

Technical Manual on Dry Deposition Flux Estimation in East Asia

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1. Introduction

1.1 Background

To assess the ecological impact caused by the acid deposition, a long term monitoring of the acid in both the wet and dry forms are required. The remarkable differences in determining the flux quantity (the amount of the acid deposition per unit area per time) between the wet and dry depositions are the techniques to collect and analyzed the samples. The wet deposition can be collected more readily by collecting the rain water in bulk or by an automatic wet only collector. The collected samples are later analyzed in a laboratory to determine the chemical species. For the dry deposition, the method is much varied from cumbersome to highly sophisticated. The methods to determine the deposition amount can be done directly or indirectly. The direct method and less sophisticated one employed natural surfaces as collectors. Collection of the rain running through the forest canopy (through fall and stem flow) is practical in collecting the samples in the forest. By this method, the dry deposition are obtained by differences between the chemical content in the rain flowing through the tree surfaces and the chemical content in the bulk rain collection. Other direct measurement is by intercepting the dry deposition onto the surrogated surfaces i.e. water surface or chemical impregnated surface. The deposition flux is determined in terms of the weight of acid collected per the area of surrogated surface per collecting time. The more sophisticated direct method is the eddy correlation which requires simultaneous measurements of rapid fluctuations of vertical wind speed and air concentrations. The product of the vertical wind speed and the concentration gradient are the flux of the atmospheric substance. Other meteorological methods are the aerodynamic and the Bowen ratio methods. They rely on the measurements of vertical concentration gradient and meteorological parameters which comprise of the wind speed, humidity, temperature, net radiation etc. The result obtained is the mean potential. When multiply with concentration gradient, the flux of acid is obtained.

The above-mentioned direct and indirect methods are not practical for a long term and regional scale monitoring, as well as required high costs and skilled technicians. In practice, the ambient concentrations are monitored continuously throughout the year. To ultimately determine the dry deposition flux using the air concentration data, the inferential method can be employed. Under this application, the deposition velocity has to be known. The product of the deposition velocity and the air concentration is the amount of the deposition flux. The deposition velocity can be obtained specifically by experiment, or by estimation using the parameterization methods. Presently, this methodology has been employed in many applications.

Chemical compositions of wet and dry components have been monitored by EANET country members since 1998 during the preparatory phase and continued to a regular monitoring basis started in the year of 2001. In the year of 2000, the wet deposition has been initially calculated and included in the data report. To complete the total acid deposition in wet and dry forms, a group of experts under the Task Force on Dry Deposition Monitoring was appointed. The task is to issue the Technical Manual on Dry Deposition Flux Estimation in the EANET region. The methodology for estimation is identified for the inferential method by the Strategy Paper for Future Direction of Dry Deposition Monitoring (Acid Deposition Monitoring Network in East Asia, 2005, 2010). The guidelines for calculating the deposition flux are provided in this manual.

1.2 Objectives

- 1) To quantify the dry deposition flux in the EANET region using the site specific meteorological and ambient concentration monitoring data.
- 2) To produce data for investigating a long term adverse effect of acid deposition in the EANET region.

1.3 Outline of the manual

The technical manual will provide a comprehensive guideline for the EANET countries to estimate the amount of acid deposition using data of EANET monitoring sites. The manual composed of 5 chapters and 3 appendices.

Chapter 1 provides general background and the objective of the work of this technical manual.

Chapter 2 describes the fundamental items for dry deposition flux estimation. The site selection and the instrumentation required for measuring the meteorological data, the priority chemical species to be assessed, the sampling period and the land use information are described in the chapter.

Chapter 3 describes the technical manual will provide templates for EANET countries to report the calculated parameterization terms of dry deposition velocity. Data checking include data flags and invalid data are to be notified in the report.

Chapter 4 describes the methodology for dry deposition flux estimation. The inferential method is outlined. The parameterization terms for dry deposition velocity calculation

referred by the inferential method are provided for both gases and particulate matters. The method to compute dry deposition flux is included. This chapter is also desired to evaluate the value of dry deposition flux obtained by the inferential method comparing with the flux value obtained from other methods, e.g. the gradient method and the throughfall method. The chapter also described the uncertainty in determining air concentration, deposition velocity and flux.

Finally, the future direction of dry deposition flux estimation is discussed in Chapter 5.

Other relevant information related to the future direction of dry deposition is compiled as appendices in the technical manual.

Appendix I reviews the methodologies to determine the dry deposition flux using natural surface, surrogate surface and micrometeorological data. The natural surface method captures the precipitation with the throughfall and stem flow techniques. The surrogate surface employed the prepared impregnated material with substrate in order to enhance its effectiveness in capturing particle and gases. The micrometeorological method estimates the flux of depositing chemical species over the earth surface using the relationship between air concentrations and meteorological data. Advantages and disadvantages for each method are described in the chapter.

Appendix II describes the use of remote sensing information and methods to calculate Normalize the Difference of Vegetation index (NDVI) and the Leaf Area Index (LAI) from the data of NDVI. The data are useful in obtaining the parameterized terms to be used for estimating the dry deposition flux.

Appendix III describes how to operate Microsoft Excel macro program for calculation of deposition velocity in order to the staffs in EANET participating countries can calculate deposition velocities by themselves.

References

- Acid Deposition Monitoring Network in East Asia (2005) Strategy Paper for Future Direction of Dry Deposition Monitoring of EANET Second Edition, Network Center of EANET, 13p.
- Acid Deposition Monitoring Network in East Asia (2010) Strategy Paper on Future Direction of Monitoring for Dry Deposition of EANET (2011–2015), Network Center of EANET, 15p.

2. Fundamental items for dry deposition flux estimation

2.1 Air quality measurements

2.1.1 Siting of air quality monitoring

Dry deposition monitoring sites are classified into three categories: remote sites, rural sites and urban sites according to the objectives of the monitoring. The siting criteria are described in the EANET guidelines (Acid Deposition Monitoring Network in East Asia, 2000).

(a) Remote sites

Remote sites are to be established for the assessment of the state of acid deposition in background areas. The monitoring data can be used to evaluate long-range transport and transmission models of acidic substances in East Asia.

The location of these sites should be selected in areas with no or least influence from local emission and contamination sources. Therefore, remote sites should be located with sufficient distance from significant stationary sources such as urban areas, thermal power plants, large factories and significant mobile sources such as major highways, ports and railways to minimize these influences. It is desirable for remote sites to be located at existing meteorological stations, in particular, upper wind monitoring stations or in their vicinity.

(b) Rural sites

Rural sites are to be established for the assessment of the state of acidic deposition in rural areas or hinterlands. The monitoring data can be used, for instance, to evaluate the effects of acid deposition on agricultural crops and forests.

The location of these sites should be selected in areas with minor influence from local emission and contamination sources. Therefore, rural sites should be sited away from significant stationary and mobile sources and should be free from these influences to the extent possible.

Some rural sites which generally satisfy the criteria for remote sites may also be used to evaluate long-range transport and deposition models of acidic substances.

(c) Urban sites

Urban sites are to be established for the assessment of the state of acidic deposition in urban areas. Urban and industrialized areas, and the areas immediately outside such areas, can be included. The monitoring data can be used, for instance, to evaluate the

effects of acid deposition on buildings and historical monuments. Monitoring data at these sites may also be useful for the assessment of acidity of precipitation and the trends in urban areas.

2.1.2 Priority chemical species for dry deposition monitoring in EANET

2.1.2.1 Major chemical species for flux estimation

The purposes of dry deposition monitoring are (i) to provide data for the evaluation of total acid deposition on soil, vegetation, etc. for the assessment of the adverse effects on specified ecosystems, (ii) to provide data for the evaluation of the regional budget of sulfur and nitrogen with the aid of numerical model. Although O_3 is not an acidic but rather an oxidizing species, it is known to be very harmful to plants. Its deposition rate on vegetation is large and is generally believed to affect ecosystems synergistically with acid deposition. Thus, it is highly recommended to evaluate O_3 deposition together with acid deposition. Moreover Na^+ in particles also concerns the regional budget of sulfur to estimate sea-salt or non-sea-salt sulfate. For these purposes, the concerned chemicals are primarily gaseous SO_2 , NO , NO_2 , O_3 , HNO_3 , HCl , NH_3 and the particulate SO_4^{2-} , NO_3^- , Cl^- , NH_4^+ , Na^+ and Ca^{2+} .

Hourly data are expected where diurnal cycles in deposition velocity are to be monitored explicitly. However for evaluation of dry deposition, the sampling period of air concentrations could be longer than one day, e.g., a week for certain circumstances at a monitoring site.

2.1.2.2 Major chemical species for air quality monitoring

From the viewpoint of air quality monitoring, major substances of concern are gaseous SO_2 , NO/NO_2 , O_3 and the particulate mass. Among these substances, SO_2 , NO_2 , O_3 and particulate matter (PM) are well-known air pollutants from the viewpoint of health impacts etc. Although NO is not harmful, it is a primary pollutants and easily converts to NO_2 in the atmosphere, as a precursor of O_3 . Thus, it is highly recommended to measure NO whenever feasible. For the measurement of PM, PM_{10} is important to detect total amounts of acid and base components in particle; $PM_{2.5}$ is effective to define national, regional and hemispheric transport characteristics.

2.1.2.3 Priority chemical species in EANET

The priority chemical species for EANET dry deposition monitoring are

recommended to be as follows (Acid Deposition Monitoring Network in East Asia, 2005, 2010):

First priority: SO₂, O₃, NO, NO₂ (urban), HNO₃, HCl, NH₃

Particulate component (SO₄²⁻, NO₃⁻, Cl⁻, NH₄⁺, Na⁺, Mg²⁺, K⁺ and Ca²⁺),
PM₁₀

Second priority: NO₂ (rural and remote), PM_{2.5}

Data of PM mass concentration can be used for comparison with particulate component concentrations and clarification of the state of air quality. Since there is currently no methodology to estimate dry deposition flux of PM mass concentration, PM mass concentration should be excluded from flux estimation.

2.1.2.4 Selected chemical species for flux estimation

Considering the practical conditions, the following chemical species for EANET dry deposition monitoring are recommended to be selected as for flux estimation:

First priority: SO₂, NO₂, HNO₃, NH₃, O₃,

Particulate components (SO₄²⁻, NO₃⁻, and NH₄⁺)

Second priority: NO, HCl, Particulate components (Cl⁻, Na⁺, Mg²⁺, K⁺ and Ca²⁺).

Only passive sampling is recommended for measuring NO₂ in rural and remote sites because the commercial chemiluminescence NO_x instrument is known to be vulnerable to overestimate the air concentration. Although the acidity of HCl is comparable to that of sulfuric acid, the ambient HCl concentration is usually negligible in East Asia. The same reason can be applied to NO.

2.1.3 Instrumentation

HNO₃, HCl, NH₃ and particulate components require the use of filter pack or denuder. Automatic instruments for SO₂ (UV fluorescence method), O₃ (UV photometric method) and NO/NO₂ (Chemiluminescence detection method) are suitable to obtain one-hour averaged values of these species for air quality monitoring. Unit of ppb for gas and ug/m³ for particle can be used for data reporting. For calibration of the ozone monitoring, traceability to the international standard of National Institute for Standard and Technology (NIST), U.S.A. should be considered. These one-hour averaged values can be surely used for the purpose of the evaluation of dry deposition after averaging over longer period (e.g. one week).

It should be emphasized that commercial “NO_x chemiluminescence instruments”

with molybdenum converter should not be used for NO₂ measurement at rural and remote sites since its NO_x mode responds not only to NO/NO₂ but also to HNO₃ and other organic nitrates unspecifically. The instruments could be used for NO/NO_x* (Total of NO, NO₂, PAN and partial HNO₃). Its use in urban sites near emission sources may be acceptable for NO₂ measurement since a major component of NO_x would be NO₂ and NO in urban area. In remote and rural areas, passive sampler could be used to measure NO₂, unless advanced research-grade methods can be used. Passive sampler also could be used to measure O₃.

Table 1. Methods suggested for concentration monitoring

Parameters	Method for automatic instrument	Manual Method
SO ₂	Ultraviolet fluorescent(UVF) method	Filter pack Denuder Passive sampler
NO	Chemiluminescence detection (CLD) Method	
NO ₂	Chemiluminescence detection (CLD) Method (urban)	Passive sampler (rural and remote)
HNO ₃	—	Filter pack Denuder Passive sampler
NH ₃	—	Filter pack Denuder Passive sampler
HCl	—	Filter pack
O ₃	Ultraviolet photometric method CLD method	Passive sampler (rural and remote)
Ionic components	—	Filter pack Denuder Low-volume sampler

(Note) The analytical methods used for wet deposition (e.g. Ion Chromatography) can also be applied to analysis of the manual methods.

2.1.4 Sampling period

In case of automatic instruments, hourly data are expected to be gathered. One-hour

averaged values can be used for the purpose of the evaluation of air concentration after averaging for one-week. It is desirable that the air concentration monitoring by automatic instruments is carried out throughout a year. If it is difficult, adequate measurement duration in every month should be determined, taking account of the situation in respective countries.

In case of concentrations of gas and particulate components by denuder and/or filter pack methods, weekly data are expected. Daily data are also acceptable.

2.2 Meteorological measurements

2.2.1 Siting of meteorological instruments

Meteorological measurements should be conducted in the clearing adjacent to the air quality measurement instruments. As in the case of wet deposition monitoring, meteorological instruments should be installed at the place well away from trees, hills, buildings, and other obstructions, e.g. distance between meteorological instrument and obstruction should be at least twice the obstruction height. It is recommended that all of meteorological instruments are installed in the observation field with area of at least 20m x 20m square covered with short grasses. Detailed instructions for installing each meteorological instrument are described in 2.2.3.

2.2.2 Meteorological parameters necessary for dry deposition flux estimation

For the purpose of calculations of deposition velocity, following meteorological parameters are required;

<Mandatory>

- Wind speed
- Temperature
- Relative humidity
- Solar radiation
- Precipitation amount

<Optional>

- Wind direction (Standard deviation of wind direction*)
- Net radiation*
- Cloud cover*
- Surface wetness

All of those parameters, except cloud cover, should be measured at the monitoring sites by using installed equipment. If available, cloud cover data measured at the nearest meteorological station should be used.

*As described in 2.2.5, atmospheric stability can be determined by three different schemes using one of three parameters, standard deviation of wind direction, net radiation and cloud cover. Therefore, it is not necessary to measure all of those three parameters simultaneously.

2.2.3 Instrumentation

The typical instruments for measuring the above selected meteorological parameters are summarized in Table 2.1.

The sensors for wind speed, wind direction, temperature and relative humidity should be installed at a top of meteorological mast or tower, 10 m above the ground. The sensors for temperature, relative humidity, solar radiation and net radiation should be installed at 1.5–2.0 m above ground outside the sun shadows of meteorological mast/tower and surrounding obstructions. The rain gauge should be installed on the ground. The surface wetness sensor should be installed at 20–30 cm above the ground.

Table 2.1 Typical instruments for selected meteorological parameters

Parameter	Instrument
<Mandatory>	
Wind speed	Anemometer
Temperature	Ventilated platinum resistance thermometer
Relative humidity	Ventilated capacitance humidity sensor
Solar radiation	Pyranometer
Precipitation amount	Tipping bucket rain gauge
<Optional>	
Wind direction (Standard deviation of wind direction)	Wind vane
Net radiation	Net radiometer
Surface wetness	Conductivity bridge

In addition to those meteorological sensors, a programmable data logger is required to record measured meteorological data on the site. The logger should be mounted in a weatherproof box at easily accessed place, e.g. at the base of meteorological mast/tower. All of instantaneous data of measured parameters and calculated various statistics (see below) should be recorded in memory card equipped in the logger.

2.2.4 Monitoring period

All meteorological parameters should be measured continuously throughout a year. The instantaneous data of measured parameters (1–10 minutes interval), and to calculate various statistics (e.g. hourly means of wind speed, wind direction, temperature, relative humidity, solar and net radiation, hourly standard deviation of wind direction, hourly sum of precipitation amount, duration of wetness) should be stored in the data logger.

At intervals no longer than one month, recorded data in memory card of the logger should be transferred to PC or other memory module and returned to laboratory for processing. All hourly data should be tabulated separately in each measurements period of air concentration of gases and particles. Based on those summarized hourly data set, hourly deposition velocities for gases and particles will be calculated (see Chapter 4).

2.2.5 Atmospheric stability

As described in Chapter 4, atmospheric stability is one of the key factors for determining dry deposition velocity. In case of adopting the inferential method for calculating dry deposition velocity, the aerodynamic resistance (R_a) and the quasi-laminar layer resistance (R_b) are estimated by the parameter of atmospheric stability, i.e., the Monin-Obukhov length (L). Using one of following estimation methods, the atmospheric stability and/or the Monin-Obukhov length will be determined.

(1) Pasquill stability classes (Seinfeld and Pandis, 2006)

In this method, the stability classes A, B, C, D, E and F shown in Table 2.2 are defined by the prevailing meteorological conditions of: (a) measured surface wind speed and (b) day-time incoming solar radiation or the night-time fraction of cloud cover.

Table 2.2 Estimation of Pasquill stability classes

Surface (10 m) Windspeed, m s^{-1}	Daytime ^c			Nighttime ^c	
	Incoming Solar Radiation ^b			Cloud Cover Fraction	
	Strong	Moderate	Slight	$\geq \frac{4}{8}$	$\leq \frac{3}{8}$
<2	A	A-B	B	—	—
2-3	A-B	B	C	E	F
3-5	B	B-C	C	D	E
5-6	C	C-D	D	D	D
>6	C	D	D	D	D

^aKey: A—extremely unstable; B—moderately unstable; C—slightly unstable; D—neutral; E—slightly stable; F—moderately stable.

^bSolar radiation: strong ($> 700 \text{ W m}^{-2}$), moderate ($350-700 \text{ W m}^{-2}$), slight ($< 350 \text{ W m}^{-2}$)

^cThe neutral category D should be used, regardless of windspeed, for overcast conditions during day or night.

Source: Turner (1969).

Once the stability class is determined, the Monin-Obukhov length (L) can be estimated according to the following correlation proposed by Golder (1972):

$$1/L = a + b \log z_0 \quad (2.1)$$

where a and b are the corresponding coefficients listed in Table 2.3, and z_0 is the roughness length.

Table 2.3 Correlation parameters for the estimation of L

Pasquill Stability Class	Coefficients	
	a	b
A (extremely unstable)	-0.096	0.029
B (moderately unstable)	-0.037	0.029
C (slightly unstable)	-0.002	0.018
D (neutral)	0	0
E (slightly stable)	0.004	-0.018
F (moderately stable)	0.035	-0.036

(2) Modified Pasquill stability classes (Japan Atomic Energy Commission, 1982)

In this modified method, although the day-time stability classes are defined by the same parameters as the original Pasquill's method, night-time procedures are modified. The night-time stability classes are defined by the measured surface wind speed (U) and the measured net radiation (Q). This method is useful for the area where no cloud cover data is available in night-time. Classification is summarized in Table 2.4.

Table 2.4 Estimation of Pasquill stability classes (Japanese method)

Wind speed (U), m/s	Solar radiation (T), kw/m^2				Net radiation (Q), kw/m^2		
	$T \geq 0.60$	$0.60 > T \geq 0.30$	$0.30 > T \geq 0.15$	$0.15 > T$	$Q \geq -0.020$	$-0.020 > Q \geq -0.040$	$-0.040 > Q$
$U < 2$	A	A – B	B	D	D	G	G
$2 \leq U < 3$	A – B	B	C	D	D	E	F
$3 \leq U < 4$	B	B – C	C	D	D	D	E
$4 \leq U < 6$	C	C – D	D	D	D	D	D
$6 \leq U$	C	D	D	D	D	D	D

(3) Method based on standard deviation of wind direction

As shown in Table 2.5, Hicks *et al.* (1987) proposed the method for estimation stability classes based on the measured standard deviation of wind direction.

Table 2.5 Estimation of stability classes (σ_θ method)

Meteorological condition	Stability
$T > 100 (\text{Wm}^{-2})$ and $\sigma_\theta > 10$ (degree)	unstable
other conditions	neutral, stable

T : solar radiation

σ_θ : standard deviation of wind direction

2.3 Land use information

Dry deposition is of our concern for different special areas, i.e., at the monitoring site and for the area of several square kilometers and for meso-scale areas. The target area may be composed of various vegetation (land use) at different vegetation fractions. Even if the air concentrations of pollutants and the meteorological parameters like wind speed are the same for areas with different vegetation and vegetation fractions, the dry deposition flux could be different. It is due to their different roughness length and characteristic length. In the case when the changes of the pollution concentration and meteorological parameters are small in a certain finite area, the dry deposition over it can be estimated by means of information on the vegetation species or the land uses which compose that region. Thus, to estimate dry deposition flux in a certain finite area, land use information is indispensable. Most of the governments in East Asia have prepared the gridded land use information, and if it is available, we can use it for the total dry deposition over the target area.

On the other hand, the global land cover characteristic data (GLCC) is available and can be easily accessed at the USGS (US Geological Survey) Web site; http://educ.usgs.gov/#/Find_Data/Products_and_Data_Available/Land_Cover_Products, which is derived from the Advanced Very High Resolution Radiometer (AVHRR) data obtained by polar orbiting NOAA satellites.

It is given for regular longitude-latitude terrain with the highest resolution of 30 sec. (about 1 km). This data set has been converted to several groups for different purposes and also for lower resolutions, i.e., 1 degree, 30-, 10-, 5- and 2-minutes. The one adopted by Zhang et. al (2001) in their dry deposition studies is mainly the Biosphere Atmosphere Transfer Scheme (BAT) (Dickinson, 1986) with the highest 30 sec. resolution. They regrouped the BAT's original 20 land use categories (LUCs) into 14 LUCs and add "urban" as the 15th land use category. Dry deposition velocity is calculated for all LUCs existing inside the certain finite area and then averaging according to the area fraction of each LUC. Here in this manual, we will follow this categorization.

Since some parameters change with season of the year, it is necessary to define several different seasonal categories. The one used here is the same as in Brook et al. (1999), which was originally reported in Wesely (1989). Five seasonal categories are defined. The 15 LUCs and five seasonal categories used here are listed in Table 2.3.1. Otherwise, global monthly vegetation fraction may also be used, but its resolution is 10-minutes (18.5 km) for 12 months at each of latitude-longitude grid points and covers only from 55 degree north to 75 degree north. To make the data file have global coverage, a zero value of vegetation fraction is usually assigned over the higher latitude area.

In the dry deposition estimation important properties for each LUC and seasonal category are the roughness length " z_o " and characteristic radius " A " such as mean radius of plant leaves and that of surface obstacles. They are listed in Table 2.3.2.

The other important parameter relevant to the vegetation is the Leaf Area Index (LAI). It is defined as the leaf area per unit ground area on which dry deposition of air pollutants mainly occurs. It also represents the activity of vegetation which changes in the course of year. On-site observation of the LAI can be done by means of canopy height, but costly. Recently, extensive trials to estimate the LAI from the satellite data have been done. Advanced Very High Resolution Radiometer (AVHRR) data and Moderate-resolution Imaging Spectroradiometer (MODIS) data are used to determine the LAI, and the detailed procedure is presented in the Appendix II. At the present stage, the estimated LAI has not reach to the level which represents accurately the on-site value, but gives its information on meso and regional scales.

Table 2.3.1 Land use categories (LUC) and seasonal categories (SC)

Category	Description
Land use categories (LUC)	
1	Evergreen-needleleaf trees
2	Evergreen-broadleaf trees
3	Deciduous-needleleaf trees
4	Deciduous-broadleaf trees
5	Mixed broadleaf and needleleaf trees
6	Grass
7	Crops, mixed farming
8	Desert
9	Tundra
10	Shrubs and interrupted woodlands
11	Wet land with plants
12	Ice cap and glacier
13	Inland water
14	Ocean
15	Urban
<i>Seasonal categories (SC)</i>	
1	Midsummer with lush vegetation
2	Autumn with cropland that has not been harvested
3	Late autumn after frost, no snow
4	Winter, snow on ground and subfreezing
5	Transitional spring with partially green short annuals

Table 2.3.2 Roughness length “ z_0 ”(m) and characteristic radius of plants “ A ”(mm) for 15 land use categories (LUC) and five seasonal categories (SC)

	LUC	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Z_0	SC1	0.8	2.65	0.85	1.05	1.15	0.10	0.10	0.04	0.03	0.1	0.03	0.01	$f(u)$	$f(u)$	1.0
	SC2	0.9	2.65	0.85	1.05	1.15	0.10	0.10	0.04	0.03	0.1	0.03	0.01	$f(u)$	$f(u)$	1.0
	SC3	0.9	2.65	0.80	0.95	1.15	0.05	0.02	0.04	0.03	0.1	0.02	0.01	$f(u)$	$f(u)$	1.0
	SC4	0.8	2.65	0.85	1.05	1.15	0.02	0.02	0.04	0.03	0.1	0.02	0.01	$f(u)$	$f(u)$	1.0
	SC5	0.8	2.65	0.85	1.05	1.15	0.05	0.05	0.04	0.03	0.1	0.03	0.01	$f(u)$	$f(u)$	1.0
A	SC1	2.0	5.0	2.0	5.0	5.0	2.0	2.0	na	na	10.0	10.0	na	na	na	10.0
	SC2	2.0	5.0	2.0	5.0	5.0	2.0	2.0	na	na	10.0	10.0	na	na	na	10.0
	SC3	2.0	5.0	5.0	10.0	5.0	5.0	5.0	na	na	10.0	10.0	na	na	na	10.0
	SC4	2.0	5.0	5.0	10.0	5.0	5.0	5.0	na	na	10.0	10.0	na	na	na	10.0
	SC5	2.0	5.0	2.0	5.0	5.0	2.0	2.0	na	na	10.0	10.0	na	na	na	10.0

Note: $f(u)$ represents a function of wind speed (u), and “na” represents not applicable.

References

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3. Data reporting

The data are processed by the personnel in charge of the laboratory (PCL), the national QA/QC managers (NAM) in each participating country and the network QA/QC manager (NEM). The PCL has to collect the measurement data of air concentrations and meteorological parameters and input these data to the reporting format. In addition, other relevant information regarding dry deposition should be reported to the NAM of each participating country. The National Center and the NAM have to report monitoring data to the Network Center. The Network Center and the NEM has to compile, compute and verify the database of dry deposition and provide a copy of the database to the participating countries when requested.

3.1 Classification of data

Data are classified into two types: (1) Data to be processed and reported by the PCL to the NEM through the NAM, (2) Data to be processed by the NEM, or supplemental data should be reported to the NEM by a request.

3.1.1 Reporting data

The data to be reported to the NAM are grouped into two types: 1) Information about sites, monitoring condition, shipping of the filter pack or other manual monitoring samples, laboratory operation, chemical analysis, etc., 2) Measurement results of air concentrations by automatic monitors and the manual monitoring, measurement results of meteorological parameters, and other parameters required to calculate dry deposition fluxes. Remarks and notes also compose major parts of the measurement results.

(1) Information about sites, sampling, shipping, laboratory operation, chemical analysis

- Name of country and site (Code of country and site)
- Name of the NAM
- Name of responsible laboratory and the PCL (Code of laboratory)
- Information of site (on-site scale, local scale and regional scale)
- Information of monitoring condition (automatic monitors, a filter pack or other manual monitoring sampling instrument, meteorological instruments, etc.)
- Information of filter pack or other manual monitoring samples history (shipping frequency, packing procedure, laboratory operation, etc.)
- Chemical analysis condition by ion chromatography (Control chart)

(2) Measurement and calculated results

- Name of country and site (Code of country and site)
- Name of the NAM
- Name of responsible laboratory and the PCL (Code of laboratory)
- Sample number (Code consist of country code, site code, year, month and number)
- Start and end of date and time of monitoring
- Date of chemical analysis for filter pack or other manual monitoring samples
- Air concentration measured by automatic monitors (SO₂, NO, NO₂ and O₃)
- Air concentration measured by the filter pack or other manual monitoring methods (SO₂, HNO₃, HCl, NH₃, and Particle matter components, etc.)
- Meteorological parameters (Wind Speed, Wind direction, Temperature, Relative humidity and Solar radiation)
- Leaf Area Index (Optional)
- Notes and any other information

3.1.2 Local circumstances information

The local circumstances information to be reported by individual country (NAM) are: (1) the information which affects air concentrations such as specific meteorological data, climate, climate vegetation and life style, (2) the information which affects measurement accuracy such as conditions of monitoring instruments, laboratory conditions. These data should be reported when there is a demand.

(1) Information of effects on air concentrations

- Specific meteorological data
- Climate (rainy and dry season, season of sand storm, volcanic condition, etc.)
- Vegetation (the type of tree, season of pollen dispersion, etc.)
- Life style (agricultural operation, biomass burning, etc.)

(2) Information on the precision of monitoring results

- Conditions of monitoring instruments, (calibration methods, failure and incidents on instruments, maintenance conditions, etc.)
- Laboratory condition (maintenance conditions for ion chromatography, instrument list, chemicals, etc.)

3.2 Data checking

Data checking or validation is based upon:

- Experience with the data from earlier measurements,
- Knowledge about spatial and temporal variation.

Records of old data can be used to create simple statistics including percentiles, mean values and standard deviations. Log-transformed data are sometimes considered. These statistics can be used in connection with control chart or in other comparisons of new data with aggregation of the old ones.

Relations between various chemical components (including ion balance of particulate matter components), relationship between sea salt components, and relationship between air concentrations from neighboring stations and time-series plots are also useful.

3.2.1 Statistical tests

The statistical tests are comparisons between new measurement and calculated results and data already stored in the database. The tests are carried out to identify possible outliers and results which may be wrong. They can be based upon assumption about the data distributions. In some cases, respective air concentrations and calculated dry deposition fluxes may be compared with earlier data using the lognormal distribution. Data which is not within the four times the standard deviations range, should be checked by comparison with air concentrations and other results obtained on earlier and later days, and results from neighboring sites.

3.2.2 Data completeness

Data completeness should be evaluated in terms of the flagged or invalid data for automatic monitoring and the filter pack or other manual monitoring methods. Data completeness describes the fraction of valid data coverage length in a certain monitoring period. Data completeness also should be described in the report form. The definition of data completeness is expressed as follows;

(Data completeness for automatic monitor)

$$= (\text{Number of valid hourly data}) / (\text{Number of total measured hourly data}) \quad (3.1)$$

(Data completeness for the filter pack or other manual monitoring methods)

$$= (\text{Number of valid measurement days}) / (\text{Number of total measurement days}) \quad (3.2)$$

In order to evaluate monthly and annual dry deposition fluxes, data completeness should

be no less than 70%. Otherwise, those data will be flagged in the report.

3.2.3 Analytical precision

The precision of laboratory chemical analyses of blank samples for the filter pack method should be tested and be reported by the method described on “the Technical Document for Filter Pack Method in East Asia” For or other manual monitoring, the similar blank test should be conducted.

3.3 Data flags and invalid data

To indicate quality information to data users, data flags and/or data comments are useful; they will indicate whether a data is valid or invalid. The function of the flags and comments is to ensure that the user has full knowledge of the data validity, and of conditions which produce that level of validity. Thereby, the user can select the data most appropriate to his/her application.

(Code number of data flags)

999	Missing measurement
781	Below detection limit
701	Less accurate than usual
699	Mechanical problem
599	Contamination of the samples or the sampling system

3.4 Data reporting form

Data reporting forms may be used for the reporting site condition (on site, local scale, regional scale), monitoring condition, sample history, chemical analysis condition and measurement results (air concentrations, meteorological parameters and other necessary results, flags and data completeness). Staffs in the Network Center in charge of dry deposition data will have responsibility for ensuring that all data elements are properly entered into the appropriate databases. Data should be submitted to the Network Center once every year.

The Network Center will provide the formatted data reporting form as a Microsoft Excel file. All data should be input in the distributed data form. The followings should be described on every data form: name of country and site (code of country and site), name of the NAM, name of laboratory (code of laboratory) and name of the PCL.

3.4.1 Information about sites, sampling, shipping, laboratory operation

(1) Site condition

Any change in the circumstances of the site should be reported every year, even if the site selection criteria remain satisfied. Name and code of each site should be given first. The category of a site should be determined by consideration of the siting criteria. No site is included in more than one class.

The area around the site should be especially described in terms of potential sources of contamination of samples on three different scales. Maps of the site and potential contamination sources should be provided to the Network Center.

- On-site scale

Description of the on-site scale is given for the area within a radius of 100 m from the site. Locations of automatic monitors, a filter pack or other manual monitoring instruments, and meteorological instruments should be given. Trees, overhead wires, buildings, and other physical obstacles should be also described. Ground cover and slope, and farmlands etc., are also important factors. Pictures of the monitoring instruments and their surroundings should be attached. Seasonal specific condition such as snowfall, dust storm and seasonal variation of site conditions should be reported. This information will be used for land use information described in Chapter 2.3.

- Local scale

Surface storage of agricultural products, fuels, vehicles, parking lots, or maintenance yards and feed lots, dairy barns or a large concentration of animals within a radius of 100 m–10 km should be described. Urban areas will be also described with population.

- Regional scale

Both stationary and mobile emission sources within 50 km should be described with emitted chemical species and emission intensities. Urban areas with population greater than 10,000 should be described. Near meteorological stations should be described on the map with available information.

(2) Sampling condition

- Automatic monitors

The following should be reported: model and manufacturer of the monitors, kind of calibration gas, calibration methods and frequency, shapes of a manifold or a sample inlet, tubing, air condition status in the monitoring station. Start and end times of sample

collection in the sampling plan should be reported. Pictures of collector and design diagrams should be attached.

- Filter pack or other manual monitoring instruments

The following should be reported: model and manufacturer of instruments, suction pump, and a flow meter, calibration method of flow rate, arrangement of instrument components, tubing, air condition status in the monitoring station. Start and end times of sample collection in the sampling plan should be reported. Pictures of collector and design diagrams should be attached.

- Meteorological instruments

The following should be reported: model and manufacturer of the instruments, maintenance records, arrangement of the instruments in the site, and monitoring frequency (duration interval). Pictures of collector and design diagrams should be attached.

(3) Sample history

Sample history plays an important role in sample handling from collection to chemical analysis of the filter pack or other manual monitoring samples.

- Shipping

Shipping frequency and packing procedure for collected samples should be also reported.

- Laboratory operation

The following should be reported: sample preparation procedure, plan of chemical analysis frequency, range of laboratory room temperature.

3.4.2 Analytical condition for filter pack samples

The chemical analysis of the filter pack or other manual monitoring samples and control chart should be reported as the laboratory QA/QC data for each sampling station. The following items are included in the form: Method applied, instrument name and type, detection limit, calibration curve (5 points), ion concentrations in the deionized water (when a dilution process is included), data obtained from analysis of standard solution (commercial Standard Reference Material) of known ion concentrations, data from duplicate or triplicate analysis of samples, data of blank filters.

3.4.3 Measurement results and flags

Air concentrations obtained by automatic monitors and analytical results obtained by the filter pack methods must be accurately input in the proper place in the formatted data report form. Items to be input are listed as follows:

- 1) Gas concentration unit with the unit of ppb (SO_2 , NO, NO_2 , NO_x^* and O_3)
- 2) Analytical results of the filter pack method with the unit of mg L^{-1} (F0: SO_4^{2-} , NO_3^- , Cl^- , NH_4^+ , Na^+ , K^+ , Mg^{2+} and Ca^{2+} ; F1: SO_4^{2-} , NO_3^- , Cl^- and NH_4^+ ; F2: SO_4^{2-} and Cl^- ; F3: NH_4^+). Duration time of the filter pack other manual monitoring.
- 3) Meteorological Parameters (Wind Speed with the unit of m/s, Wind direction, Temperature with the unit of $^{\circ}\text{C}$, Relative humidity with the unit of %, Sunshine duration with the unit of hours, Solar radiation with the unit of MJ/m^2) Duration time of meteorological measurement.
- 4) Class of vegetation.

If data are flagged or failed results, corresponding cells must be kept the blank. The reporting form should also include sampling conditions, date of chemical analysis, and other remarks. An example of the data report form is shown in the attached electric file. Each national center should keep all the raw measurement data for future reference.

References

- Acid Deposition Monitoring Network in East Asia (2000) Technical Manual for Wet Deposition Monitoring in East Asia, 68p.
- Malé Declaration (2004) Technical Documents for Wet and Dry Monitoring, Chapter 4, pp.16–23.

4. Methodology for dry deposition flux estimation in EANET

4.1 Fundamental items of the inferential method in EANET

Based on the Strategy Paper for Future Direction of Dry Deposition Monitoring of EANET (Acid Deposition Monitoring Network in East Asia, 2005, 2010), the inferential method is adopted to estimate the dry depositions of the target species defined in Chapter 2.1. The method can estimate the dry depositions around the monitoring sites using the data measured at the sites. Since deposition velocity depends on height, a reference height $z_{\text{ref}} = z - d$, where z is height above ground d is the so-called zero-plane displacement height, should be determined. This manual recommended that the target area and the reference height for the flux estimation should be set at 1 km around the sites and 10 m, respectively.

4.2 Parameterization of dry deposition velocity

Suitable parameterizations to estimate the deposition velocities of the target species were arranged by Matsuda (2008) fundamentally based on Wesely's parameterization (Wesely, 1989). The parameterizations were arranged taking into account previous field studies performed in East Asia (e.g. Takahashi et al., 2002; Matsuda et al., 2002; Sorimachi et al., 2003; Sorimachi et al., 2004; Takahashi and Wakamatsu, 2004; Matsuda et al., 2005; Matsuda et al., 2006; Hayashi et al., 2006). The parameterizations are described in this section. Fundamental theory of the deposition velocity parameterizations were introduced in the texts of Erisman and Draaijers (1995) and Seinfeld and Pandis (1998).

4.2.1 Gaseous species

Framework of deposition velocity parameterizations for gases is determined by the equation on the basis of Wesely and Hicks (1977):

$$V_d^i(z) = (R_a(z) + R_b^i + R_c^i)^{-1}, \quad (4.1)$$

where $V_d^i(z)$ is deposition velocity of i species at $z (= z_{\text{ref}} + d)$, height above the ground, R_a is the aerodynamic resistance, R_b is the quasi-laminar layer resistance and R_c is the surface resistance. R_a is obtained by the following equation (Erisman & Draaijers, 1995):

$$R_a(z) = (ku^*)^{-1} [\ln((z-d)/z_0) - \Psi_h((z-d)/L) + \Psi_h(z_0/L)], \quad (4.2)$$

where k is the Von Karman constant, u_* is the friction velocity, z_0 is the roughness length, d is the zero-plane displacement height, L is the Monin-Obukhov length and ψ_h is the stability correction function for heat. Friction velocity is obtained by the following equation (Erisman & Draaijers, 1995):

$$u_* = (ku(z)) [\ln((z-d)/z_0) - \Psi_m((z-d)/L) + \Psi_m(z_0/L)]^{-1} \quad (4.3)$$

where $u(z)$ is wind velocity at height z , Ψ_m is the stability correction function for momentum. Ψ_h and Ψ_m are calculated from the following equations.

$$\Psi_m((z-d)/L) = \Psi_h((z-d)/L) = -5.2 (z-d)/L \quad (4.4)$$

for stable conditions and,

$$\begin{aligned} \Psi_m((z-d)/L) &= 2\ln((1+x)/2) + \ln((1+x^2)/2) - 2\arctan(x) + \pi/2 \\ \Psi_h((z-d)/L) &= 2\ln((1+x^2)/2) \\ x &= [1 - 16(z-d)/L]^{0.25} \end{aligned} \quad (4.5)$$

for unstable conditions.

R_b is obtained by the following equation (Erisman & Draaijers, 1995):

$$R_b = (2/k u_*) (Sc/Pr)^{2/3}, \quad (4.6)$$

where Sc is the Schmidt number and Pr is the Prandtl number.

When the method based on standard deviation of wind direction is adopted for the atmospheric stability estimation, R_a and R_b can be calculated directly by following empirical formulas without estimation of the Monin-Obukhov length (L):

$$\begin{aligned} R_a &= 4/ (u\sigma_\theta^2) & (\text{neutral, stable}) \\ R_a &= 9/ (u\sigma_\theta^2) & (\text{unstable}) \end{aligned} \quad (4.7)$$

$$R_b = (2/ ku_*)(Sc/Pr)^{2/3} \quad (4.8)$$

On the basis of Wesely (1989), R_c is expressed by:

$$R_c = [(R_s + R_m)^{-1} + (R_{lu})^{-1} + (R_{dc} + R_{cl})^{-1} + (R_{ac} + R_{gs})^{-1}]^{-1}, \quad (4.9)$$

where the first and second terms are resistances in the upper canopy, which include the stomatal (R_s), mesophyll (R_m) and outer surface (R_{lu}) resistances; the third term represents resistances in the lower canopy, which include the resistance to transfer by buoyant convection (R_{dc}) and the resistance to uptake by exposed surfaces (R_{cl}); and the fourth term represents resistances to transfer (R_{ac}) and uptake (R_{gs}) on the ground. Each resistance ($s\ m^{-1}$) is calculated from the following equations (Wesely, 1989):

$$R_{st}^i + R_m^i = R_{st} (D_{H_2O}/D_i) + 1/(3.3 \times 10^{-4} H_i^* + 100 f_0^i) \quad (4.10)$$

$$R_{st} = R_j [1 + (200/(G+0.1))^2 (400/(T(40-T)))]$$

$$R_{lu}^i = R_{lu} / (10^{-5} H_i^* + f_0^i) \quad (4.11)$$

$$R_{dc} = 100 (1+1000/(G+10)) / (1+1000\theta) \quad (4.12)$$

$$R_{cl}^i = (10^{-5} H_i^*/R_{cls} + f_0^i/R_{clo})^{-1} \quad (4.13)$$

$$R_{gs}^i = (10^{-5} H_i^*/R_{gsS} + f_0^i/R_{gsO})^{-1} \quad (4.14)$$

where, D_{H_2O}/D_i is the ratio of the molecular diffusivity of water to that of specific gas, H^* is the effective Henry's law constant ($M\ atm^{-1}$) for the gas, and f_0 is normalized (0 to 1) reactivity factor for the dissolved gas, G is solar radiation ($W\ m^{-2}$), T is temperature (Celsius degree), θ is the slope of the local terrain (rad). H^* and f_0 are based on Wesely (1989). Input resistances (R_j , R_{lu} , R_{cls} , R_{clo} , R_{ac} , R_{gsS} , R_{gsO}) in each land use type and each seasonal category are also defined by Wesely (1989).

For soluble gaseous components (not only SO_2 but also NH_3), the R_{lu} parameterization (Wesely, 1989) should be changed to following parameterizations taking into account the effect of enhanced uptake by wet canopies: the canopy cuticle or external leaf resistance defined by Erisman (1994) for SO_2 and the single non-stomatal resistance for uptake primarily to water films defined by Smith (2000) for NH_3 .

R_{lu} for SO_2 (Erisman, 1994):

during or just after precipitation: $R_{lu} = 1$

in all other case:

$$\begin{aligned} R_{lu} &= 25000 \exp [-0.0693 RH] \quad \text{at } RH < 81.3\% \\ R_{lu} &= 0.58 \times 10^{12} \exp [-0.278 RH] \quad \text{at } RH > 81.3\% \end{aligned} \quad (4.15)$$

R_{lu} for NH_3 (Smith, 2000):

$$\begin{aligned}
R_{lu} &= 10^{\log(T+2)} \exp[(100 - RH)/7] \quad \text{at } T > 0 \\
R_{lu} &= 200 \quad \text{at } -5 < T < 0 \\
R_{lu} &= 1000 \quad \text{at } T < -5
\end{aligned} \tag{4.16}$$

where RH is relative humidity (%).

For HNO_3 , the default parameterization could be used because the surface resistance is extremely small and negligible compared with the R_a and R_b .

4.2.2 Particulate matter

Framework of deposition velocity parameterization for particulate matter is determined by modified Slinn (1982) model (Erisman et al., 1997):

$$V_d = (V_{ds}^{-1} + R_a)^{-1} + V_s \tag{4.17}$$

where V_s is the deposition velocity due to sedimentation. For fine particle, V_s can be negligible.

V_{ds} for grass surface is calculated from Wesely et al. (1985):

$$V_{ds} = u_* / 500 \tag{4.18}$$

for stable conditions and,

$$V_{ds} = (u_* / 500) [1 + (300 / (-L))^{2/3}] \tag{4.19}$$

for unstable conditions.

V_{ds} for forest surface is calculated from (Ruijgrok et al., 1997):

$$V_{ds} = E u_*^2 / u_h \tag{4.20}$$

where u_h is the wind speed at canopy height; E is the collection efficiency. E parameterization is given for deferent components and conditions. Erisman et al. (1997) mentioned that the most important parameters used for generalization to other forests are well represented in the parameterization of E . Matsuda et al. (2010) also mentioned the parameterization is considered to be most applicable to a deciduous forest in Japan based on the direct measurements of $\text{PM}_{2.5}$ sulfate.

4.3 Computation of dry deposition flux

4.3.1 Setting of fundamental parameters at each monitoring site

At first the following parameters at each monitoring site should be set taking the conditions in Chapter 2.3 and Wesely (1989) into account.

- Land use type around 1 km from each site,
- Seasonal category based on the climate at each site,
- Roughness length (z_0),
- Zero-plane displacement height (d).

Typical values for the zero-plane displacement height (d) and the roughness length (z_0) range from $0.5 h_c$ to $0.85 h_c$ and $0.05 h_c$ to $0.12 h_c$, respectively. A parameterization is available to determine d and z_0 as functions of profile shape and LAI (Meyers et al., 1998). LAI data obtained by satellite remote sensing is introduced in Appendix II.

Next deposition velocities are calculated by observed data at each site such as wind speed, solar radiation, temperature, relative humidity and precipitation. Net radiation or cloud coverage information should be obtained to determine Pasquill class taking the conditions in Chapter 2.2 into account.

Updated external resistance and E parameterization request the information of period of wet or dry surface. For example, the period during or just after precipitation (3 hours) is regarded as wet surface, otherwise dry surface.

4.3.2 Computation of dry deposition flux

Dry deposition flux of gaseous and particulate species is calculated from the product of air concentration and deposition velocity:

$$F_i = V_d^i \times C_i \quad (4.21)$$

where F_i is flux of i species and C_i is concentration of i species. At first, hourly deposition velocity is calculated, and then the deposition velocity is averaged in the time resolution of air concentration.

4.3.3 Program file for dry deposition velocity and flux calculation

The Network Center provides the program file for dry deposition velocity and flux calculation. This program file is executed by a Microsoft Excel macro file, and one can download at the EANET Web site. If the necessary data listed in Chapter 3.4.3 and the

macro program is run, the dry deposition velocities and dry deposition fluxes for respective components will be automatically calculated. If an error is occurred, one should check the input data. Detailed explanation of how to operate Microsoft Excel macro is shown in Appendix III.

The methodology of dry deposition flux estimation has room for improvement. Further details of data handling and data reporting will be established on the basis of preliminary monitoring activities. Therefore, the program file for dry deposition velocity and flux calculation will be updated as needed.

4.4 Evaluation of dry deposition flux determined by the inferential method

4.4.1 Uncertainties of the inferential method

4.4.1.1 Air concentration

Evaluation of measured air concentrations is achieved by the QA/QC protocol of EANET. The precision and accuracy of measurement can be raised technically. It is, however, impossible to perfectly eliminate the uncertainty attributed to air concentration. Because, the inferential method assumes that there is no process except dry deposition which changes the air concentrations of target substances. But, such processes exist in the atmosphere and cause errors in estimation of dry deposition flux. Three possible processes are as follows:

- Emission from the ground surface, e.g., NH_3 , NO , HNO_2 .
- Gas-particle interconversion in the atmosphere, e.g., NH_3 -particulate, NH_4^+ , HNO_3 -particulate, NO_3^- , SO_2 -particulate, SO_4^{2-} , HCl -particulate, Cl^- , and HNO_2 -particulate, NO_2^- .
- (Photo-) chemical reactions in the atmosphere and on surfaces, e.g., photochemical formation and removal of HNO_2 , and heterogeneous reaction of NO_2 on surfaces.

Regarding the surface emission, Andersen et al. (1999) reported the occasional NH_3 emission even from a forest as natural vegetation. Surface emission directly increases air concentrations of corresponding substances near the ground surface. It is, however, difficult to separate the effect of emission from the observed air concentrations. For NH_3 , well known its occasional surface emission, an alternative inferential method has been developed to correct the deposition velocity by considering the effect of surface emission instead of correcting air concentrations (e.g., Sutton et al., 1998; Spindler et al., 2001). In the improved method, the effect of surface emission is treated as an enhancing factor of surface resistance.

Regarding the gas-particle interconversion, the one-way formation of ammonium sulfate from the condensation of NH_3 with SO_2 (or H_2SO_4) is relatively well known (e.g., Cape et al., 1998; van Oss et al., 1998). Gas-particle interconversion also exists in the NH_3 – HNO_3 – NH_4NO_3 system. The condensation of particles reduces gaseous concentrations which results in an underestimation of gaseous deposition when the inferential method was applied, and inversely, the evaporation of particles causes an overestimation of gaseous deposition.

Regarding the chemical reactions, changes in air concentration of target substances due to the formation and removal in the atmosphere and/or on surfaces lead to errors of gaseous deposition. There is some trial (e.g., Watt et al., 2004) that the effects of formation and removal were eliminated using the flux gradient, not the concentration gradient, and then the actual exchange fluxes not affected by chemical reactions were estimated.

4.4.1.2 Deposition velocity

To begin with, the inferential method was originally developed to substitute micrometeorological techniques such as the gradient method requiring high cost of devices. In the inferential method, deposition velocity is obtained by the resistance model based on parameterization (Hicks et al., 1987; Wesely, 1989). Therefore, the derived deposition velocity perhaps has considerable degree of uncertainty. Furthermore, direct evaluation of the derived deposition velocity is impossible unless micrometeorological measurements are used at the same time.

The resistances in the resistance model are broadly divided into the aerodynamic resistance (R_a), the sub-laminar resistance (R_b), and the surface resistance (R_c); R_c also composes of sub-resistances such as stomatal, mesophyll, cuticular, and soil resistances (see Chapter 4.2).

R_a governed by the eddy diffusion in the atmosphere is common regardless of gaseous species. Although, further research themes remain in the field of micrometeorology, R_a can be determined relatively strictly.

R_b is slightly different among gaseous species due to the differences in molecular diffusion in sub-laminar flow. However, these differences can be determined precisely using the theory of molecular diffusivity.

In contrast, each of the sub-resistances composing R_c largely varies among gaseous species due to the differences in physicochemical properties such as hydrophobic or hydrophilic and high or low in chemical activity. The early studies of inferential method treated the surface resistance as the residue subtracting R_a and R_b from the total resistance derived from micrometeorological measurements; this is why R_c was once

called the residual resistance. The early studies such as Wesely (1989) and Erisman et al. (1994) provided the excellent parameterizations for each of sub-resistances of R_c , which have been widely adopted in relevant studies; a revised parameterization is now also available (e.g., Zhang et al., 2003). However, a large part of the uncertainty of inferential method might be attributed to the parameterization of R_c . It is expected that the parameterization of R_c well agrees with the actual status when applied to the same conditions in relation to vegetation type and climate. On the other hand, it is concerned that the parameterization of R_c established in Europe and the U.S.A. causes considerable uncertainty when applied to East Asia where its climate, vegetation, and soil conditions are diverse and largely different from those in Europe and the U.S.A.

For example, Wesely (1989) gives the stomatal resistance (R_{stom}) with a correction function of global solar radiation and air temperature which affects the stomatal aperture. This correction function is common regardless of plant species. The optimum solar radiation and temperature should vary among plant species. Erisman et al. (2004) also recognized the large uncertainty in surface resistance parameterizations, and concluded that the uncertainty would be 40% or more. It is important to compare the parameterizations, particularly for the surface resistance, with micrometeorological measurements.

4.4.1.3 Flux calculation

In the inferential method, a deposition flux is expressed as the product of air concentration and deposition velocity. Although, high-frequency measurements are ideal, weekly or biweekly measurement for air concentrations seems realistic in a long-term monitoring activity. A flux is expressed by the product of “mean” concentration by “mean” deposition velocity during one measurement. However, this calculation causes the uncertainty of calculated deposition flux. More specifically,

$$C = \bar{C} + C' \quad (4.22)$$

$$V_d = \bar{V}_d + V_d' \quad (4.23)$$

$$\bar{F} = \overline{C \cdot V_d} = \bar{C} \cdot \bar{V}_d + \overline{C' \cdot V_d'} \quad (4.24)$$

where C , V_d , and F denote the air concentration, the deposition velocity, and the deposition flux, respectively. The crossbar and apostrophe are the mean value and the deviation from mean value, respectively. What we want to know is the leftmost side in Eq. 4.24, i.e., the mean flux. But, what we can know from a long-term measurement is only the first term in the rightmost side in Eq. 4.24, i.e., the product of the mean air

concentration by the mean deposition velocity. The second term in the rightmost side in Eq. 4.24, i.e., the covariance between air concentration and deposition velocity has a potential of uncertainty. In other words, the correlation between air concentration and deposition velocity results in the uncertainty of flux calculation, regardless whether the correlation is apparent or actual.

Many gaseous species show a diurnal change in air concentration, and simultaneously atmospheric stability also has a diurnal change and then leads to a diurnal change in vertical transportation including deposition velocity, i.e., large in daytime and very small in nighttime. Therefore, the uncertainty due to the correlation between air concentration and deposition velocity tends to increase when an averaging time becomes long, particularly in the case of lumping day and night. For example, a positive correlation was found for NH_3 (e.g., Hayashi et al., 2009a). Hansen et al. (1998) reported that the uncertainty in annual flux of NH_3 at a heath in Denmark based on weekly monitoring was estimated to 10–50%. Clarke et al. (1997) also reported that the uncertainty in dry deposition of SO_2 and HNO_3 for all data in CASTNET in the U.S.A. based on weekly monitoring underestimated the flux by 5–15% and 5–20%, respectively.

4.4.2 Evaluation of the inferential method for gaseous species

The target gaseous species for dry deposition flux estimation are primarily what monitored in the EANET activities, i.e., SO_2 , HNO_3 , HCl , NH_3 , NO , NO_2 , and O_3 . It is also suggested that HNO_2 should be involved in dry deposition flux estimation in future activities.

The inferential method gives a dry deposition flux as the product of the air concentration at a target height and the deposition velocity at the same height (Hicks et al., 1987; Wesely, 1989). The air concentration is obtained from observations. On the other hand, the deposition velocity is inferred from parameterization. It is, therefore, desirable that the estimated flux by inferential method is evaluated with comparisons to fluxes obtained from the other methods, e.g., the gradient method (e.g., Matsuda et al., 2005; Hayashi et al., 2009b) and the throughfall method (e.g., Horváth, 2003; Schmitt et al., 2005; Zimmermann et al., 2006). In particular, the gradient method on the basis of micrometeorology gives exchange fluxes of high precision. On the other hand, the throughfall method is practically and then only applicable to forests, which also has uncertainly originated from the nutrient exchange within the canopy.

Direct measurements of fluxes are required for improved parameterizations of the inferential method, and micrometeorological approaches have been used extensively (Wesely and Hicks, 2000). It is, however, unrealistic on the cost front that the gradient

method is conducted at all monitoring stations; consequently, the inferential method is chosen as the possible best way. Moreover, East Asia has diverse climate and vegetation. It is, hence, suggested that quantitative evaluation of the uncertainty of inferential method is made in conjunction with the gradient method (or other adequate micrometeorological technique) per typical combination of climatic zone and vegetation type. Once the uncertainty could be evaluated, the results can be applied to correct the dry deposition flux obtained from the inferential method.

When a deposition flux is obtained by a direct measurement (e.g., gradient method) (F_{obs}), the deposition velocity derived from the observation ($V_{\text{d obs}}$) is expressed by,

$$V_{\text{d obs}} = \frac{F_{\text{obs}}}{C} \quad (4.25)$$

Since the inferential method gives well estimation of R_a and R_b (Acid Deposition Monitoring Network in East Asia, 2005), R_c based on the observation ($R_{\text{c obs}}$), i.e., the residue of the total resistance, is expressed by,

$$R_{\text{c obs}} = \frac{1}{V_{\text{d}}} - R_a - R_b \quad (4.26)$$

A systematic error of dataset between $R_{\text{c obs}}$ and R_c by the inferential method is ascribed to the parameterization of the inferential method which should be revised.

4.4.3 Evaluation of the inferential method for particulate matter

The most well known equation to calculate the deposition velocity of particulate matters is obtained by Wesely et al. (1985), which was derived from the measurements of fine fraction of particulate sulfate at short grassland. It is, therefore, possible that this equation causes errors when applied to coarse particles. Furthermore, it is highly possible that this equation underestimates the deposition velocity of particulate matters when applied to vegetation with large aerodynamic roughness, e.g., forest. Erisman et al. (1997) pointed out that the early parameterization for deposition velocity of particles underestimated the particulate dry deposition in forests. Unfortunately, little is known about the canopy exchange of particulate matters.

Temporal changes in air concentration of particulate matters are smaller than those of gases. It is, hence, expected that the uncertainty originated from the flux calculation, i.e., the product of mean air concentration by mean deposition velocity, is small for particulate matters compared to gases. On the other hand, the gas-particle interconversion in the atmosphere also affects the estimated dry deposition of particulate matters as well as gases.

The deposition velocity of particulate matters is expressed by the aerodynamic resistance (R_a) and the sub-laminar resistance (R_b) adding the effect of atmospheric

stability (e.g., Wesely et al., 1985). It can be said that the surface resistance (R_c) is out of consideration for dry deposition of particulate matters. However, particulate matters are not necessarily unreactive with various surfaces on the ground. The reaction of particulate matters on the ground surfaces, which may affect the deposition velocity of particulate matters, is an important theme in future studies.

Earnest trials to revise the estimation of deposition velocity of particulate matters have recently been increasing (e.g., Zhang et al., 2001; Pryor, 2006; Pryor et al., 2008). It is desired strongly that relevant studies will progress in East Asia.

4.4.4 Evaluation of the inferential method for total deposition

A method for direct measurement requires instruments of high expense such as ultrasonic anemometer-thermometer. A comparison between the inferential method and the throughfall method (e.g., Zimmermann et al., 2006; Schmitt et al., 2005) is a cost-effective way to evaluate the inferential method; however, this procedure is limited to forest vegetation. In this procedure, the required measurements other than the inferential method related are wet-only deposition, throughfall, and stemflow. It is, therefore, desired that this procedure should be performed at a site where monitoring both for wet and dry deposition is conducted. This procedure handles the total deposition, i.e., the sum of wet and dry deposition in addition to the canopy exchange. Furthermore, the fraction of dry deposition is the sum of dry deposition of gases and particles. For example, the total deposition of ammoniacal nitrogen composes of the wet deposition of ammonium ion, the dry deposition of ammonia, the dry deposition of particulate ammonium, and the canopy exchange of ammonium ion.

The fraction of dry deposition to the total deposition of component i is expressed by (modified from Schmitt et al., 2005),

$$DD(i) = TD(i) - WD(i) \quad (4.27)$$

$$TD(i) = TF(i) + SF(i) - CE(i) \quad (4.28)$$

where, DD , TD , WD , TF , SF , and CE denote the dry deposition, total deposition, wet-only deposition, throughfall, stemflow, and canopy exchange (a positive value indicates uptake). WD , TF , and SF are obtained by observation, whereas CE should be estimated by an adequate model, e.g., the canopy budget model (EC-UN/ECE, 2001); however, the canopy exchange of sulfate and nitrate is assumed negligible in the canopy budget model. In this regard, the main target substance for the canopy exchange model is ammoniacal nitrogen.

A simple way of the canopy budget model, which excludes weak acid leaching, is as

follows (modified from EC-UN/ECE, 2001; please refers to the original report for the model including weak acid leaching):

$$BC_{TD} = \frac{Na_{TF} + Na_{SF}}{Na_{WD}} BC_{WD} \quad (4.29)$$

$$NH_{4TD} = NH_{4TF} + NH_{4SF} + NH_{4CE} \quad (4.30)$$

$$NH_{4CE} = \frac{NH_{4TF}}{NH_{4TF} + H_{TF} \cdot xH} BC_{CE} \quad (4.31)$$

$$BC_{CE} = BC_{TF} + BC_{SF} - BC_{TD} \quad (4.32)$$

where, BC denotes Ca, Mg, and K (as equivalent concentration), xH , an efficiency factor of H in comparison to NH_4 (= 6).

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5. Future direction of dry deposition flux estimation

This manual recommends available and practical method for dry deposition flux estimation taking present situations into account. In order to elaborate the method in the future, the following items should be considered.

- For atmospheric stability estimation, available method is selected based on Chapter 2.2.5 at first. After that most practical method should be determined taking the experiences of the first attempt into account.
- Suitable sites to estimate dry deposition should be selected taking the experiences of the first attempt into account. Criteria of the site selection should be determined taking into account the step-wise approaches defined by the Strategy Paper for Future Direction of Dry Deposition Monitoring of EANET (Acid Deposition Monitoring Network in East Asia, 2005, 2010).
- Reference height should be reconsidered taking into account advantage and disadvantage of recommended height (10 m).
- The parameterization in Chapter 4 should be updated based on future studies on comparison with direct measurement method, introduced in Appendix I, at special sites defined by the Strategy Paper for Future Direction of Dry Deposition Monitoring of EANET (Acid Deposition Monitoring Network in East Asia, 2005). Especially the studies on aerosols and nitrogen compounds should be encouraged, because of their large uncertainties. Advanced deposition velocity estimation by use of a multilayer numerical model would be considered for the next step of deposition velocity estimation.
- In order to elaborate setting of seasonal categories (SC), roughness length (z_0) and zero-plane displacement height (d), LAI data from satellite remote sensing could be used (see Appendix II).

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Appendix I Direct measurement for determining dry deposition flux

Dry deposition can be measured explicitly by employing the natural surface, the surrogated surface, or the micrometeorological instrument to determine the flux of the material. Analysis of material deposited on natural surfaces represents a method to infer dry deposition rates especially when micrometeorological methods are difficult to apply i.e. the complex terrain, forest edges (Seinfeld and Pandis, 1998). The surrogate surface method is to use filter substrates to collect depositing material. The method is most suitable when the aerodynamic resistance is prevailed. Measurement of fluxes by micrometeorological approach can be carried out either by direct or indirect methods. The direct methods, known as eddy correlation, require simultaneous measurements of rapid fluctuations of vertical windspeed and the eddy concentration. The product of the magnitude of the fluctuations is a direct measurement of the instantaneous flux at the point. The indirect methods rely on the measurement of vertical gradients of the mean concentration of the depositing material, and relating these quantities to the flux. They are usually referred to as the aerodynamic gradient and Bowen ratio methods. The following section describes differences of various direct measurement methodologies.

AI.1 Natural surface method

Throughfall and stemflow

The quantity and quality of precipitation as it passes through vegetative cover are important components of nutrient budget studies. Water dripping from leaves and branches, and falling through gaps in the canopy is referred to as throughfall, whereas water running down tree trunks is called stemflow. Both throughfall and stemflow methods have been used also to measure the amounts of total atmospheric deposition. The difference between the total deposition and the open field wet deposition provides information on the dry deposition.

The throughfall and stemflow methods have some advantages over the micrometeorological methods in several aspects: 1) the methods are more suitable to measure the atmospheric deposition in a complex terrain whereas the micrometeorological method requires a continuous flat land to ensure the constant flux; 2) the throughfall and stemflow contain all gases and particles deposited, including coarse particles; 3) the throughfall and stem flow methods allow fairly easy continuous monitoring, and thus provide the opportunity to study deposition processes; and 4) the throughfall and stem flow methods are less expensive than micrometeorological measurements and relatively easy to implement. For this reason, the throughfall method

is well suited for monitoring at a large number of sites (Erisman and Draaijers, 1995).

The throughfall and stemflow methods also have several weak points. The methods are related to the large spatial variability of fluxes, usually observed within forest stands, and hence the deposition estimates will be obscured (Beier et al., 1992). The presence of litterfall, insects, pollen, bird excrements in the throughfall and stemflow will affect the chemical contents in the collected rain water and hence alter the true causes of acid deposition. Figure AI.1 presents the technique of throughfall and stemflow sample collection.



Throughfall collection¹



Stemflow collection²

Figure AI.1 Measurement of acid deposition by throughfall and stemflow methods.

Source 1: http://www.fs.fed.us/psw/topics/air_quality/resin_collectors/index.shtml

Source 2: <http://www.ucd.ie/ferg/Research/Projects/FOREM/Stemflow.html>

AI.2 Surrogate surface method

The surrogate surface method is to use flat plates, petridishes or other devices intended to represent natural surfaces to collect depositing material. The approach is most suitable when the aerodynamic resistance is the dominant resistance. If other resistances such as canopy resistance are significant, a surrogate surface is unlikely to represent the correct behavior of the natural surface of interest.

The surrogate surface is mostly impregnated with substrates in order to enhance its effectiveness in capturing particle and gases. This is to prevent rebounding of the

particle captures and re-evaporating of depositing gases.

The wide range of dry deposition rates estimated from the variety of deposition surfaces emphasizes the uncertainty of the surrogate surface techniques. In spite of these limitations, surrogate surfaces provide an estimate of sulfate flux rates not currently obtainable from natural surfaces. Figure A1.2 depicts the experimental set up to collect dry depositions near the small industrial areas in the northern Thailand.



Figure A1.2 Measurement of dry deposition using surrogate surface in the northern Thailand.

A1.3 Micrometeorological method

Surface methods and micrometeorological methods are distinguished, in part, by the spatial scales over which they are representative. Surface methods yield dry deposition fluxes representative of the spatial scales of the foliage or surrogate surface element sampled, typically on the order of fractions of a square meter. If these surface elements dominate the overall surface, they may account for more area than just the local area where the measurement was made. The micrometeorological methods most commonly used provide data that are representative of the flux over a larger spatial scale than that associated with a surface method. This is because the turbulent eddies responsible for the flux inherently include a degree of spatial averaging of the conditions in the vicinity of the measurement site. The nondivergence assumption depends on the degree of variability within this vicinity. The micrometeorological methods generally meet the nondivergence criterion more frequently than do surface sampling methods (Seinfeld

and Pandis, 1998).

The flux of the total system may be determined by relationship between air concentrations and meteorology. A flat homogeneous terrain is required for the constant flux layer over a canopy to exist. Over a uniform stand of level vegetation, fluxes of momentum, heat, water vapor and any other entrained gas are constant with height. Bulk rates of exchange between the canopy and the air flowing over it can be determined by measuring vertical fluxes in this part of the boundary layer (Moneith and Unsworth, 1999). Several micrometeorological measuring methods exist for measuring dry deposition. The eddy correlation, eddy accumulation, aerodynamic gradient and Bowen ratio methods are summarized here. Figure A1.3 shows the micrometeorological instruments installed at a tower for measuring the deposition flux.

Eddy correlation method

Eddy correlation is the most common of the direct micrometeorological techniques to measure dry deposition rates. This method relies on measurements of the vertical wind velocity, w and the concentration, C of the gas or particles. The deposition flux can be considered as the sum of two components, the product of mean vertical wind speed \bar{w} and concentration \bar{C} and the fluctuating components about the means of the same quantities, $\overline{w'C'}$. Hence, the instantaneous heat flux is

$$F = \bar{w}\bar{C} + \overline{w'C'} \quad (\text{AI.1})$$

where w' and C' are the instantaneous vertical wind velocity and the departure from the mean concentration, respectively. This technique requires fast-response instrumentation i.e., the sonic anemometer and gas analyzer to resolve the turbulent fluctuation that contributes primarily to the vertical flux. These requirements are particularly severe under stable conditions where response times on the order of 0.2 s or less may be required. In practice, it is often possible to use somewhat slower instruments and apply various corrections to the computed fluxes as compensation (Seinfeld and Pandis, 1998). The deposition velocity can be obtained from the measured value of F by dividing the mean concentration at a chosen reference height.

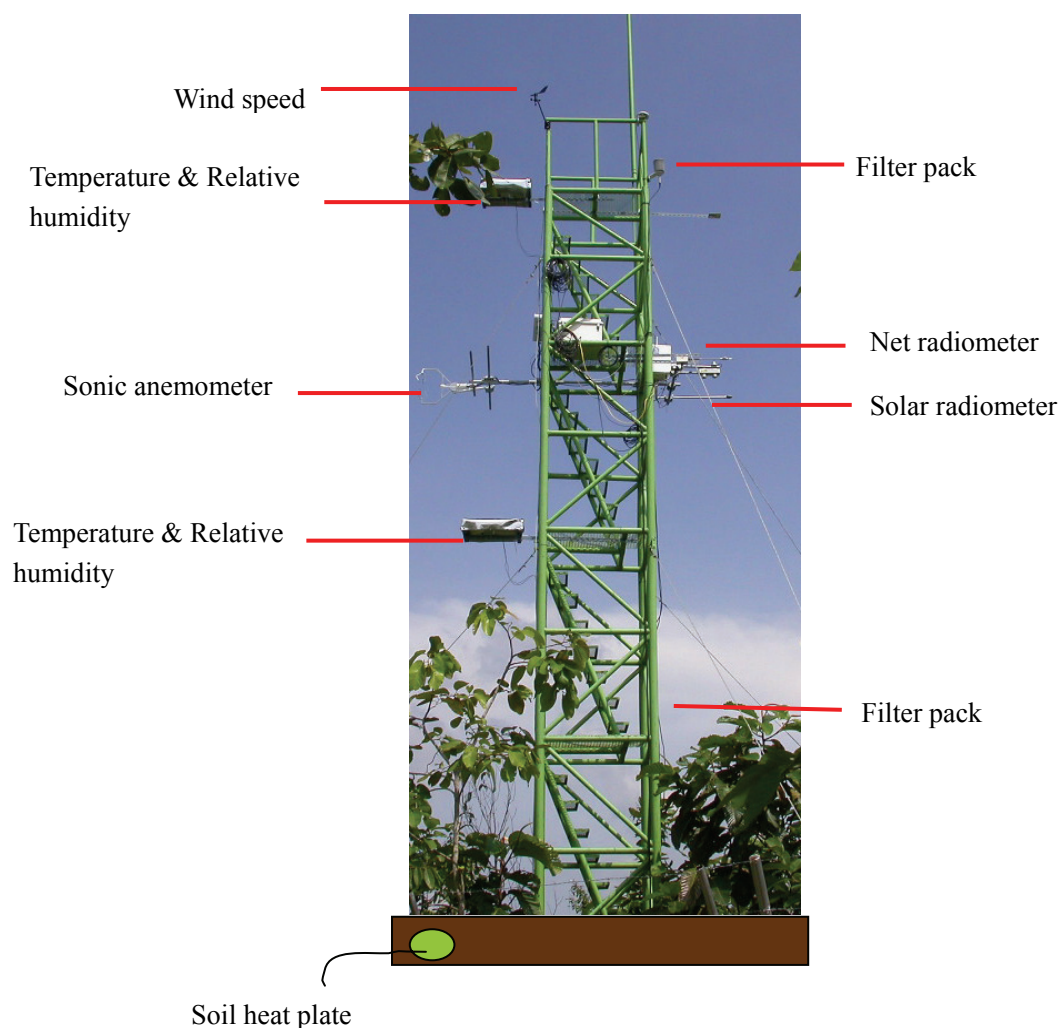


Figure AI.3 Micrometeorological instruments installed at a tower for measuring the deposition flux in Ratchaburi site, Thailand.

Relaxed eddy accumulation method

Eddy accumulation depends on essentially the same conditions and assumptions as those for eddy correlation. In this method, air is collected on two separate filters, with the vertical velocity determining which filter receives the sampled air (Hicks and McMillen, 1984, Myles et. al, 2007, Zhu et. al, 2000). One filter is used for positive vertical velocities and the second is used for negative vertical velocities; the instantaneous sampling rate for each filter is proportional to the magnitude of the velocity. The deposition flux of the aerosol can be calculated by

$$F = b\sigma_w (C_{\text{up}} - C_{\text{down}}) \quad (\text{AI.2})$$

where b is the experimental coefficient obtained from the probability distribution of vertical wind velocity, σ_w is the standard deviation of the vertical wind velocity and C_{up} and C_{down} are the average concentration of the pollutant depending on the wind velocity is upflow or downflow. The filter is then analyzed for the species of interest and the results are used to calculate the net flux (Businger, 1986).

Aerodynamic gradient method

In the aerodynamic gradient method, the flux is determined by measuring the vertical concentrations gradient and the meteorological variables at a height above the canopy. The following derivation to determine the flux by aerodynamic gradient method is obtained from Erisman and Draaijers (1995). For the mass flux,

$$F = K_c \frac{\partial C}{\partial z} \quad (\text{AI.3})$$

where K_c is diffusion coefficient for turbulent transfer in air. For the momentum transport of the deposition species,

$$\tau = K_m \partial(\rho u) / \partial z \quad (\text{AI.4})$$

where τ is the shear stress or momentum flux. It is defined as the drag force per unit area of a horizontal plane caused by horizontal air motion.

For sensible heat flux (H),

$$H = K_h \frac{\partial(\rho c_p T)}{\partial z} \quad (\text{AI.5})$$

In neutral stability, $K_c = K_m = K_h$ and consequently,

$$\frac{\rho(\partial u / \partial z)}{\tau} = \frac{\rho c_p (\partial T / \partial z)}{H} = \frac{(\partial C / \partial z)}{F} \quad (\text{AI.6}).$$

The shear stress, τ is related to the air density and the effectiveness of vertical turbulent exchange in the air flow over the surface: $\tau = \rho u_*^2$.

The eddy velocity or friction velocity associated with the momentum flux is u_* . In neutral stability, u_* can be estimated from the wind profile alone, and so the gradient method requires only two sets of profiles: concentration of deposition species at series of heights above the canopy, and wind speed measured at identical heights. The friction velocity is found from the wind profile by differentiating the wind profile equation,

$$\frac{u_*}{k} = \frac{\partial u}{\partial [\ln(z-d)]} \quad (\text{AI.7})$$

where k is thermal conductivity constant of air. Hick defined the term u_* in terms of gradient theory,

$$u_* = L \frac{\partial u}{\partial z} \quad (\text{AI.8})$$

where L is the mixing length for momentum, or rather the effective eddy size, at level z . The value for L may be given by,

$$L = \frac{\kappa(z-d)}{\phi_m} \quad (\text{AI.9})$$

where κ is the Von Karman constant, established experimentally to be about 0.41 (Pasquill and Smith, 1983) ϕ_m is the empirically estimated dimensionless correction for stability effects upon this ratio, while d is the zero displacement height.

U_* is derived as

$$u_* = \frac{(z-d)}{\phi_m} \frac{\partial u}{\partial z} \quad (\text{AI.10})$$

The eddy diffusivity K_m may be found from

$$K_m = \frac{\kappa(z-d)u_*}{\phi_m} \quad (\text{AI.11})$$

This equation may be used to estimate K_c , given the similarity between K_m , K_h and K_c . For K_c , ϕ_h is used rather than ϕ_m . Thus given the equality of ϕ_h and ϕ_c :

$$K_c = \frac{\kappa(z-d)u_*}{\phi_h} \quad (\text{AI.12})$$

which may be substituted into the mass flux, yielding

$$F = \frac{\kappa(z-d)u_*}{\phi_m} \frac{\partial C}{\partial z} \quad (\text{AI.13})$$

The eddy concentration can be defined as

$$C_* = \frac{\kappa(z-d)}{\phi_h} \frac{\partial C}{\partial z} \quad (\text{AI.14})$$

and thus the mass flux becomes,

$$F = u_* C_* \quad (\text{AI.15})$$

As a result, the flux of a pollutant may be derived from information on the wind profile, the concentration gradient and the effect of stability. The stability function is a correction for the departure of the neutral profile. Under the neutral conditions $\phi_m = \phi_h = \phi_c = 1$. The stability correction is a function of height. Therefore ϕ should be included in the integration. In the literature, results obtained by Dyer and Hicks (1970) are widely used (e.g. Thom, 1975; Denmead, 1983). Under stable conditions,

$$\phi_m = \phi_h = \phi_c = 1 + 5.2 \frac{(z-d)}{L} \quad (\text{AI.16})$$

and unstable conditions, ϕ_h and ϕ_c are represented by the square of ϕ_m ,

$$\phi_m = \phi_h = \phi_c = \left[1 - 16 \frac{(z-d)}{L} \right]^{-0.5} \quad (\text{AI.17})$$

where L is the Monin-Obukhov stability length used as stability parameter ($L > 0$:

stable; $L < 0$: unstable; $|L| \rightarrow \infty$: neutral), given as

$$L = \frac{-T\rho_a c_p u_*}{\kappa h H} \quad (\text{AI.18})$$

where T is the absolute temperature and g , the acceleration of gravity. The sensible heat flux H can be calculated from the net radiation using the Priestly-Taylor model parameterized (modified by Holtslag and De Bruin, 1988). This modified model was tested using experiments as a meteorological mast and Cabauw in the center of the Netherlands. The model was used in subroutines for the calculation of H , L and u_* by Beljaars et. al. (1987).

Integration between the roughness length z_0 and z , yields:

$$u_* = \frac{\kappa u}{\ln\left(\frac{z-d}{z_0}\right) - \Psi_m\left(\frac{z-d}{L}\right) + \Psi_m\left(\frac{z_0}{L}\right)} \quad (\text{AI.19})$$

$$\text{and} \quad C_* = \frac{\kappa C}{\ln\left(\frac{z-d}{z_0}\right) - \Psi_h\left(\frac{z-d}{L}\right) + \Psi_h\left(\frac{z_0}{L}\right)} \quad (\text{AI.20})$$

$$\text{where} \quad \Psi_m\left(\frac{z-d}{L}\right) = \Psi_h\left(\frac{z-d}{L}\right) = -5.2 \frac{(z-d)}{L} \quad (\text{AI.21})$$

for stable conditions and

$$\Psi_m\left(\frac{z-d}{L}\right) = 2 \ln\left(\frac{1+x}{s}\right) + \ln\left(\frac{1+x^2}{2}\right) + \ln\left(\frac{1+x^2}{2}\right) - 2 \arctan(x) + \frac{\pi}{2} \quad (\text{AI.22})$$

$$\Psi_h\left(\frac{z-d}{L}\right) = 2 \ln\left(\frac{1+x^2}{2}\right) \quad (\text{AI.23})$$

$$x = \left[1 - 16 \frac{(z-d)}{L} \right]^{0.25} \quad (\text{AI.24})$$

For unstable conditions, $\Psi_m((z-d)/L)$ is the integrated stability correction for momentum and $\Psi_h((z-d)/L)$ is the integrated stability correction for heat. Whereas the gradient method is theoretically straightforward, it requires relatively accurate concentration values at two or more heights, since the difference between such values can be very small if the deposition rate is small. For example, for a deposition velocity of 0.2 cm s^{-1} and u_* of 0.4 m s^{-1} , the concentrations at 2 and 4 m above the surface will differ by less than 1% under neutral conditions. The difficulty of achieving such relative accuracy can be addressed by using a single detector for the species of interest, thereby eliminating inter-instrument differences, to sample the air at different heights, for example, with a movable sample probe or with a mechanism that switches between sampling lines. The gradient method tends to be impractical over extremely rough surfaces, because the measuring heights should satisfy the criterion $z / z_o \gg 1$; however, that condition can actually place the measurement above the constant-flux layer. In such a case, the turbulent diffusivity based on the gradient-transport assumption may then be poorly known.

Bowen ratio method

The Bowen ratio method for flux measurement is derived from the energy balance above the canopy,

$$R_n - G = 1 + \lambda E \quad (\text{AI.25})$$

Where R_n is the net radiation, G is the soil heat flux, λ is the latent heat of vaporization of water and E is the flux of water vapor per unit area.

It can be rewritten in the form

$$\lambda E = (R_n - G) / (1 + \beta) \quad (\text{AI.26})$$

where β is the Bowen ratio equivalent to $(1/\lambda E)$. The net radiation of $(R_n - G)$ and β is obtained from measurements of temperature and vapor pressure at a series of heights within the constant flux layer. Assuming that the transfer coefficients of heat and vapor are equal,

$$\beta = 1 / \lambda E = \gamma \partial T / \partial e \quad (\text{AI.27})$$

and $\partial T / \partial e$ is found by plotting the temperature at each height against vapor pressure at the same height.

By writing the heat balance equation as

$$R_n - G = -D_w \rho_a C_p (\partial T / \partial z) - D_H \rho (\partial e / \partial z) \quad (\text{AI.28})$$

or

$$R_n - G = D \rho_a C_p (\partial T_e / \partial e) \quad (\text{AI.29})$$

where D is a turbulent transfer coefficient and T_e is the equivalent temperature of $(T + (e / \gamma))$. The Bowen ratio equation can be derived from following equation.

$$T_e = T + (e / \gamma) \quad (\text{AI.30})$$

By writing the sensible heat flux as $1 = -D_H \rho_a C_p (\partial T / \partial z)$ and forming similar expressions relating the heat flux λE to $\gamma^{-1} (\partial e / \partial z)$ and the flux F of any other gas to $\partial C_s / \partial z$, it can be presented with equation,

$$L = (R_n - G) (\partial T / \partial T_e) \quad (\text{AI.31})$$

$$\lambda E = [(R_n - G) (\partial T / \partial T_e)] / \gamma \quad (\text{AI.32})$$

$$F = [(R_n - G) (\partial T / \partial T_e)] / \rho C_p \quad (\text{AI.33})$$

In the addition, the flux, F can be written in form of the absolute gaseous concentration C ,

$$F = [(R_n - G) / \rho_a C_p] (\partial C_s / \partial T_e) \quad (\text{AI.34})$$

$$\Delta T_e = \Delta (T + e / \gamma) \quad (\text{AI.35})$$

$$\begin{aligned} \Delta T_e &= \Delta T + \Delta [e / (C_p P / \lambda E)] \\ &= \Delta T + \Delta (\lambda E e / C_p P) \\ &= \Delta T + (\lambda E e / C_p P) / C_p P \end{aligned} \quad (\text{AI.36})$$

$$F = [(R_n - G)(C_{s1} - C_{s2})] / [\rho_a C_p [\Delta T + (\lambda \varepsilon / C_p P) \Delta e]] \quad (\text{AI.37})$$

$$F = [(R_n - G)(C_{s1} - C_{s2})] / [\rho_a C_p \Delta T + (\lambda \varepsilon / C_p P) C_p P \Delta e] \quad (\text{AI.38})$$

$$F = [(R_n - G)(C_{s1} - C_{s2})] / [\rho_a \lambda (0.622 / P)(e_1 - e_2)] + [\rho C_p (T_1 - T_2)] \quad (\text{AI.39})$$

By Fick's law, $F = -v_d \bar{C}$ and, thus $F = -D \Delta C / \Delta z$ (AI.40)

Therefore, $D_{1-2} = (R_n - G) / [\rho_a \lambda (0.622 / P)(e_1 - e_2)] + [\rho_a C_p (T_1 - T_2)]$ (AI.41)

where D_{1-2} is the transport velocity between two heights.

$$D_{1-2} = (R_n - G) / [\rho_a \lambda \Delta e (0.622 / P) + \rho C_p \Delta T] \quad (\text{AI.42})$$

where 0.622 is a ratio of the molecular weight of water to the molecular weight of air, P is a total pressure; and C_{s1} and C_{s2} are the absolute gaseous concentrations at height z_1 and z_2 , respectively.

The Bowen ratio methods of flux calculations are generally applied to measurements averaging for periods of a half to one hour. Fluctuations in the measuring parameters, especially on a day of intermittent cloud cover, will affect the estimation of mean fluxes for shorter periods. The diurnal changes make time-averaging objectionable for periods of more than two hours, especially near sunrise and sunset (Moneith and Unsworth, 1999).

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Appendix II Use of remotely sensed information

AII.1 Use of remote sensing

Land Use/Land Cover (LU/LC) data obtained by satellite remote sensing can express surface characteristics which will be used for dry deposition flux estimation. In this chapter, resources of LU/LC data and important parameters for calculation of dry deposition flux will be introduced.

Some of LU/LC datasets can be obtained via the Internet. The two major resources of LU/LC dataset are listed below.

Advanced Very High Resolution Radiometer (AVHRR)

AVHRR is an electromagnetic radiation sensor mounted on polar orbiting satellites operated by National Oceanic and Atmospheric Administration (NOAA). The AVHRR instrument measures the reflectance of the Earth in 5 spectral bands. The first two are located in the visible (0.6 micrometer) and near-infrared (0.9 micrometer) regions, the third one is located around 3.5 micrometer, and the last two monitor thermal radiation emitted from the earth and are located around 11 and 12 micrometers. The highest ground resolution that can be obtained from the AVHRR is 1.1 km. AVHRR data have been collected continuously since 1981. Initially, the NOAA/AVHRR satellites were designed to observe the Earth's weather in the form of cloud patterns. However, further researches on the sensors clearly demonstrated that they could be used for more than just monitoring weather phenomena. Today the AVHRR are used in many applications such as monitoring land-surface processes and other characteristics of the Earth.

Institute of Industrial Science (IIS) at the University of Tokyo has been receiving the AVHRR data in Tokyo, Japan since 1984, and Asian Institute of Technology in Bangkok, Thailand since 1997. The combined AVHRR datasets cover whole East Asian region. Raw data of the AVHRR can not be freely downloaded because of their enormous data size. Furthermore, the raw data have to be conducted data processing such as general array processing operations, reading and writing properly formatted files, conversion of radiometer counts to radiance, reflectance and brightness temperature and mapping data from the satellite coordinate system to standard geographic coordinates. IIS developed an AVHRR data processing system, named as PaNDA, on the Web site. Using this system enables us to process the AVHRR data on the Web site with a few input parameters and download the processed data by FTP access. The URL of PaNDA is <http://webpanda.iis.u-tokyo.ac.jp/index.php>.

Moderate-resolution Imaging Spectroradiometer (MODIS)

MODIS is a payload scientific instrument launched into an Earth orbit by National Aeronautics and Space Administration (NASA). It is on board the Terra satellite in 1999 and the Aqua satellite in 2002. Terra's orbit around the Earth is timed so that it passes from north to south across the equator in the morning, while the Aqua passes south to north over the equator in the afternoon. The Terra MODIS and the Aqua MODIS are viewing the entire Earth's surface 1 or 2 times a day, which enables more frequent monitoring than AVHRR. The MODIS instrument acquire data in 36 spectral bands ranging in wavelength from 0.4 μm to 14.4 μm and at varying spatial resolutions (2 bands at 250 m, 5 bands at 500 m and 29 bands at 1 km). They are designed to provide measurements in large-scale global dynamics including changes in Earth's cloud cover, radiation budget and processes occurring in the oceans, on land, and in the lower atmosphere. The detailed information of MODIS is provided at the NASA Web site (<http://modis.gsfc.nasa.gov/index.php>).

IIS has been retrieving also MODIS dataset in Tokyo and Bangkok since 2001, and it covers whole East Asian region. As well as AVHRR data processing system (PaNDA), IIS also developed an MODIS data processing system including following functions: spectral subset (250 m, 500 m and 1000 m resolutions), radiometric correction to radiance, spatial subset of geo-referenced data as a rectangular area with latitude-longitude grid system in HDF format and generation of a quick look file in JPEG format. Using this system enables us to process the MODIS data on the Web site with a few input parameters and download the processed data by FTP access. The URL of WebMODIS site is <http://webmodis.iis.u-tokyo.ac.jp/index.php>.

AII.2 How to calculate Normalized Difference of Vegetation Index (NDVI)

The AVHRR and MODIS sensor is also a useful tool for monitoring vegetation, land cover, and climate, and enables scientists to observe how these three elements interact. These data can be used to assess the quantity and vigor (photosynthesis activity) of vegetation through a measure of "greenness", referred to as the vegetation index or the Normalized Difference Vegetation Index (NDVI). Using AVHRR and MODIS, one can monitor the growing season of crops, which will change with variations in regional climates, and which can also provide potentially life saving information to developing countries that heavily rely on an abundant and reliable harvest. From AVHRR data, it is relatively easy to identify green vegetation and non-vegetated features such as water, barren land, ice, snow, and clouds.

NDVI is a non-linear transformation of the visible (red) and near-infrared bands of

satellite information. NDVI is defined as the difference between the visible (*red*) and near-infrared (*nir*) bands, over their sum, as shown in the equation AII.1. NDVI is an alternative measure of vegetation amount and condition. It is associated with vegetation canopy characteristics such as biomass, leaf area index and percentage of vegetation cover.

$$\text{NDVI} = (\text{nir} - \text{red}) / (\text{nir} + \text{red}) \quad (\text{AII.1})$$

For vegetation monitoring, the NDVI obtained by the combination of Channels 1 (0.54–0.68 μm for AVHRR, 0.62–0.67 μm for MODIS) and 2 (0.725–1.10 μm for AVHRR, 0.841–0.876 μm for MODIS) expressed as $[(Ch_2 - Ch_1)/(Ch_2 + Ch_1)]$, and respective visible and near infrared data of AVHRR and MODIS are commonly used. The NDVI is representative of plant assimilation condition and of its photosynthetic apparatus capacity and biomass concentration (Groten, 1993; Loveland et al., 1991). In particular vegetation index dynamics in time are correlated with the canopy Leaf Area Index (LAI) and other functional variables (Cihlar et al. 1991). These variables are strongly conditioned by the behavior of precipitation, temperature and daily radiation of the observed area (Davenport et al., 1993). Vegetation index, therefore, is representative of plants' photosynthetic efficiency, and it is time varying due to changes in meteorological and environmental parameters. The NDVI values range from -1 to $+1$ (equivalent to pixel values of 0–255).

AVHRR data is particularly suited to monitoring seasonal and inter-annual changes in land cover/land use because of its low cost and temporal and spatial characteristics. There have been a number of studies which have directly linked AVHRR-NDVI to plant phenology (DeFries 1995; Reed et al., 1994). For instance, the number of periods when the NDVI exceeded a threshold might indicate the number of growing seasons, the time integrated NDVI might indicate gross primary production and the length of the period when NDVI exceeded a threshold might indicate the length of the growing season. Seasonal and inter-annual variations can be derived from multi-temporal series of NDVI that can be associated with other ecological variables (Mora and Iverson 1995).

AII.3 How to calculate Leaf Area Index (LAI) from the data of NDVI

Leaf Area Index (LAI) is defined as the leaf area per unit ground area. LAI is a factor that indicates how many leaf (or photosynthetically active) surfaces are in a column extended from, the ground area under the canopy diameter, up through the canopy. LAI is the necessary index to calculate dry deposition velocity as shown in Chapter 4.2.

LAI can be estimated from NDVI because NDVI represent the relative seasonal

changes in vegetation rather than vegetation amount. There is a significant relationship between NDVI and LAI. Assuming that NDVI/LAI relationship is linear (Wiegand C.L. 1979, Tucker 1980, Wardley and Curran 1984), and the maximum NDVI value in a season corresponds to the maximum LAI of vegetation cover (Justice 1986). LAI can be inferred from NDVI as (Zhangshi and Williams 1997) the following equation.

$$LAI_i = LAI_{\max} * (NDVI_i - NDVI_{\min}) / (NDVI_{\max} - NDVI_{\min}) \quad (AII.2)$$

Where max, min and 'i' are the maximum, minimum and period values observed, respectively.

The maximum and minimum NDVI values can be determined by multi-temporal NDVI observations from the AVHRR sensor. The formulation of LAI as a fraction of the maximum NDVI observed in a season facilitates the integration of data from different sensors. High and coarse resolution satellite observations can be combined to get more reliable estimates of LAI patterns in a landscape.

LAI_{\max} can be determined empirically by assigning different values to a land cover categories, and LAI_i can be then obtained by combining NDVI information from different dates.

Even when a linear relationship between $NDVI/LAI$ is often assumed, the relationship is not always linear since the vegetation indices approach a saturation level asymptotically for LAI ranging from 2 to 6, depending on the type of vegetation cover, and environmental conditions (Clevers 1989; Carlson and Ripley 1998). However, by assuming a non-linear relationship, the LAI estimates from NDVI are then highly dependent upon certain factors such as canopy geometry, leaf and soil optical properties, sun position and cloud coverage. The variation of NDVI as a function of LAI can be expressed by the modified Beer's law (Baret and Guyot 1991) as shown below:

$$NDVI = NDVI_a + (NDVI_{bs} - NDVI_a) * \exp(-K_{ndvi} * LAI) \quad (AII.3)$$

Where $NDVI_{bs}$ is the vegetation index corresponding to that of the bare soil, $NDVI_a$ is the asymptotic value of NDVI when LAI tends towards infinity, and K_{ndvi} is the coefficient that controls the slope of the relationship (extinction coefficient).

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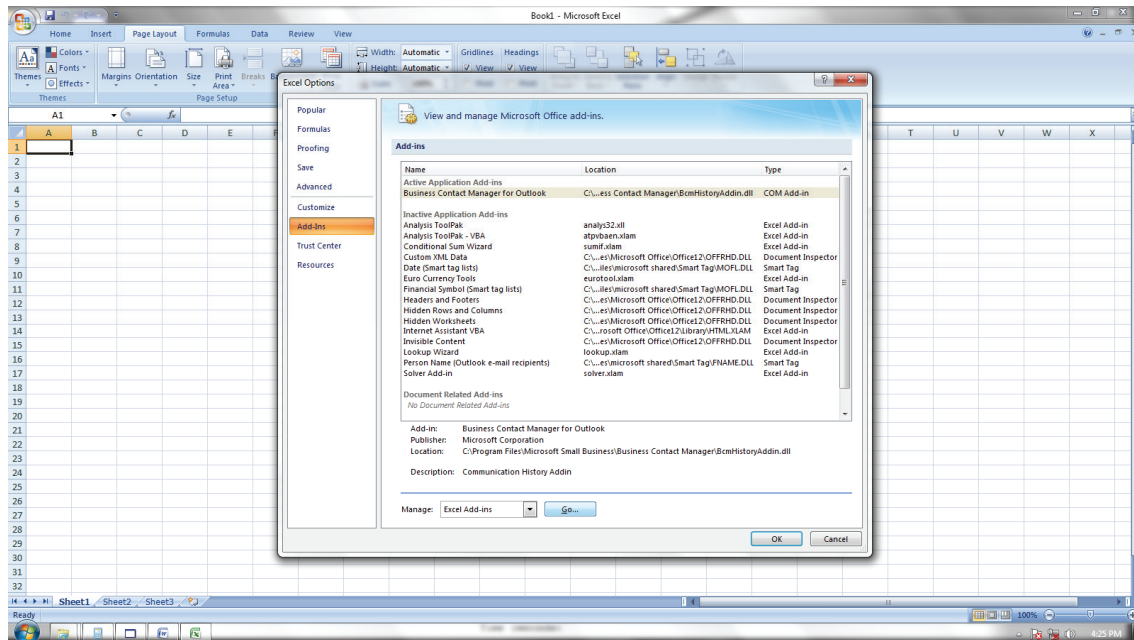
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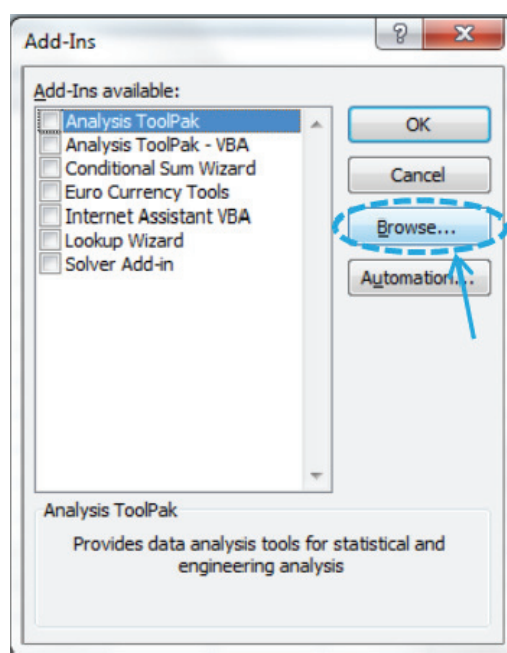
Appendix III How to operate Microsoft Excel macro program for calculation of deposition velocity

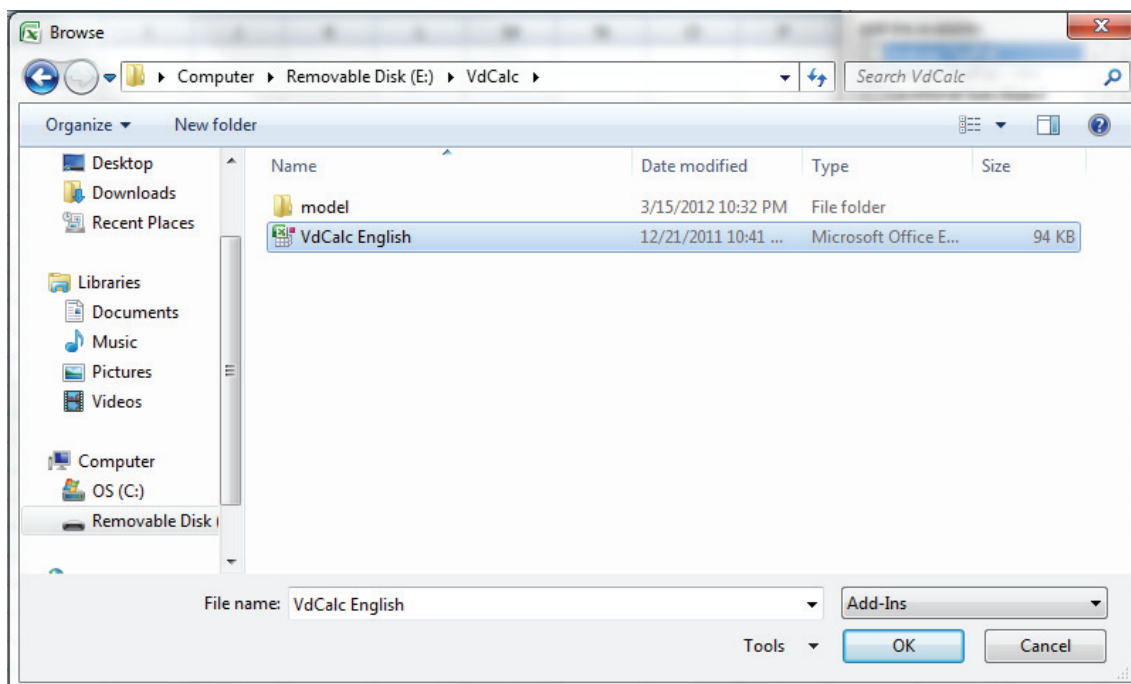
(1) Installation of Vd calc add-in program

Copy the macro package in the folder of “C:\Program Files\VdCalc”, and open the Add-Ins menu of MS Excel as shown below.

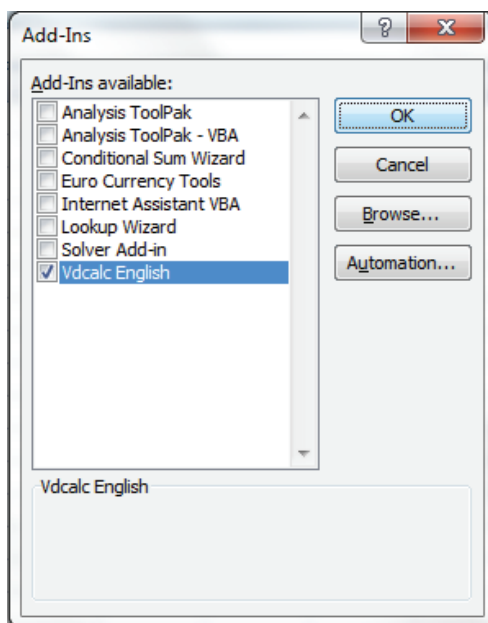


Choose browse menu, and then choose the file “C:\Program Files\VdCalc\VdCalc English.xla”.

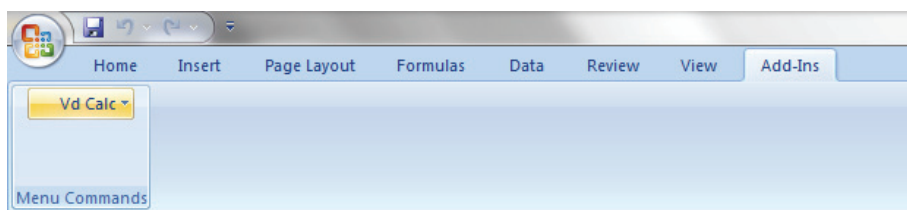




Choose Vdcalc English.

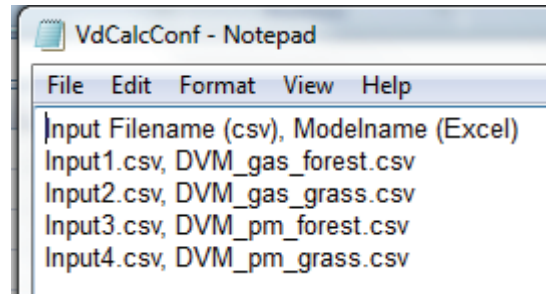


Vd Calc menu will be shown in the Add-Ins tab.



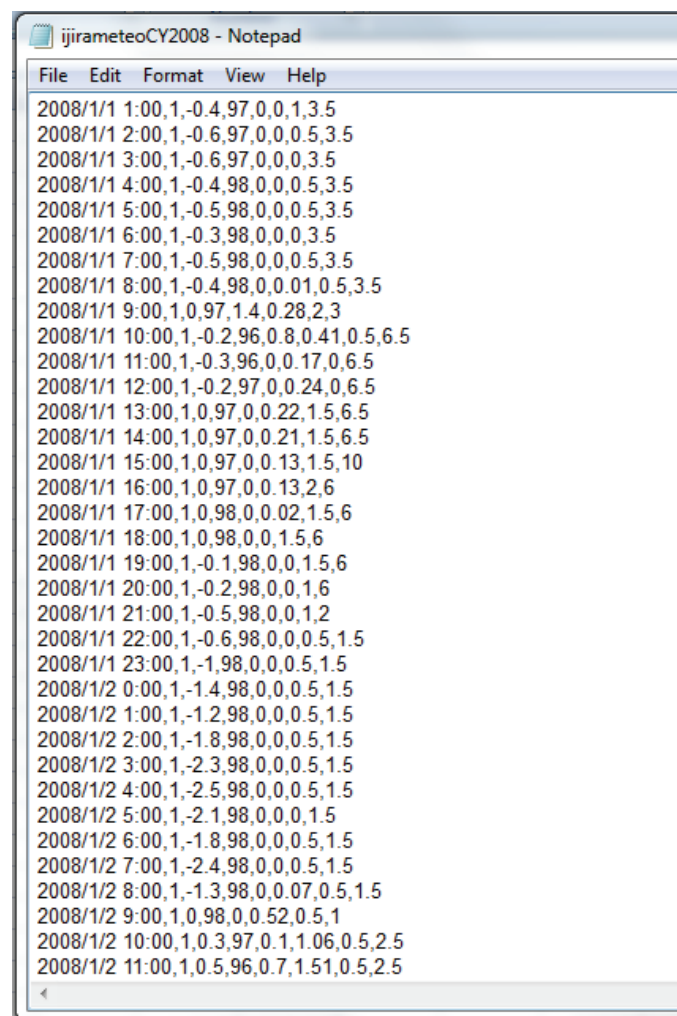
(2) Input of initial condition data

Input setting of a system definition file (VdCalcConf.csv) for calculation by the selected folder. Create “File name of input csv file” and “File name of Vd calculation model”.

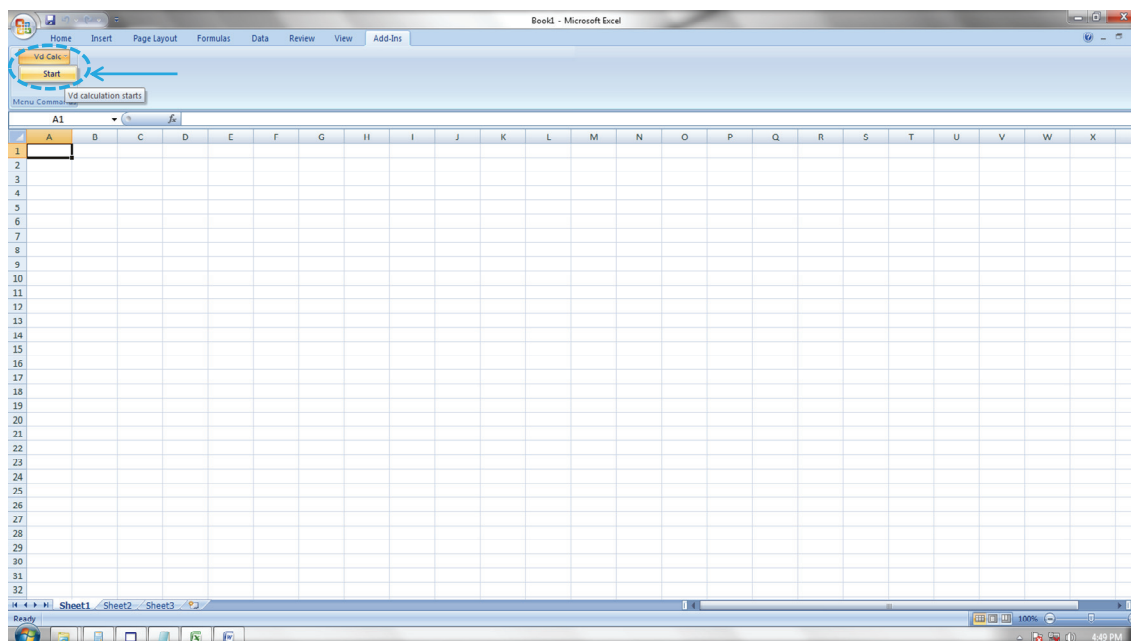


Input meteorological parameters as shown below. Each parameter should be divided by comma.

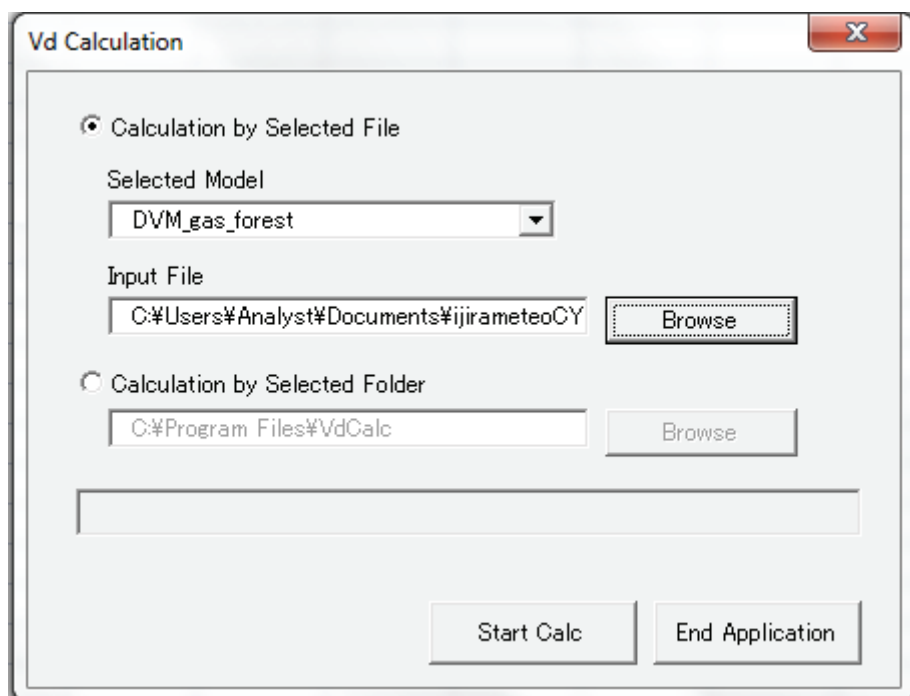
(Date and Time, Temperature [degree], Relative Humidity [%], Wind Speed [m/s], Solar radiation [MJ/m²], Precipitation [mm], Cloud coverage [0~10])



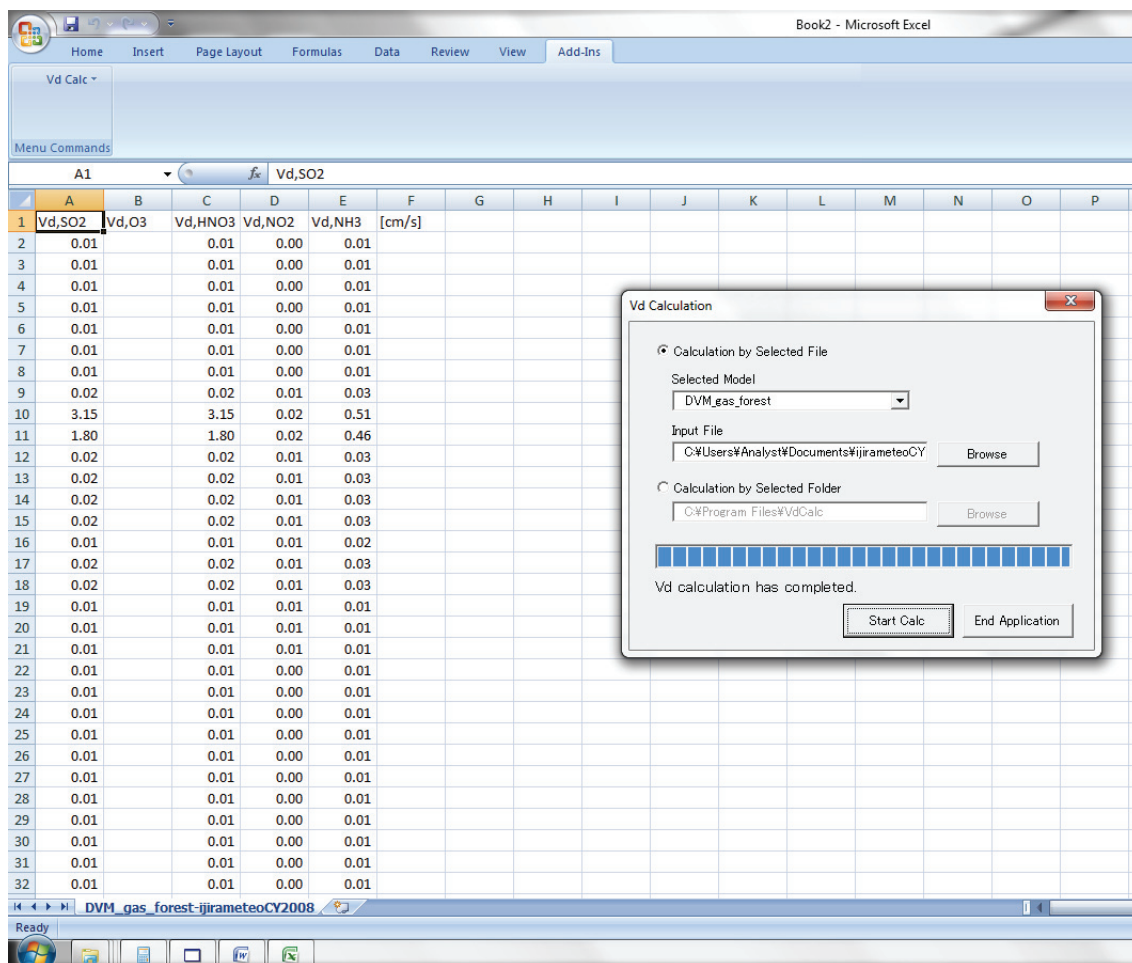
Choose the Add-Ins tab, and then choose the start icon of Vd calculation.



Choose the Vd model and input meteorological data file for calculation by the selected file or choose the folder including the system definition file for calculation by the selected folder.



When Vd calculation has completed, the below display was shown. Calculated Vd results will be obtained on hourly basis.



The Network Center will provide the MS Excel macro program for calculation of deposition velocity local staffs who want to calculate by themselves. The macro program is protected by the password, and the Network Center will distribute it upon the request.

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