1.16   DRY DEPOSITION OF FLY ASH DEPENDING ON BOUNDARY-LAYER STRATIFICATION AND UNDERLAYING SURFACE ROUGHNESS: A MODEL VALIDATION STUDY

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INTRODUCTION

The effects of the underlying surface to the dry deposition of airborne ingredients are known at least in qualitative sense: rougher surface initiates stronger wind-induced turbulence and therefore, as a rule, larger deposition velocities. Numerous measurements (McMahon & Denison, 1979) have shown that at other similar conditions the dry deposition fluxes are larger on the areas with high plant cover (forest) than on the areas with lower one (grassland). The detail model calculations (Ruijgrok et al., 1995) show that deposition fluxes of particles with diameter about 10 μm can be as much as ten times larger in the forest than in the open landscape.

Direct measurements of deposition velocities are technically complicated. Even more complicated is the theoretical interpretation of results, including several parameters (e.g. Monin-Obukhov length, surface roughness, friction velocity), which cannot be measured directly and are rather sensitive in respect to the values of measured or modelled meteorological parameters.

Measurements of medium-term (several days or weeks) average deposition fluxes of coarse solid particles have been carried out in north-eastern part of Estonia since 1985. These studies base on accumulation of ash components in typically stable wintertime snow cover in this region. Due to relatively flat nearly-natural landscape, sparse rural inhabitation and a few large point sources with specific source signature (oil-shale-fired thermal power plants) this area is proven to be appropriate for deposition studies (Kaasik & Sõukand, 2000). In 1996 there were found systematically higher deposition fluxes at forested areas than on open bog (Kaasik *et al.*, 2000), which is consistent with theoretical assumptions.

FIELD MEASUREMENTS

In order to clarify the effects of the thermal stratification and underlying surface roughness to dry deposition of particles, two time intervals with extremely different meteorological conditions were selected for field study. As the wet deposition is evidently not affected by surface roughness (as far as precipitation amount is not affected), a criterion for selection the sampling intervals was a small frequency of precipitation events during these periods.

Summer campaign (August 2-12, 2002): 4 deposition samples were taken from polluted area and 2 samples from background site (140 km away) using the slightly modified EMEP precipitation sampling method – the funnel collector, which was carefully washed at the end of the sampling to collect entirely all dryly deposited matter.

Winter campaign (December 2-14, 2002) - 12 deposition samples were taken from polluted area and 5 samples from background site. Samples were taken from natural snow layer, accumulation time was identified from meteorological data.

The chemical analysis (ion chromatography) of precipitation samples and also microscopic counting of deposited spheroidal fly ash particles (chemically inert spheres originating from high-temperature combustion, Alliksaar et al., 1998) with size over 5 μm were carried out.

METEOROLOGICAL CONDITIONS
Meteorological data were gained from the output of meteorological model HIRLAM (version 4.6.2) applied for regular weather forecast in Finnish Meteorological Institute (horizontal resolution 22 km, 31 levels, meteorological output fields with 6 hours time step). Usually 6 hour forecast data were used, longer forecasts filled a few gaps in database.

August 2 – 12, 2002
Meteorological data refer to dominantly convective conditions with short and shallow night-time inversions (Figure 1, A): the sun was 8 hours below the horizon, leaving not very much time to form a deep inversion. No rainfall was registered, negligible precipitation amounts formed from night-time dew.

December 2 – 14, 2002
Due to rather stable Siberian-type anticyclon, short day (less than 7 hours sunshine) and solar elevation not exceeding 10° a long-lasting thermal inversion formed (Figure 1, B). Weak day-time heating of underlying surface could not break the inversion above during several days. About a half of the thin snow layer (HIRLAM suggests 6 – 7 mm of precipitation during the entire time interval) accumulated during a snowfall event in the very beginning of sampling period. Another snowfall event with slightly unstable stratification occurred in December 12.
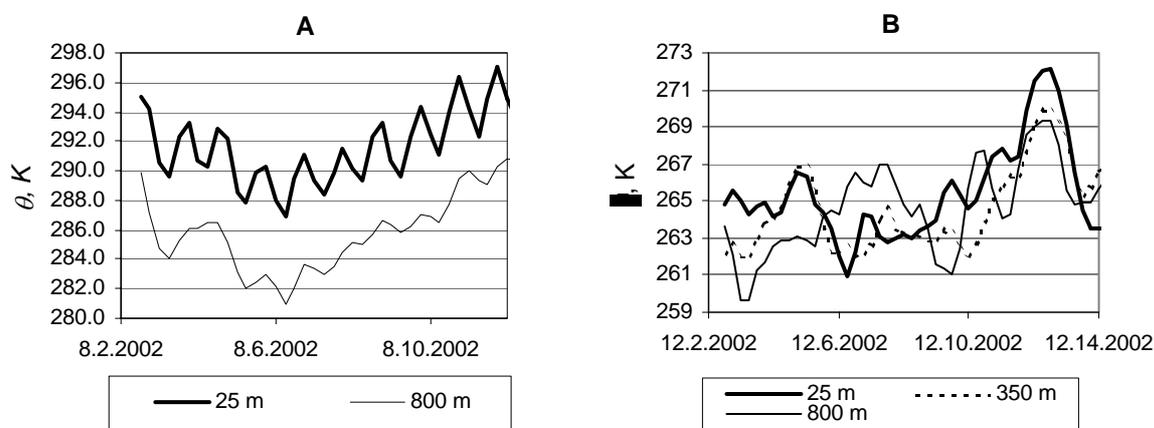
Figure 1. Potential air temperatures at different levels during August (A) and December (B) campaigns, 2002.

MODELLING
The SILAM model used is a Lagrangian particle (random-walk) model originally developed in Finnish Meteorological Institute for transport and deposition calculations of radionuclides (i.e. fine particles). For present study (coarse particles) the gravitational settling according to Stokes law was applied in lower model layer (0 – 50 m). Fractional aerosol spectrum was introduced. The size spectrum of oil shale fly ash (Aunela et al., 1995) was approximated into two fractions: 3 – 7 μm (26% of the total mass) and 7 – 11 μm (74% of mass). Deposition fluxes were calculated in HIRLAM 22 km grid, but concentrations in the air with finer resolution – in 5 km cells.

Emission data on Narva power plants (main sources of fly ash, 24 600 tonnes in 2002) and 6 minor sources were gained from the report of Estonian Ministry of Environment (Kohv et al., 2003).

RESULTS

Measured (averaged over the period) deposition fluxes of calcium and sulphate ion and spheroidal fly ash particles as well as modelled fly ash concentrations and deposition fluxes are presented in Tables 1 and 2. Modelled concentrations are averages over lower 200 m layer of the atmosphere.

Table 1. Measured deposition fluxes of fly ash components and modelled (SILAM) concentrations and deposition fluxes of fly ash, August 2 – 12, 2002. Approximate distance from nearest of two main power plants is indicated in brackets.

Sample No.	Site type	Ca^{2+}, mg/m^2 per day	SO_4^{2-}, mg/m^2 per day	Spheroidal particles, 10^3 no./m^2 per day	Modelled fly ash concentration, μg/m^3	Modelled fly ash flux, mg/m^2 per day
1	Bog forest (30 km)	10.9	4.7	3.9	4.15	0.320
2	Open bog (30 km)	14.7	5.5	2.6	4.15	0.320
3	Bog forest (30 km)	11.6	2.4	3.6	4.15	0.320
4	Open bog (30 km)	14.3	4.8	1.9	4.15	0.320
6	Bog forest (background)	13.7	-	0.9	0.20	0.026
5	Open bog (background)	14.8	-	0.3	0.20	0.025

Table 2. Measured deposition fluxes of fly ash components and modelled (SILAM) concentrations and deposition fluxes of fly ash (average and standard deviation), December 2 – 14, 2002. Number of samples and approximate distance from nearest of two main power plants is indicated in brackets.

Site type (number of samples)	Ca^{2+}, mg/m^2 per day		SO_4^{2-}, mg/m^2 per day		Spheroidal particles, 10^3 no./m^2 per day		Modelled fly ash concentration, μg/m^3		Modelled fly ash flux, mg/m^2 per day	
Woodland (6, 15-20 km)	6.37	±0.99	6.88	±1.22	49.2	±8.8	13.05	±0.65	1.29	±0.06
Open land (5, 15-20 km)	6.83	±0.80	6.60	±0.53	62.7	±6.7	12.68	±1.27	1.28	±0.06
Woodland, backgr. (2)	0.22	±0.03	0.42	±0.02	0.6	±0.2	0.51	±0.00	0.04	±0.00
Open land, backgr. (3)	0.42	±0.01	0.58	±0.21	0.9	±0.3	0.53	±0.01	0.05	±0.00

DISCUSSION

In August, 2002 the deposition fluxes of calcium ion (Table 1) were high compared to previous studies in the same area and far from the nearly 1:1 rate with sulphate flux found typical for oil-shale fly ash deposition (Kaasik & Sõukand, 2000). Thus, we have enough reason to think that most of deposited calcium originates from other sources than oil shale combustion. Hot dry weather during long-staying summertime high-pressure area over all the north-eastern Europe caused the occurrence of severe forest fires and intense soil erosion in Baltic countries and nearby Russia. Therefore the only reliable indicator of combustion sources is the flux of spheroidal particles. Unfortunately, we do not have absolute calibration for these spheres as the concentrations of these particles are measured only in electrostatic filter ashes of power plants and not in emitted fly ash. Deposition fluxes of spheroidal particles seem higher in forested areas, but opposite tendency appears for calcium flux. More summertime measurements are needed to clarify the situation.

In December (Table 2) the measured deposition fluxes of calcium were lower (in spite of closer distances to sources) and the fit with the sulphate was almost perfect. Thus, most of deposition probably originated from regional power plants. Keeping in mind standard deviations, there is no significant forest to open land difference between deposition fluxes neither of calcium and sulphate ions, nor of spheroidal particles. The modelled deposition fluxes are severely underestimated, regarding the 22% calcium content of Estonian oil-shale fly ash (Pets et al., 1985). Reasons are not fully understood yet but probably related to the different initial purpose of SILAM model. It is oriented to regional-to-continental scales of transport where the small particles play the major role. This study constitutes only the initial stage of tuning this model for applications to dispersion of coarse particles at local scales. As SILAM applies only aggregated roughness length over HIRLAM grid cell, there is impossible to include directly the effects of forest and open land patches. Small differences in Table 2 occur only due to locations fall into different grid cells.

More successful was the run of AEROPOL model (Kaasik, 2004), which has local to regional Gaussian dispersion scheme, 1 km grid resolution and has been many times validated against the deposition data of oil-shale combustion products (Sofiev et al., 2003). A different (coarser resolution) meteorological data set of NOAA Air Resources Laboratory (look at http://www.arl.noaa.gov/ready.html) was applied. The results of comparison are presented in Table 3.

Table 3. Measured and modelled (AEROPOL) deposition fluxes of fly ash (average and standard deviation), December 2 – 14, 2002. Number of samples and approximate distance from nearest of two main power plants is indicated in brackets.

Site type (number of samples)	Measured flux (estimation based on Ca^{2+} flux), mg/m^2 per day		Modelled flux, mg/m^2 per day (AEROPOL)	
Woodland (6, 15-20 km)	29.0	±4.5	22.0	±3.1
Open land (5, 15-20 km)	28.3	±3.6	19.8	±3.7

High horizontal resolution is not sufficient to reproduce the roughness-dependent deposition flux, whenever such exists. Applying the aerodynamic and quasi-laminar sublayer resistances (r_a, r_b) in sequence and inverse gravitational settling velocity $1/v_g$ in parallel to these, we get for total deposition velocity v_d:

$$v_d = 1/(r_a + r_b + r_a r_b v_s) + v_s \qquad (1)$$

The sensitivity in respect to surface roughness depends very much on how precise parameterisation schemes for resistances are applied. Preparing the deposition velocities for AEROPOL model, in addition to the molecular diffusivity through Schmidt number Sc, the inertial impaction was taken into account, including the Stokes number (St) similarity:

$$r_b = 1/\left[u_*(Sc^{-2/3} + 10^{-3/St}) \right] \qquad (2)$$

Without the inertial impaction term (Slinn, 1982) the quasi-laminar sublayer resistance becomes highly dominating over aerodynamic one, leaving only gravitational settling velocity evident, which does not depend on underlying surface properties.

ACKNOWLEDGEMENTS

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