DRY AEROSOL DEPOSITION OVER THE NORTH SEA ESTIMATED FROM AIRCRAFT MEASUREMENTS

Carlos M. Rojas, Philippe M. Otten and René E. van Grieken

Department of Chemistry, University of Antwerp (UIA)
Universiteitsplein 1, B-2610 Antwerp-Wilrijk
Belgium

and

Remi Laane

Tidal Waters Division, Rijkswaterstaat
P.O. Box 20907
The Hague, The Netherlands

ABSTRACT

A mathematical approach based on the Monin-Obukhov similarity theory is used to predict the wind speed, friction velocity and drag coefficient, which are then introduced in the well-known deposition model of Slinn and Slinn (1980), to calculate the dry deposition of heavy metals into the North Sea. This model is perfectly suitable for aircraft sampling considering the fact that usually due to safety reasons, flights at the reference height used in deposition models (10 m), are not possible. To check this approach, deposition velocities were calculated based on the airborne concentrations of Cu, Cd, Zn and Pb obtained by sampling with the aid of an aircraft over the Dutch continental shelf of the North Sea. Results are in agreement with those found in the literature. A rough estimation of the atmospheric input for these heavy metals and comparison with riverine inputs and direct discharges is also included.

INTRODUCTION

There has been an increasing interest in studying the fate and removal mechanisms of gases and particulate matter over the oceans both in remote areas (Arimoto et al., 1985; Buat-Ménard and Duce, 1986) and polluted areas such as the Western Mediterranean (Dulac et al., 1987; Gomes et al., 1988). In the coastal and shelf systems of the North Sea there has been a growing concern about large quantities of heavy metals being deposited from the atmosphere. Several authors, such as Van Aalst et al. (1983), concluded that the atmosphere plays an important role in the pollution of the North Sea and estimated that the input of heavy metals is comparable to that from river and direct discharges. Due to the natural limitations imposed on sample collection at sea, it has been necessary to develop mathematical models in order to estimate such heavy metal inputs. These theoretical approaches began more than 15 years ago (Sehmel and Sutter, 1974) and were reviewed by Sehmel (1980). Models for dry deposition on natural waters such as that of Slinn and Slinn (1980) have recently been used in the Western Mediterranean Sea (Dulac et al., 1987) and the North Sea (Steiger et al., 1989; Dedeurwaerder, 1988). Williams (1982) proposed another model which has been discussed by Slinn (1983) and Liss et al. (1988). In order to assess the input of heavy metals to the North Sea Van Aalst et al. (1983) and Van

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Jaarsveld et al. (1986) used a trajectory model based upon anthropogenic emisions and in-land meteorological data. Krell and Roeckner (1988) performed stochastic trajectory modeling estimates for wet and dry deposition of Pb and Cd.

In this paper we report on the use of the Monin-Obukhov (1954) similarity theory to predict the wind speed, friction velocity and drag coefficient at a certain height (usually 10 m), taking into account the air temperature, atmospheric stability, wind speed, atmospheric pressure and relative humidity, measured at higher altitude, and the sea temperature. This can be applied directly to aircraft samplings since safety reasons usually impede to fly at altitudes lower than 20 m. The calculated parameters are then introduced in Slinn and Slinn's formulation to determine the dry deposition velocity of airborne particulate matter. A preliminary estimation of dry deposition velocities and atmospheric inputs based on average concentrations of Cu, Cd, Zn and Pb for a few sampling flights on the North Sea is included.

THEORETICAL BACKGROUND

The shape of the wind profile over sea depends on the aerodynamic surface roughness length and atmospheric stability, which in turn depends on the vertical density (temperature) gradient and on wind stress. Thus, from the Monin-Obukhov (1954) similarity theory, the profiles for wind, temperature and humidity are:

$$\frac{\kappa z}{u_*}\frac{\partial u}{\partial z} = \psi_m\left(\frac{z}{L}\right) \qquad (1)$$

$$\frac{z}{t_*}\frac{\partial \theta}{\partial z} = \psi_t\left(\frac{z}{L}\right) \qquad (2)$$

$$\frac{z}{q_*}\frac{\partial q}{\partial z} = \psi_q\left(\frac{z}{L}\right) \qquad (3)$$

$$\kappa u_* t_* = -\overline{t'w'} \qquad (4)$$

$$\kappa u_* q_* = -\overline{q'w'}$$

where, z is the height, $\kappa = 0.4$ von Kármán's constant, u_* = friction velocity, t_* and q_* are the scaling temperature and humidity, respectively. The subscripts m, t and q stand for momentum, temperature and humidity; t', q' and w' are the vertical temperature, humidity and wind speed variations. The mean wind speed is denoted by u, ψ represents the stability function and L the Monin-Obukhov length.

The integration of (1), (2) and (3) yields,

$$\frac{u(z)}{u_*} = \frac{1}{\kappa}\left[\ln\left(\frac{z}{z_0}\right) - \psi_m\left(\frac{z}{L}\right)\right]$$

$$\frac{\theta(z) - T_s}{t_*} = \ln\left(\frac{z}{z_{ot}}\right) - \psi_t\left(\frac{z}{L}\right) \qquad (5)$$

$$\frac{q(z) - q_s}{q_*} = \ln\left(\frac{z}{z_{oq}}\right) - \psi_q\left(\frac{z}{L}\right)$$

where z_0 is the aerodynamic surface roughness length; $\theta(z)$ and T_s are the potential temperature of the air and sea surface, respectively and q_s is the absolute humidity of air saturation at sea surface temperature.

For the surface roughness height we used Charnock's (1955) formula, with a parameter $\alpha = 0.0185$ (Wu, 1982) instead of the original 0.011. This was motivated by the wide range of wind speeds used by Wu to determine this parameter. The resulting surface roughness length from the formulae of Charnock (1955) and Businger (1973) for smooth surfaces is:

$$z_0 = 0.0185 \frac{u_*^2}{g} + \frac{v}{9u_*} \tag{6}$$

where v is the dynamic air viscosity and g the gravitational acceleration.

Using the following parameterizations:

$$\begin{aligned} C_d(z)u^2(z) &= u_*^2 \\ C_t(z)u(z)\left(T_s - \theta(z)\right) &= -\kappa u_* t_* \\ C_\theta(z)u(z)\left(q_s - q(z)\right) &= -\kappa u_* q_* \end{aligned} \tag{7}$$

for the momentum, heat and humidity fluxes and solving iteratively (5), (6) and (7) including the expressions of Large and Pond (1982) for ψ_m, ψ_t and ψ_q for stable and unstable atmospheres and a stability parameter (z/L) given by Williams (1982), one can predict the drag coefficient and friction velocity at a given height.

For the dry deposition model we used Slinn and Slinn's (1980) formulation, including a slip correction factor for the diffusion sub-layer. Brownian diffusivity was calculated using Seinfeld's (1986) approach. The effect on particle deposition due to particle growth by condensation has been taken into account assuming that particles are either hydrophobic or hygroscopic. In the latter case particles were considered as having the same hygroscopic properties as ammonium sulphate particles and their wet diameter was calculated using the results of Fitzgerald (1975).

EXPERIMENTAL

From September 1988 to August 1989 several aircraft samplings have been carried out over the Dutch continental shelf of the North Sea, using a twin engine Piper Chieftain PA 31-350 aircraft belonging to the company Geosens BV (Rotterdam, The Netherlands). During the flights SO_2 and O_3 were monitored. Wind speed was determined with the aid of a Doppler radar, together with height above sea level. Also dew point temperature and air temperature were measured.

Each flight was scheduled based on 36-h backward trajectories provided by the Royal Dutch Meteorological Institute (KNMI), and consisted of 6 tracks equally spaced between the inversion height and the sea level. The normal flight height for the lowest track was 20 m.

Samples were collected on 0.4 µm pore-size Nuclepore filters using an isokinetic inlet located on the top of the fuselage. The flow rate was of 50 $l.min^{-1}$ and the average sampling time per track was 25 min. The collected airborne matter was first analyzed by X-ray fluorescence (XRF) using a Tracor Northern 5000 X-ray spectrometer equipped with a Rh target X-ray tube and a Si(Li) solid state detector; more details on the analysis protocol can be found elsewhere (Rojas et al., 1990). Then, samples were analyzed by Particle-Induced X-ray Emission (PIXE) using a 2.4 MeV proton beam supplied by the isochronous cyclotron of the Institute of Nuclear Sciences at the University of Ghent, Belgium. Details on this analytical technique are given by

Maenhaut (1989). The sea temperature data were supplied by Management Unit for the North Sea and Scheldt Estuary Mathematical Model, belonging to the Belgian Ministry of Public Health and Environment.

RESULTS AND DISCUSSION

In all calculations an atmospheric pressure of 1013 hPa, relative humidity of 75% for the turbulent layer and a particle density of 2.5 g/cm^3 were assumed. Deposition velocities were computed for 0.5 μm particle aerodynamic diameters, which correspond to the centroid of the size distribution of Pb particles in the North Sea reported by Steiger et al. (1989).

Table 1 summarizes the results of the model calculation for 5 different flights, with altitudes ranging from 20 to 50 m above sea level. Here only the second row corresponded to an unstable atmosphere. For the 4 cases of stable atmosphere the average difference in wind speed with respect to the reference height is 20%, with a minimum of 6% corresponding to an air-sea temperature difference of 0.4 °C. The surface roughness length values are of the same order of magnitude as those reported by Krauss (1972) for oceanic conditions. Drag coefficients varied from 0.18 x 10^{-3} for stable to 1.33 x 10^{-3} for unstable conditions, respectively. Dry deposition velocities are, as expected, a function of wind speed. The average difference in dry deposition velocity for hydrophobic and hygroscopic particles was 32%. The smallest difference (16%) was observed for the unstble case with a relatively warm sea.

Preliminary results of 4 size segregated samplings of airborne particulate matter collected with a cascade impactor (Dierck et al., 1990) and the average bulk concentration for the first 8 flights for Cu and Cd taken from Otten et al. (1989) and the average of PIXE and XRF for Zn and Pb from Dierck et al. (1990) were used to determine the average weighted dry deposition velocities for Cu, Cd, Zn and Pb, shown in Table 2. The dry deposition velocities of Cu, Cd and Pb are very close to those used by Van Jaarsveld et al. (1986) and Krell and Roeckner (1988) (0.2 cm/s). The deposition velocity for Zn is about 40% higher. Table 3 shows some rough estimates of the atmospheric input calculated for both particle categories and computed from the average concentration for tracks 1-5 (first column), for tracks 1-6 (second), while the third corresponds to track 6 (nearest to the sea). These heavy metals inputs are within the variation range of those reported by Kersten et al. (1988) for Cd, Zn and Pb in both particle categories and somewhat lower for Cu. If we assume that particles are indeed hydrophobic then, the atmospheric

Table 1. Results obtained using data corresponding to 5 flights. Here (1) and (2) stand for hydrophobic and hygroscopic particles, respectively.

Given Data				Calculated Data				
Z (m)	T°C Sea	T°C Air	u m/s	u at 10 m	z_0 (m) x 10^{-4}	C_d x 10^3	V_d (1) cm/s	V_d (2) cm/s
20	13	16	4.1	2.9	0.4	0.18	0.003	0.006
20	13	10	8.9	8.5	2	1.33	0.008	0.01
20	10.4	10.8	9.3	8.7	1.8	1.17	0.008	0.009
40	10	12	5.9	4.3	0.4	0.47	0.004	0.007
50	6.8	9.2	14	11	3.6	0.98	0.008	0.01

Table 2. Calculated dry deposition velocities (cm/s) for both particles categories.

Element	Hydrophobic particles	Hygroscopic particles
Cu	0.18	0.30
Cd	0.18	0.27
Zn	0.36	0.52
Pb	0.27	0.38

Table 3. Rough estimation of the atmospheric input (Tonnes/Y) of these heavy metals, based upon the average airborne concentration for 8 flights. Here 1-5 stand for the average from track 1-5, 1-6 for the average from track 1-6 and 6 is the average for the last track.

	Hydrophobic particles			Hygroscopic particles		
	1-5	1-6	6	1-5	1-6	6
Cu	190	180	175	320	310	290
Cd	21	21	21	31	31	31
Zn	3900	4200	5400	5700	6000	7800
Pb	2000	2300	2700	3000	3200	3900

inputs of Cu and Cd are lower compared to riverine inputs and direct discharges, whereas that of Zn lays between them. The input of Pb is higher than the other two, probably due to leaded fuel combustion.

FINAL REMARKS

A mathematical approach using similarity theory has been used to predict the wind speed, drag coefficient and friction velocity to be introduced in the well-known deposition model of Slinn and Slinn (1980). In this way, it is possible to determine the dry flux of heavy metals when the sampling conditions (aircraft) do not reach the required reference height of 10 m. Dry deposition velocities were similar to those of Van Jaarsveld et al. (1986) and Krell and Roeckner (1988). Even though atmospheric input determinations in this work have a preliminary nature, since they are based on a reduced data base, results seem to be promising.

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DISCUSSION

G. SCHAYES — Up to which height did you apply surface layer formulation on your data?

C.M. ROJAS

Since this approach is based on turbulent features of air parcels, it can be successfully used provided certain boundary conditions are met. For example, problems may arise in trying to estimate the wind speed at a certain height when the wind speed at a higher altitude (which in our case did not exceed 50 m) is low (a few m/sec), and atmospheric conditions are stable. Then, the two heights will be separated by a layer of undisturbed air, which makes any calculation difficult.

J. ALCAMO

I noticed that the literature values for V_d that you quote in your paper (Table 1) are a factor of ~50 lower than your calculations (Table 2). What is the reason for this?

C.M. ROJAS

The values you mention have not been cited in the literature; they are the result of this model applied to the centroid of the size distribution reported in the literature. The main difference is that values shown in Table 2 correspond to a weighted average taking into account the whole size distribution (and a set of size distributions), which might be in our case shifted towards larger particles.

C.J. WALCEK

Do you see evidence that particles you collect are affected by evaporation or growth due to condensation or evaporation of water vapor during collection? If so, how might this artifact affect your calculated deposition fluxes?

C.M. ROJAS

We have not yet taken into account such effects. However, if particle growth due to condensation of water vapor really takes place during collection, I would expect higher concentrations on the first impactor stages, which could lead to a biased particle size distribution and therefore an overestimation of the dry flux.