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MARINE AEROSOL FLUXES DETERMINED BY SIMULTANEOUS MEASUREMENTS OF EDDY COVARIANCE AND GRADIENT METHOD

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Abstract

In this paper the results of the aerosol flux measurements in near water boundary layer are presented. Measurements were conducted using two micrometeorological methods in the southern Baltic Sea region. The preliminary results of comparisons of two methods are discussed. The eddy covariance (EC) method is shown as a method for the calibration of the gradient method (GM). The reason for proceeding with such calibration is to show that the GM, which is an indirect measuring method, is comparable with a direct method such as the EC. In the EC measurements the condensation particle counter (CPC, measuring range 0.05 - 3 μm with 1 Hz counting speed) and the Research Anemometer (50 Hz measurement speed) were used. For the GM, a Classical Scattering Aerosol Spectrometer was applied (measuring range from 0.5 to 47 μm diameter, within 36 measuring channels). Data from scientific cruises of the r/v Oceania in the southern Baltic Sea between 2008 and 2011 were analyzed.

Key words: air-sea interaction, marine aerosol flux, marine boundary layer, sea spray aerosol

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1. Introduction

Knowledge about sea spray aerosol (SSA) fluxes is important for many fields of geosciences, such as air-sea interaction processes, modeling cloud microphysical properties or aerosol radiative influences (Tsigaridis et al., 2013; Zielinski, 2004; Zielinski and Zielinski, 2002). In literature there are few experiments showing simultaneous measurements of SSA fluxes based on different methodologies. Petelski and Piskozub (2006) successfully compared the dry deposition, whitecap method and the gradient method (GM). Between these three methods only the GM (Petelski, 2003) is a micrometeorological method and is based on the Monin-Obukhov Theory (M-O theory) (Monin and Obukhov, 1953). In recent years the GM was successfully applied in different marine regions (Markuszewski et al., 2017; Petelski, et al., 2014; Savelyev et al., 2014).

The eddy covariance (EC) is the most popular method of measuring SSA fluxes (de Leeuw et al., 2007; Kaimal and Finnigan, 1994; Norris et al., 2012; Whitehead et al., 2012) and it was first applied to marine measurements by Nilsson et al. (2001).

In this paper we present comparison of the GM and the EC methods based on the simultaneous measurements. Using these results it is possible to conduct a calibration method which allows to depict the differences between the two approaches. It has been shown, that this method provides correct results.

2. Experimental region and instruments

The aerosol data used in this work were gathered during 3 scientific cruises on board of r/v Oceania. The cruises were conducted in the southern Baltic Sea region at three different stations: Southern Middle Bank: from 17.09.2008 to 24.09.2008 and 11.04.2010, Slupsk Furrow: 5.04.2011 and near the

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Hel Peninsula - 7.04.2011. All places and measurements are presented in Fig. 1.

The Scattering Aerosol Spectrometer (CSASP) was placed on a special lift on board of the vessel. Methodology of gathering data for the GM on board r/v Oceania has been described by Petelski (2003). The CSASP device has been set for counting aerosol particles for 10 seconds at each of the measurement ranges (there are 4 ranges within 15 channels - range #0: 2 µm -47 µm with 3.0 µm step, range #1: 2 µm - 32 µm with 2.0 µm step, range #2: 1 µm -16 µm with 1.0 µm, #3: 0.5 µm -8 µm with 0.50 µm step).

The initial measurements with the EC method on board r/v Oceania were conducted using condensation particle counter (CPC), TSI model 3771 and the GILL R3-50™ 3D Research Anemometer. The GILL was situated on the bow of the vessel at a 10 m elevation. The CPC was situated at the same elevation but placed on the research balcony 4 meters behind the GILL. The delay between the instruments was taken into account. The CPC made by TSI, model 3771 allows to measure with 1 Hz speed, which is enough to provide satisfactory data for covariance determination (de Leeuw et al., 2007).

All measurements are based on ambient aerosol, no dryer was used. To eliminate the influence of humidity, concentration data were corrected to 80% relative humidity (Fitzgerald, 1975; Petelski, 2005). Moreover, measurements conducted when humidity was higher than 95% were excluded.

3. Processing data using the MG and the ED

In processing data we used a total of 55 hours of measurements including 35 measurement hours of the vertical aerosol concentration profiles and 20 hours of simultaneous GM and EC measurements.

The gradient method is based on aerosol fluxes derivation from vertical aerosol concentration gradients (Petelski, 2003).

Therefore, the measurements were made at five elevations: 8, 11, 14, 17 and 20 m above sea level (a. s. l.), with a single measurement at each level lasting 2 minutes. The vertical aerosol concentration gradient was obtained from a minimum of 4 measurement series. Thus each result consists of a one-hour series with the average sampling time at each elevation of 8 minutes. Based on such aerosol data the determination of aerosol gradients from aerosol concentration profiles is made. Using aerosol gradients it is possible to determine aerosol gradient flux F_g (Petelski, 2003) (Eq. 1):

$$F_g = N_* u_* \tag{1}$$

where N_* is a scale of aerosol concentration, u_* is a friction velocity.

In the EC method the aerosol flux F_e is determined by the covariance calculation between turbulent fluctuating aerosol concentration c' and the turbulent vertical wind speed w' (de Leeuw et al., 2007) (Eq. 2):

$$F_e = cov(w,c) = \langle w' \cdot c' \rangle \tag{2}$$

In the EC flux determination half-hour data series were used. The delay related with distance between the CPC and the GILL was corrected and in the further data processing the rotation and the tilt angle were applied. Both methods are applied only for steady conditions in the near sea boundary layer. Non-representative data connected i.e. with ship-tracks or dust advection were rejected. The main uncertainty of the EC measurements is connected with movements of the vessel on the sea surface during high wind conditions (rolling, pitching and yawing). The GM fluxes are free from such errors due to one-hour averaging. The calculated fluxes are presented in Fig. 2. Results for the EC fluxes are scattered due to the ship's movement on the wavy surface of the sea.

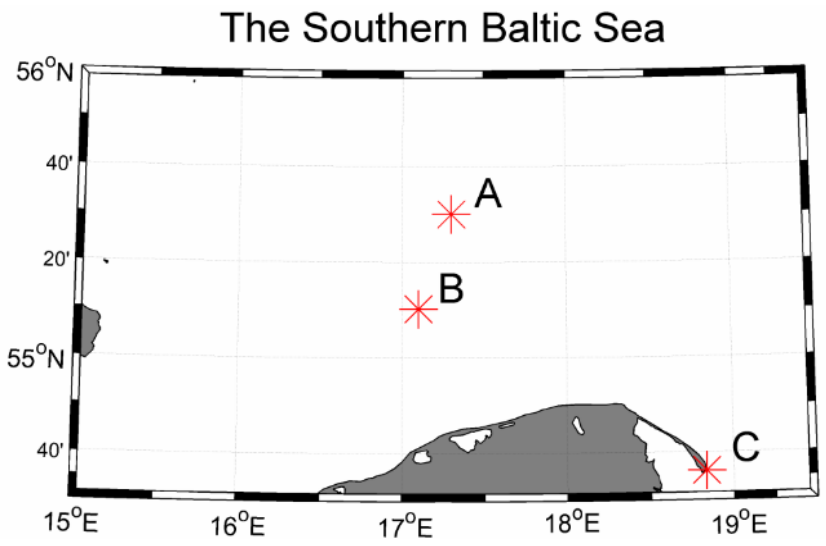

Fig. 1. Locations of the measurement stations (3 points in the Southern Baltic Sea). A: the Southern Middle Bank region, B: Slupsk Furrow, C: the Hel Peninsula region

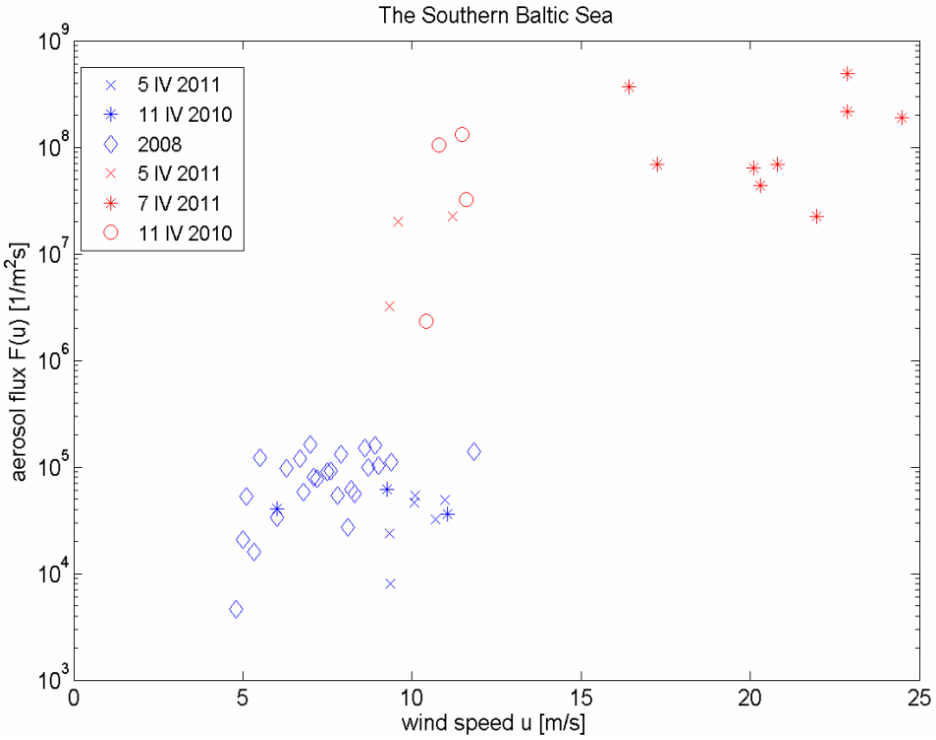

Fig. 2. Results of EC and GM flux measurements on board r/v Oceania. Eddy correlation fluxes are represented by red points. Gradient fluxes are represented by blue points

The highest measurements for the MG is for wind speed u=12m/s. Conducting the GM measurements during higher wind speeds was impossible for safety reasons - two people must be present on the measuring balcony during the measurements.

4. The EC method for calibrating the GM

The main purpose of this paper is to propose a calibration method for the GM. The main goal of such calibration is showing a relationship between the most popular direct EC method and passive and a better GM for on board measurements.

The idea of the calibration method is to find a calibration coefficient x which allows for result transformation from the gradient fluxes to the EC fluxes. Because the measurement ranges of the CPC and the CSASP-100-HV are different the x coefficient is defined as a ratio between a definite integral in the range of the CPC measurements range to the CSASP-100-HV (0.5 µm to 8 µm) range measurements from a theoretical function fitted f_{fit} to a gradient function (Eq. 3):

$$x = \left[\int_{0.05}^{3} f_{fit}(d')dd'\right] / \left[\int_{0.5}^{8} f_{fit}(d')dd'\right] \qquad (3)$$

The first step is to fit the theoretical function. For this purpose the analyses of partial fluxes measured by the CSASP-100-HV must be made. The advantage of this device is the ability to conduct measurements in 36 channels. In this work only 15 channels for diameters from 0.5 µm to 8 µm were used. The remaining concentration of bigger aerosol particles (8 µm -34 µm) was only around 0.01% of total aerosol concentration. In the fitting procedure an exponential f_{exp} and power f_{pow} function were chosen. Results of the fitting are presented in the Fig. 3. Fitting for both functions is characterized by a good determination coefficient (for f_{exp} r=0.99 and f_{pow} r=0.96). Total fluxes ratio, which have been derived using the function fitting method, for size ranges for both methods, is the measure of the difference between the EC and the GM methods. The total flux value is represented by a definite integral for a given size range (Eq. 3). Based on this equation the x coefficient was determined for both functions. The results are presented in Table 1.

Table 1. Comparison of theoretical total flux values and x coefficient (Eq. 3) for the fitted functions

function	*integration range [µm]*	*result [1/m²s]*	*x*
$\int f_{pow}(d')dd'$	(0.5, 8)	$5.1*10^4$	x_{pow}=713
	(0.05, 3)	$3.7*10^7$	
$\int f_{exp}(d')dd'$	(0.5, 8)	$4.3*10^4$	x_{exp}=6
	(0.05, 3)	$2.4*10^5$	

5. Results and discussion

The main aim for this procedure is to use gradient fluxes to obtain the EC flux scale. To check the quality of the results the $f_1(u^2)$ function is fitted to

the GM fluxes using the diagram of flux versus wind speed. The reason to use the $f_1(u^2)$ function is only for obtaining better quality results. However, it is also supported in the literature: Mulcahy et al. (2008), Petelski and Piskozub (2006), Petelski et al. (2005). Such function is multiplied by the calibration coefficient x. The end result should give a curve which will be in the range of the EC fluxes. This procedure is presented in Fig. 4. As we can see in Fig. 4 the transformation of $f_1(u^2)$ using factor x_{pow}=713 is much better than using the factor x_{exp}=6. We presented first approach to our calibration method. In Fig. 4 the differences between experimental EC values and the fitted function (red line) differ by one order of magnitude.

This problem comes inter alia from imperfection of the fitting for coarse aerosol mode only. If the measurements for wider aerosol size spectrum were available, it could be possible to use the log-normal fitting.

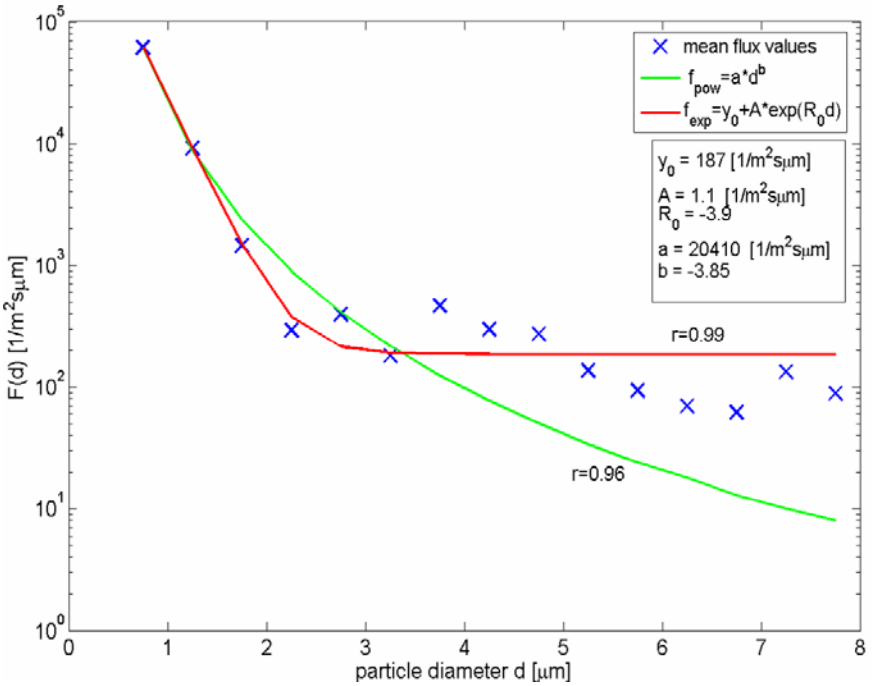

Fig. 3. Results of the functions fitting to partial aerosol fluxes

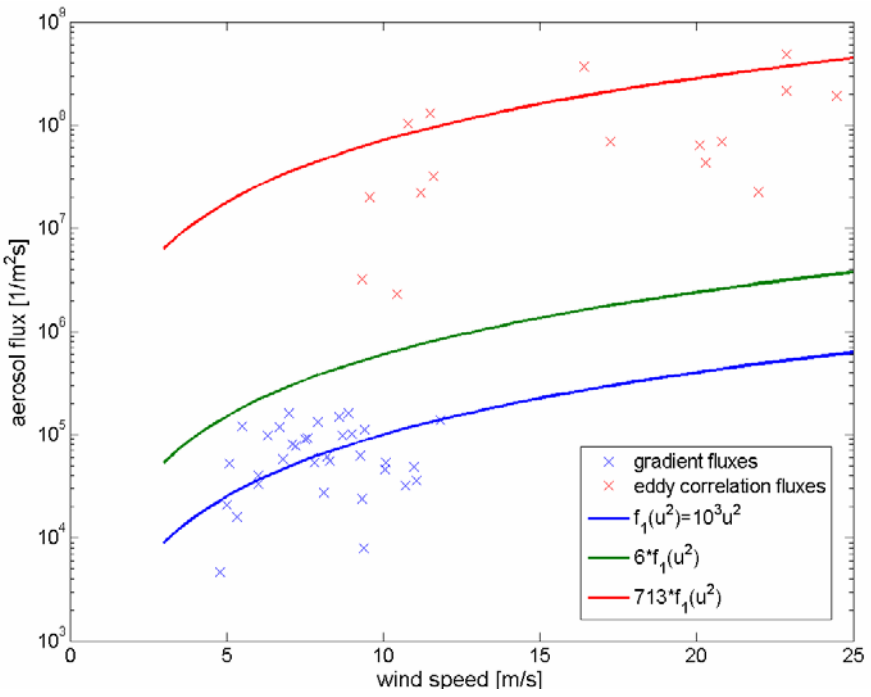

Fig. 4. The experimental aerosol fluxes versus wind speed with square function fitted and functions generated using the calibration coefficient x

Another problem involves a small spectrum of wind speeds available for gradient measurements. Conducting aerosol measurements in higher wind speed conditions (higher than 7 in the Beaufort Scale) were impossible for safety reasons. Nevertheless, based on existing data it was possible to show that this methodology provides satisfactory results.

6. Conclusions

In this paper we presented, that applying of the calibration method is justified. The results obtained from the gradient measurements with narrow range of sizing can be transformed to more general eddy correlation results. Power function describes better the aerosol size composition in the range 0.5 µm – 8 µm in the first approach of the calibration than the exponential one. Such calibration method could be used in other areas of research, everywhere when simultaneous methodologies with different devices are applied.

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