) is the most reliable (Krewski et al. 2000; Pope et al. 1995, 2002). The value from Krewski et al. (2009), the most recent cohort study based on the database, was adopted in this research for all regions and linearity was assumed without a threshold. For morbidities that occurred due to exposure of $PM_{2.5}$ and PM_{10}, only the items recommended by the ExternE update report (Bickel et al. 2005) were used. The CRFs for PM_{10} were converted for $PM_{2.5}$ by applying 0.6 as a ratio of $PM_{10}/PM_{2.5}$. This is a typical ratio value for the concentration ratios of PM_{10} and $PM_{2.5}$ (Bickel et al. 2005).

Table 1 Concentration response function of each disease

Disease[a]	Affected age groups	Concentration response function %/(μg/m³)	Concentration response function (case/cap)/(μg/m³)	Size of particulate matter[b]
Chronic mortality	Adult only	7.5×10^{-1}	–	$PM_{2.5}$
Chronic bronchitis	Adult only	–	2.7×10^{-5}	PM_{10}
Medication usage by people with asthma	4.5 % of adult	–	9.1×10^{-2}	PM_{10}
	20 % of child	–	1.8×10^{-2}	PM_{10}
Lower respiratory symptoms	30 % of child	–	1.3×10^{-1}	PM_{10}
	Child	–	1.9×10^{-1}	PM_{10}
Restricted activity days	Adult only	–	9.0×10^{-2}	$PM_{2.5}$
Respiratory hospital admissions	All ages	–	7.0×10^{-6}	PM_{10}

[a] The CRF of chronic mortality is collected from Krewski et al. (2009) and others are from ExternE (2005)

[b] The ratio of CRF slopes $PM_{10}/PM_{2.5}$ which should be taken as 0.6 is used to change the CRF from PM_{10} to $PM_{2.5}$

CRFs of mortality and morbidity were both assumed with no regional variation in this research. For chronic mortality, there are few cohort studies published in developing countries. This is because of its long and costly investigation period. There have been interesting morbidity studies found recently that indicate variability between some regions. However, a result of discussions carried out in the update report of ExternE showed that it was not sufficient enough to allow for generalizations about other regions.

2.4 Damage analysis

Data of population and DALY per case of disease and death were collected. DALYs were then calculated as health damage from the results of the effect analysis. Gridded population data on a 2.8° grid size compiled from 0.25° grid data which is provided by Gridded Population of the World Version 3 in the year 2000 (CIESIN et al. 2005) was used. While the DALY per case of disease and death may have variation among regions, very few data are available. Hofstetter (1998) provides a rough estimation of years of life lost (YLL) per case and years lost occurring due to disability (YLD) per case for targeted items of mortality and morbidity. The estimation is based on literature mainly published in Europe. On the other hand, the World Health Organization (2008) provides the values of YLD and numbers of case only for very limited diseases, like asthma. However, these were done for each of the 14 WHO sub-regions. In order to estimate the DALY per case on a regional level, we assumed that the ratio of YLD per case for asthma obtained between Hofstetter (1998) and each WHO sub-region could be applied to all the targeted diseases and death. Table 2 shows the rough estimated DALY per case for each disease and death in 14 regions. It also indicates that regions with lower economic growth have a high value.

3 Results

3.1 Concentration change

Figure 2 showed the global changes in annual average surface mass concentration for sulfate $PM_{2.5}$ per unit emission of SO_2 from every source region in Fig. 1. In general, the affected spheres of emission increments in one source region extend leeward of the prevailing wind around the region. For instance, the emissions of SO_2 in Central and South America (R2) or Africa (R6) have apparent influence on the low-latitude maritime area west of these source regions because of the trade winds. Moreover, a remarkable tendency of sulfate transportation affected by westerly winds was found for source regions located in the mid-latitudes of both hemispheres. In particular, SO_2 emissions in the regions of North America (R1), Europe (R3), Russia (R4), China (R8), and Japan (R10) caused increases in sulfate $PM_{2.5}$ in their downwind areas. Although it is not visible in the annual average concentrations in Fig. 2, there were large seasonal differences in the shape and range of the affected spheres for source regions that are strongly influenced by the monsoonal circulation, such as India (R7) and Oceania (R9).

The results of concentration change including which produced from additional NO_x and BCOC emissions (see Fig. S1, Fig. S2 in the Electronic Supplementary Material) show the differences in the range of affected spheres for each $PM_{2.5}$ species. In general, the shapes of the affected sphere are similar to each other; however, the range of it for nitrate $PM_{2.5}$ is apparently narrower than those for the others. This is mainly due to the difference in the residence time of each $PM_{2.5}$ species in the atmosphere. Nitric acid (HNO_3), precursor of nitrate $PM_{2.5}$, is highly soluble in cloud droplet and readily wet deposited from the atmosphere which results in short residence time and small existing area around NO_x source region both for HNO_3 and nitrate $PM_{2.5}$.

Table 2 Rough-estimated DALY per case for each disease on 14 regions

Unit**		Disease considered in this study						
		Asthma	Chronic mortality	Respiratory hospital admissions	Chronic bronchitis	Lower respiratory symptoms	Medication usage by people with asthma	Restricted activity days
		YLD (0,0)/case	YLD (0,0)/case	YLD (0,0)/case	YLD (0,0)/case	YLD (0,0)/case	YLD (0,0)/case	YLD (0,0)/case
Hofstetter (1998)		*0.03**	*6.6**	*0.01**	*2.0**	*0.0001**	*0.0003**	*0.0003**
	AFR-D	0.07*	13.4	0.03	5.4	0.0004	0.0007	0.0007
	AFR-E	0.05*	9.9	0.02	4.0	0.0003	0.0005	0.0005
	AMR-A	0.04*	7.3	0.02	3.0	0.0002	0.0004	0.0004
	AMR-B	0.05*	10.1	0.02	4.1	0.0003	0.0006	0.0006
	AMR-D	0.05*	10.8	0.02	4.4	0.0003	0.0006	0.0006
	EMR-B	0.06*	11.2	0.02	4.5	0.0003	0.0006	0.0006
WHO (2008)	EMR-D	0.06*	12.3	0.03	5.0	0.0003	0.0007	0.0007
	EUR-A	0.03*	6.6	0.01	2.7	0.0002	0.0004	0.0004
	EUR-B	0.05*	9.0	0.02	3.6	0.0003	0.0005	0.0005
	EUR-C	0.04*	7.0	0.02	2.8	0.0002	0.0004	0.0004
	SEAR-B	0.05*	10.4	0.02	4.2	0.0003	0.0006	0.0006
	SEAR-D	0.06*	11.5	0.03	4.7	0.0003	0.0006	0.0006
	WPR-A	0.03*	6.6	0.01	2.7	0.0002	0.0004	0.0004
	WPR-B	0.05*	9.4	0.02	3.8	0.0003	0.0005	0.0005

*Italic values are collected from Hofstetter (1998) and WHO (2008)

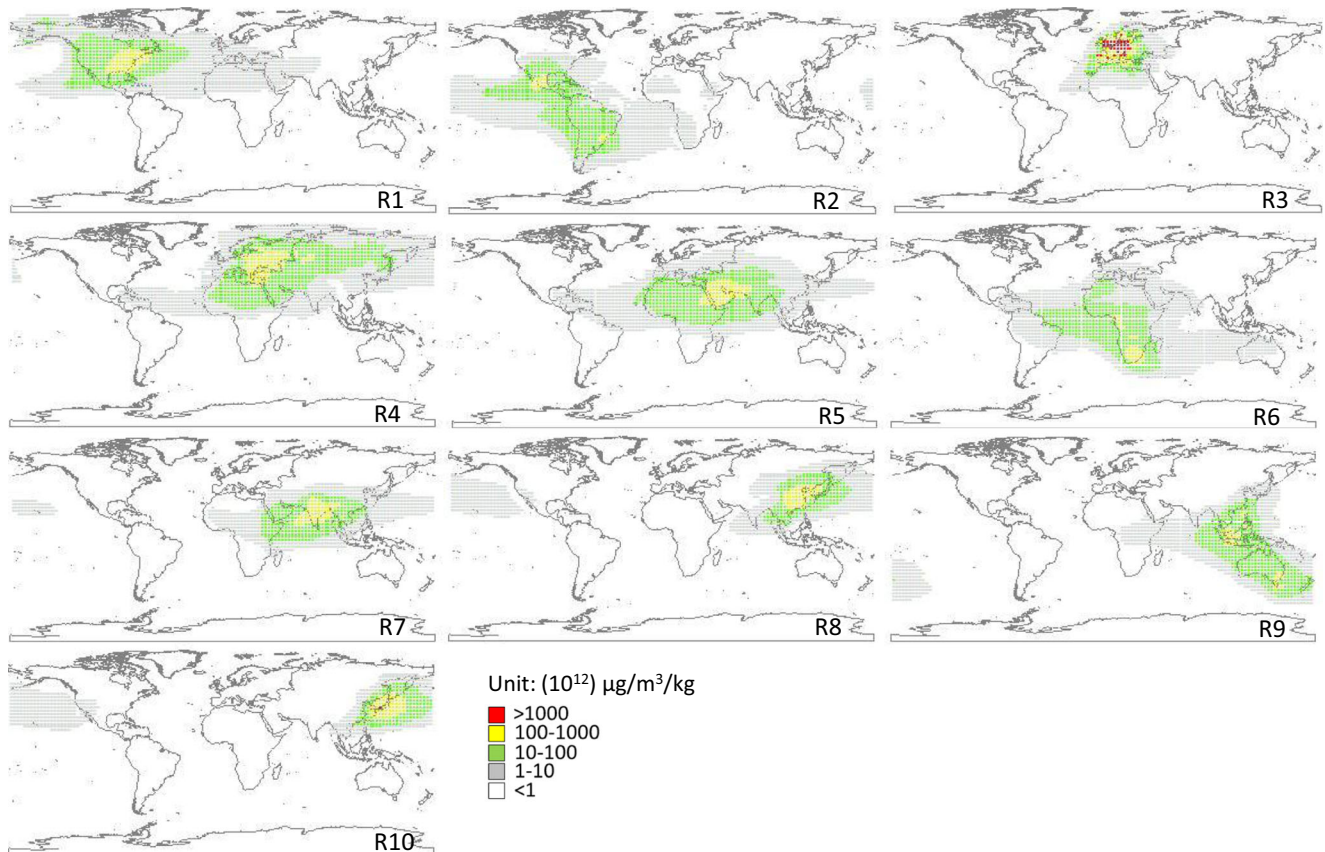**(0,0) stands for a calculation without discounting and no age weighting

Fig. 2 Annual average concentration of PM2.5 (sulfate) per kilogram emission of SO_2 for ten regions in the year 2000

Unit: (10^{12}) µg/m³/kg
- >1000
- 100–1000
- 10–100
- 1–10
- <1

3.2 Damage factors

DFs for three substances with its breakdown of diseases are shown in Fig. 3. SO_2 and NO_x showed similar values ranged $2.7 \times 10^{-5} \sim 2.2 \times 10^{-4}$ (DALY/kg) and $1.2 \times 10^{-5} \sim 3.3 \times 10^{-4}$ (DALY/kg), while BCOC resulted $7.8 \times 10^{-5} \sim 1.6 \times 10^{-3}$ (DALY/kg). The DFs of BCOC are around five times larger than those of NO_x and SO_2. This is mainly because the concentration change in primary $PM_{2.5}$, resulted from BCOC, was estimated to be larger than that of the secondary $PM_{2.5}$ produced by NO_x and SO_2. For details of diseases, it was clear that chronic mortality damage was the largest. However, morbidity damage was not small. In particular, the damage of chronic bronchitis, the highest of the morbidities, was estimated at around a quarter than that of chronic mortality.

Regional differences of DFs are also shown in Fig. 3. The ranges of DFs for ten regions were estimated around one order for all three substances. Regions of India (R7) and China (R8) showed the biggest values of damage. This is because of their large amount of exposure population and high values of DALY per case of disease and death, even though the increments of concentration per kg emission are not high. Damages on regions of Europe (R3) and Japan (R10) were the second largest mainly due to their high increments of concentration. Those of other regions were small, except the Middle East region (R5).

Furthermore, the difference in the ranges of transportation among substances also impacted on DFs. Figure 4 shows the percentage of damages for SO_2, NO_x, and BCOC caused by one source region that affected the ten regions. For most regions, SO_2 showed that the largest share of damage occurred outside of the emission region because of its broad range of transportation. However, because the emission location is close to other regions, it is possible that, for limited regions such as Russia (R3) and Africa (R6), the share of damage outside of BCOC and NO_x is larger than that of SO_2. For all three substances, the regions of Europe (R3), Russia (R4), and the Middle East (R5) showed the higher share of damage occurred in their downwind areas than that of other regions. Especially, the Middle East region had the highest share of damage in the India region because of its high population density and DALY per case. The results indicated that, if the transboundary effects of $PM_{2.5}$ are ignored, around 25~60 % of DF may be underestimated.

4 Discussion

4.1 Fate analysis

The DFs estimated in this study largely relied on the results of the fate analysis. Therefore, it is important to observe and evaluate how representative modeled concentrations and temporal variations of $PM_{2.5}$ are. The tropospheric aerosol transport model SPRINTARS, which uses the same procedure of aerosol calculation as our model, has been evaluated as having the ability to represent the observed features of atmospheric aerosols through comparisons with ground-based or satellite observations (Takemura et al. 2003; Goto et al. 2011). These
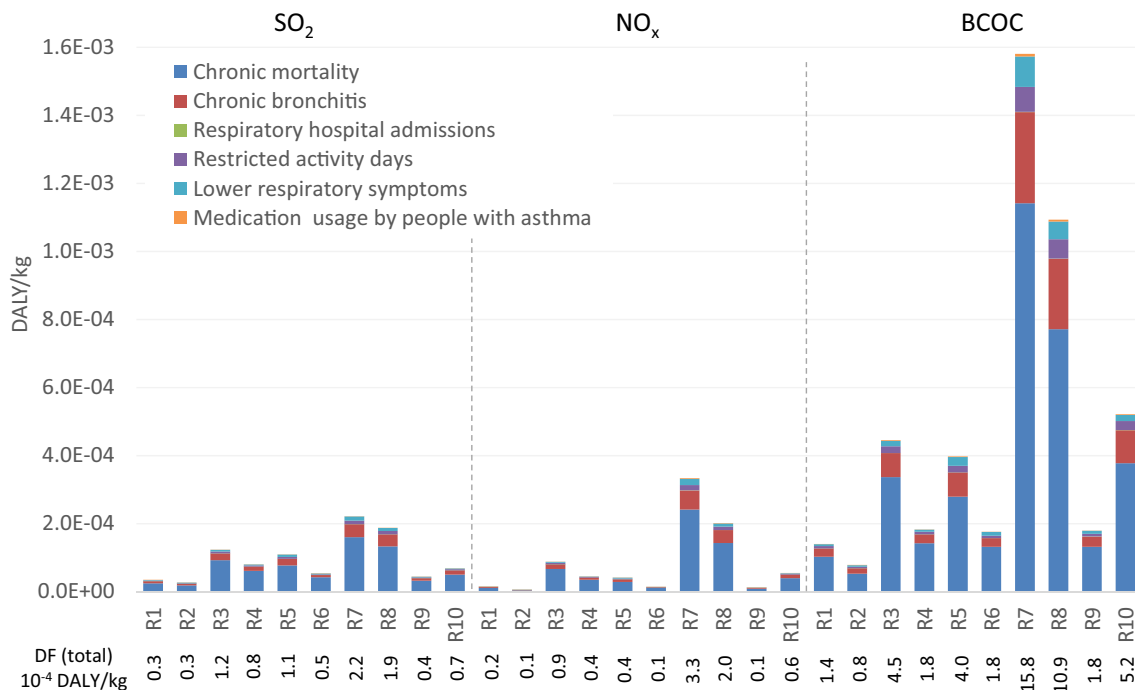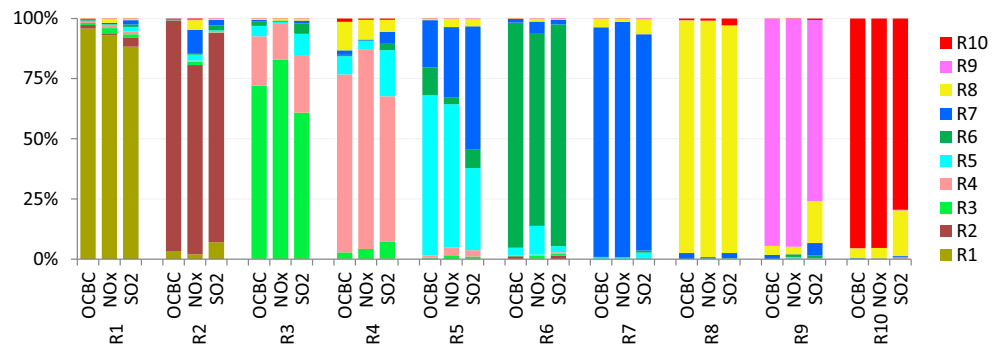

Fig. 3 Damage factors of SO_2, NO_x, and black carbon and organic carbon (BCOC) for ten regions and its breakdown of diseases for the year 2000

Fig. 4 Ratio of damage affects on ten regions caused by one source region for SO_2, NO_x, and black carbon and organic carbon (BCOC) in the year 2000

studies show that the model has the ability to represent observed features in aerosol mass concentration or optical thickness, which is an indicator of the total aerosol loading in the overhead atmosphere. However, these comparisons were not specifically targeted at $PM_{2.5}$. Instead, they were targeted at all aerosols. This was because of the limited availability of $PM_{2.5}$ observations. Although the observation of $PM_{2.5}$ has been rapidly strengthened in recent years, it is still not enough to validate the global or inter-continental scale distribution of $PM_{2.5}$. In that sense, the verification of large horizontal scale fate analysis of $PM_{2.5}$, such as in our study, is a problem for the future.

There are several issues in the design of the fate analysis, which would have had some influences on the results. Firstly, the aerosol processes in the atmosphere are non-linear, especially for secondary aerosols. Also, a 20 % increment in emissions is usually larger than the unit emission increase. Therefore, the concentration change occurring due to additional unit emissions that were estimated in this study might have been overestimated. However, using ozone as an analogy, which is governed by more non-linear processes (Yamaji et al. 2012), a 20 % increment would be small enough to estimate the impact of unit emission increases. In recent years, the emission of air pollutants has been largely changed in several areas in the world, such as large increase in East Asia (Ohara et al. 2007). Therefore, emission data from years more recent than the year 2000 would be preferable. Furthermore, because the meteorological conditions can greatly change year by year, the affected spheres of emission increments in source regions should be estimated with meteorological data from multiple years. Doing so can provide the uncertainty range of the analysis. Finally, the variation among present day global CTMs is still large (Dentener et al. 2010). Thus, estimating the range of DFs by considering different CTM results is important work for the future.

There is few existing LCIA research that shows the results of fate analysis, except for Itsubo and Inaba (2010). They showed that the changes in the mass concentration of $PM_{2.5}$ averaged over Japan because of the unit emissions of primary species (BC and OC) and the precursors of secondary $PM_{2.5}$,

SO_2 for sulfate, and NO_x for nitrate. In Japan, they were 3.5×10^{-9} ($\mu g/m^3/kg$), 1.0×10^{-9} ($\mu g/m^3/kg$), and 0.2×10^{-9} ($\mu g/m^3/kg$) respectively. In this research, they were estimated at 3.0×10^{-9} ($\mu g/m^3/kg$), 0.6×10^{-9} ($\mu g/m^3/kg$), and 0.3×10^{-9} ($\mu g/m^3/kg$) respectively; very similar results were obtained.

4.2 Effect analysis

In order to calculate the regional differences of DFs, not only the variation of transport for $PM_{2.5}$ but also the regional differences of CRF, especially for chronic mortality, should be considered. However, because the values of CRF estimated by cohort studies are not sufficient enough to allow for generalizations, this research applies to the CRF obtained from the USA's cohort study to all ten regions and assumes linearity without threshold. Recently, some new CRFs focus on long-term exposure to $PM_{2.5}$ and natural cause mortality have been provided. A meta-analysis (Hoek et al. 2013) covering worldwide range showed a CRF which similar to the one used in our study. While the value of CRF from another analysis (Beelen et al. 2014) of 22 European cohort studies was about double that of the first. On the other hand, a rare cohort study in China (Cao et al. 2011) showed a CRF of $PM_{2.5}$ which is quite a lot smaller than the one this research used. Although a tendency on regionality of CRFs is indicated, since there are still no detailed explanation of the differences between the above studies have been provided, it seems difficult to make generalization about other regions based on the studies.

On the other hand, a new Global Burden of Disease Study (Lim et al. 2012) estimated health damage occurring due to $PM_{2.5}$ by using a non-linear function between the concentration and risk of disease. They noted that it is better to apply a non-linear function because the CRF is usually estimated by a low yearly average concentration (roughly 5 to 30 $\mu g/m^3$). Actually, much higher concentrations of ambient particulate matter have been recorded in polluted cities in Asia and elsewhere. It means that the CRF could be different depending on the region and concentration of $PM_{2.5}$. As discussed above, it is important for future work that a CRF is selected with consideration given to the variation between the regions.
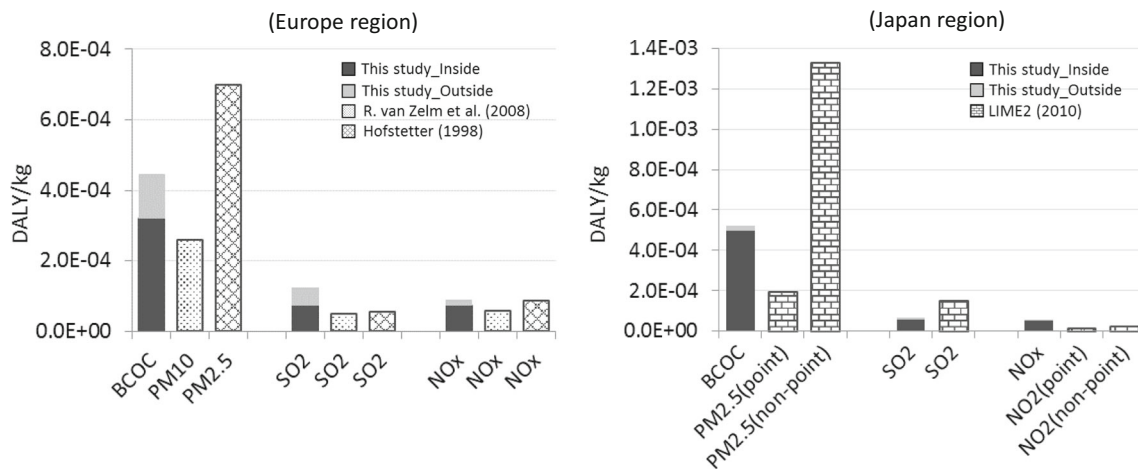
Fig. 5 Comparison with existing researches for regions of Europe and Japan

4.3 Damage factors

Because of the limited regions, where providing DFs in existing researches, we only compare the DFs in the regions of Europe and Japan (Fig. 5). In order to show the effect of transboundary, DFs of this research are divided into "inside" and "outside." These stand for the damage that occurred inside and outside of its area of emission region.

DFs for Europe, using a European Monitoring and Evaluation Program (EMEP) scale, were provided by Hofstetter (1998) and R. van Zelm et al. (2008). They focus on the damage caused by $PM_{2.5}$ and PM_{10} respectively. It is difficult to compare exactly with the results because this research focuses on damage occurring due to $PM_{2.5}$ and considers a smaller Europe region, which excepts Eastern Europe. However, the following two major tendencies can be found. First is that the DFs of existing research are similar to our inside value, except for the DF of primary $PM_{2.5}$ from Hofstetter (1998). It is indicated that the DFs will be underestimated if the transboundary of $PM_{2.5}$ is not taken into account. Furthermore, even though DFs of SO_2 are estimated to be smaller than NO_x in existing researches, when considering transboundary of $PM_{2.5}$, it is very possible that they are larger than that of NO_x in Europe.

For Japan, the DF provided by Itsubo and Inaba (2010) is compared. The DF of primary $PM_{2.5}$ for this research is between $PM_{2.5}$ (point source) and $PM_{2.5}$ (non-point source). For SO_2, the DF for this research is small because a similar CRF is used. However, the concentration change per unit emission is smaller than that of existing research. In contrast, the DF of NO_x in this research is bigger. This is because the concentration change per unit emission is almost same, but a small CRF provided by Pope et al. (1995) is used in existing research.

For the regionality among DFs, the regions with higher population density and emission strength showed bigger value, a general tendency similar to studies on sub-regional scale

with higher resolution was indicated. The study of Krewitt et al. (2001) is one of few researches which estimated the health damage per kilogram emission of NO_x, SO_2, and PM_{10} for continental regions including Europe based on one model. It showed that the result of Japan is roughly the same as that of Europe. The values of China and South America are three times bigger and five times smaller than that of Europe respectively. Although the effect of transboundary is not clear in their research, it was disclosed that the relation between regions was very similar to this research.

5 Conclusions

The health damage factors of ten regions occurring due to $PM_{2.5}$ produced by SO_2, NO_x, and primary particulate (black carbon and organic carbon) were obtained by using a global chemical transport model with consideration to their transboundary effects. The DFs of primary particulates are about five times larger than those of the secondary $PM_{2.5}$ that are produced by SO_2 and NO_x in all regions. However, one digit of difference was found among these regions for all substances.

The regions of India and China showed the largest DFs because of their huge exposure population. DFs in Europe, the Middle East, and Japan were the second largest. High concentration changes per unit of emission were contributed to Europe and Japan. However, the Middle East was largely affected by the high exposure population that occurred in India due to transboundary of $PM_{2.5}$. DFs in other regions showed relatively lower values. For effect of transboundary, it was found that Europe, Russia, and the Middle East showed large shares of damage outside of the emissions region in all three substances. This was around a quarter, a quarter, and a third respectively.

Since the LCA study focuses on products with a global supply chain, it is important to estimate the DFs of $PM_{2.5}$ on a continental level, covering the whole world. In order to express the variation of DFs between regions, not only the transport of air pollutants but also the regional differences of CRF and DALY per case could be important for future research.

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