Science of the Total Environment 466–467 (2014) 673–680

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Validity of using annual mean particulate matter concentrations as measured at fixed site in assessing personal exposure: An exposure assessment study in Japan

Takehiro Michikawa [a,*], Satoshi Nakai [b], Hiroshi Nitta [a], Kenji Tamura [a]

[a] Center for Environmental Health Sciences, National Institute for Environmental Studies, Onogawa 16-2, Tsukuba, Ibaraki 305-8506, Japan
[b] Graduate School of Environment and Information Sciences, Yokohama National University, Tokiwadai 79-1, Hodogaya-ku, Yokohama, Kanagawa 240-8501, Japan

HIGHLIGHTS

- This was the first large-scale assessment of exposure to PM anywhere in Asia.
- For PM and NO_2, the annual correlations between fixed site and outdoor were good.
- Correlations of fixed site with personal exposure were not estimated to be high.
- Our findings are useful to estimate the effects of resulting measurement errors.

ARTICLE INFO

Article history:
Received 21 March 2013
Received in revised form 24 July 2013
Accepted 25 July 2013
Available online 25 August 2013

Editor: Lidia Morawska

Keywords:
Particulate matter
Nitrogen dioxide
Long-term exposure
Ambient air monitoring
Personal exposure monitoring

ABSTRACT

From 2003 through 2005, we compared annual mean particulate matter (PM) and nitrogen dioxide (NO_2) concentrations as measured at fixed-site monitoring stations in 6 Japanese cities with those measured inside and outside subject residences and during personal monitoring. A total of 65 households participated in indoor and outdoor residential exposure monitoring. In summer and autumn, we also performed personal monitoring of one resident of each household. On each day, personal samplers were used to collect 24-h samples of PM and NO_2 simultaneously from the fixed sites, indoor and outdoor, and from those undergoing personal monitoring. We found good correlations between the fixed-site and outdoor measurements for annual mean (average of 7-day × 4-season) concentrations of $PM_{2.5}$, $PM_{10-2.5}$, PM_{10} and NO_2 (Spearman's rank correlation coefficients (ρ) ≥ 0.75). However, the correlations between the fixed-site and indoor measurements were moderate to low. In summer and autumn, the correlations between the fixed-site and personal mean concentrations of $PM_{2.5}$ ($\rho = 0.62$), PM_{10} ($\rho = 0.58$), and NO_2 ($\rho = 0.70$) were acceptable. However, because people spend most of their time indoors, these correlations for annual mean concentrations were not estimated to be high. Our results are important in allowing researchers to estimate the effects of resulting measurement errors of PM and NO_2.

1. Introduction

Particulate matter (PM) is a threat to public health, with the Global Burden of Disease Study estimating that approximately 3.2 million deaths worldwide in 2010 can be attributed to ambient PM exposure (Lim et al., 2012). Short-term exposure to PM is associated with adverse health outcomes (Rückerl et al., 2011), but long-term exposure is likely to have even more serious effects on health (Brook et al., 2010). In spite of growing interest in the association between long-term PM exposure and health outcomes, few epidemiological cohort studies have been carried out in Japan to assess such outcomes as respiratory disease, cardiovascular disease, malignancy, and pregnancy-related problems. To investigate these outcomes, accurate assessments of exposure to PM at the personal level are important.

Most of the earlier epidemiological studies of long-term PM exposure and health outcomes (Dockery et al., 1993; Gehring et al., 2006; Lepeule et al., 2012; Nishiwaki et al., 2013; Pope et al., 2002; Ueda et al., 2011) based their PM exposure assessments on ambient fixed-site measurements: the exposure of individual subjects was determined on the assumption that ambient PM concentrations as monitored at the fixed site had similar temporal and spatial patterns within the defined area (Brook et al., 2010). However, because personal exposure to PM is affected not only by ambient sources but also by non-ambient sources (Wilson et al., 2000), fixed-site measurements are not necessarily

Abbreviations: PM, particulate matter; NO_2, nitrogen dioxide.

* Corresponding author at: Center for Environmental Health Sciences, National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki 305-8506, Japan. Tel.: +81 29 850 2314x2712; fax: +81 29 850 2214.
E-mail address: tmichikawa@nies.go.jp (T. Michikawa).

0048-9697/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.scitotenv.2013.07.084

representative of personal exposure (N.A. Janssen et al., 1998; Jantunen et al., 1998; Sexton et al., 1984). Clearly, measurement of what a subject is actually breathing will yield a more accurate assessment of that person's true PM exposure (Dockery and Spengler, 1981; Howard-Reed et al., 2000; N.A.H. Janssen et al., 1998; Lee et al., 2006; Mage, 1985; Wheeler et al., 1999). Ideally, epidemiological studies would use direct measurements of PM in the breathing zone of each subject, but it is not feasible to carry out such personal monitoring of large numbers of participants over the long-term. If ambient PM concentrations measured at a fixed site are to be used instead as a surrogate for personal exposure, validation of these measurements, i.e. an understanding of the relationship between fixed-site measurements and actual personal exposure, is essential. Evidence validating PM exposure assessments in epidemiological studies would allow us to determine more accurately the association between long-term PM exposure and health outcomes.

We measured concentrations of PM and of another major air pollutant, nitrogen dioxide (NO_2), in 7 Japanese cities throughout 4 seasons. To our knowledge, this was the first large-scale assessment of exposure to PM anywhere in Asia. We compared the annual mean PM and NO_2 concentrations at the fixed sites with PM and NO_2 concentrations at individual sites, including inside and outside subjects' residences and personal monitoring (summer and autumn).

2. Materials and methods

2.1. Study design

From 2003 through 2005, we assessed exposure to PM in 7 Japanese cities: Toride City, Ibaraki Prefecture; Ichikawa City and Urayasu City, Chiba Prefecture; Joetsu City, Niigata Prefecture; Nagoya City, Aichi Prefecture; Moriguchi City, Osaka Prefecture; and Hyuga City, Miyazaki Prefecture (Fig. 1). These cities were selected on the basis of data on certain pollutants (mainly $PM_{2.5}$, i.e. fine particulate matter collected with a sampler with a 50% cut-off point of 2.5 μm) so that comparisons could be made among them. All of the cities except one (Urayasu) had an ambient air pollution monitoring station. We sent a letter requesting cooperation in our study to approximately 200 households in each city, that were located within a 5-km radius of the monitoring station (in the case of Urayasu, the households were located within a 10-km radius of the monitoring station in Ichikawa, which is adjacent to Urayasu). We selected households that included children aged 3 years old who participated in the survey by the Japanese Ministry of the Environment. Of the households from which we received replies, we selected for inclusion in this study those that met the following criteria: i) the household included no smokers (tobacco smoke is a major indoor sources of PM) (Brauer et al., 2000; Ono et al., 2008), and ii) no other obvious sources of PM, such as incense sticks (*senko* in Japanese), were present inside the house. A total of 133 households (18 in Hyuga; 20 in Ichikawa; 18 in Joetsu; 19 in Moriguchi; 20 in Nagoya; 18 in Toride; and 20 in Urayasu) were selected for this exposure assessment study.

Fig. 1. Location of study cities.

In Japan, studies involving only environmental measurements and no collection of personal information on health status did not request approval by any ethical review board before starting the present study. Thus, this study has not been subject to ethical review. However, before starting the environmental measurements, we explained the purpose of our research to the members of each household and included only those that agreed to cooperate.

2.2. Measurements of $PM_{2.5}$, $PM_{10-2.5}$, PM_{10}, and NO_2

Measurements of PM exposure levels were made with a personal sampler connected to a filter folder with cascade impactors of 10 μm and 2.5 μm (ATPS-20H, Sibata Scientific Technology Ltd., Saitama, Japan) and a portable pump (MP-Σ3, Sibata Scientific Technology Ltd., Saitama, Japan) providing a constant airflow of 1.5 L/min. PM larger than 10 μm in aerodynamic diameter was initially collected on a metal impaction plate coated with silicon grease immediately downstream of the inlet. $PM_{10-2.5}$ (i.e., thoracic coarse particulate matter, particulate matter collected between 50% cut-off points of 10 and 2.5 μm) that was not captured by the impaction plate was collected on a 10-mm diameter Teflon filter (TX40HI20, Pall Corporation, Port Washington, NY, USA) with a 50% cut-off point of 2.5 μm. $PM_{2.5}$ that was not captured by the 10-mm Teflon filter was collected on a similar 19-mm diameter Teflon filter. We also measured NO_2 with a filter badge (Advantec Toyo Co. Ltd., Tokyo, Japan).

Using the personal samplers for PM and NO_2, we carried out 24-h measurements of PM and NO_2 for 7 consecutive days at three measurement points simultaneously (inside and outside the selected houses, and at the fixed-site ambient air pollution monitoring station) in each city. In each house, one set (PM and NO_2) of samplers was put on a cabinet or television in the room where the inhabitants spent the majority of their time (usually the living room) for indoor measurements, and another set was installed under the eaves for outdoor measurements. We used the same personal samplers to measure PM and NO_2 at each fixed monitoring site so that accurate comparisons could be made with the indoor and outdoor measurements taken at each residence. Field staff visited the residences and the monitoring stations daily between 0900 and 1200 to change the sampling equipment. Measurements were conducted in 4 seasons: the 1st survey in spring (March through May, 2003), the 2nd in winter (December, 2003 through February, 2004), the 3rd in autumn (September through November, 2004), and the 4th in summer (June through August, 2005). During the 3rd and 4th surveys, we also performed personal monitoring of one inhabitant of each household. Each participant carried the sampler throughout the course of each day, both indoors and outdoors. Since the sampler was also placed in the participant's bedroom while he/she slept, we took steps to minimize the pump noise by using a case insulated with foam. During the personal measurement period, the participants kept records of their location every 15 min.

PM filters were pre- and post-weighed with an electronic microbalance (UMX-2, Mettler-Toledo, Inc., Columbus, OH, USA [detection limit: 0.1 μg]) after a storage period of 24 h in an environmentally controlled