Ångström coefficient as a tracer of the continental aerosols

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ABSTRACT

The variation of the extinction coefficient with wavelength can be presented as a power law function with a constant (related to the power factor) known as the Ångström coefficient. When the particle size distribution is dominated by small particles, usually associated with pollution, the Ångström coefficients are high; in clear conditions they are usually low. Long residence time of air masses over land and in particular the passage over large urban areas cause high concentrations of fine particles and thus high values of the Ångström coefficients. The opposite effect can be observed over water. The longer the time that the air masses spent over water the more evident is a change in the aerosol size distribution caused by the deposition of continental aerosols. As a result of this process the measured Ångström coefficient values become much smaller. Therefore this parameter is a good tracer for the concentration of aerosols originated over land. The relation between the Ångström coefficient and TOS (time over sea) is demonstrated on three data sets. The first data set includes measurements collected at the Irish Atlantic coast in 1994 and 1995, the second one, data collected within the Rough Evaporation Duct (RED) experiment that took place off Oahu, Hawaii in 2001. The third one represents data collected at the Baltic Sea during cruises in 1997and 1998.

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Keywords: Ångström coefficient, continental and industrial aerosols, deposition process, time over sea

1. INTRODUCTION

The first treatise in English showing that the spectral dependence of extinction by particles may be approximated by a power law function was written by Anders Ångström[1] in 1929. Although the constant in the power factor is now known as the Ångström coefficient (alternative names existing in the literature are Ångström exponent and Ångström parameter), the author himself refers to the earlier laboratory studies by Lindholm and his observations of power law relationship for the absorption of thin powders at Uppsala in 1912. Lindholm has never described his results in English and this is probably the reason that the full credit is given to Ångström[2].

In 1955 Junge was the first to demonstrate the relationship between the Ångström coefficient and polydisperse aerosol size distributions[3]. The power law relationship describing the number density of aerosols as a function of radius has been widely used and known as the "Junge distribution"[4,5]. However, more recent studies[5,6] show that atmospheric aerosols are rarely monomodal and that their distributions are more appropriately described by multimodal lognormal distributions. The latter distribution is also assumed to represent aerosol distributions measured within the network of sun photometers called AERONET[7] (http://aeronet.gsfc.nasa.gov). The aerosol volume distributions, retrieved from spectral optical depth measurements combined with the angular distribution of sky radiance measured at different wavelengths, are modelled as a bimodal lognormal function[8,9,10]. The two modes are called "fine mode" and "coarse mode", respectively. The fine and coarse modes are divided by a radius r=0.6 μm[11]. When the particle size distribution

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is dominated by the fine mode (small particles), the Ångström coefficients are high. In the opposite situation, when the coarse mode dominates, the Ångström coefficients are usually low.

Both the spectral aerosol extinction (aerosol optical thickness) measurements and the Ångström coefficient values reflect the history of air masses. Long residence times over land and in particular the passage over large urban areas cause high concentrations of fine particles and high values of the Ångström coefficients. The opposite effect can be observed over water. The longer the time that the air masses spent over water the more evident is a change in the aerosol size distribution caused by the deposition of continental aerosols12. On the other hand, the larger is the wind speed the more significant is also the sea-salt aerosol production. Therefore these two main processes, i.e. production and deposition, determine the mutual relation between fine and coarse modes contributions.

In view of the dependence of the aerosol optical properties on the size distribution and the chemical composition, the Ångström coefficient α can be regarded as a tracer of the continental and industrial aerosols. Reid et al.13 showed that the Ångström coefficient for smoke particles in Brazil is well correlated with their size, single-scattering albedo and the backscatter ratio. Kusmierczyk-Michulec et al.14,15 demonstrated that the Ångström coefficients values can be used to estimate the mass concentration of sea salt aerosol (SSA), black carbon (BC) and organic carbon (OC), expressed in terms of their ratio to the total particular matter (TPM). These empirical relations were derived from simultaneous optical and chemical measurements on the Baltic Sea in July 1997 and March 1998 and successfully tested for a different area15. The idea of simultaneous optical and chemical measurements was further tested at the Dutch North Sea coast. The sun photometer measurements at the AERONET station in The Hague (The Netherlands) for the period January 2002 to July 2003 were coupled to chemical aerosol data, i.e. PM10 (particulate matter with an aerodynamic diameter of less than 10 μm) and BC, from a nearby station of the Dutch National Air Quality Network16. The relation between the Ångström coefficients and BC/PM10 for the clean days (clean air according to the EU air quality norm requires that PM10<50 μg/m3) is similar to that derived from observations over the Baltic Sea with coefficients that are within the experimental error.

This paper focuses on the (coastal) marine environment. The purpose of this paper is to demonstrate the relation between the Ångström coefficients and two main processes: deposition of continental particles and production of marine aerosols. The best meteorological parameter representing the deposition of small particles of continental origin is time over sea (TOS). The second process is related to the third power of the wind speed values[17]. The paper discusses the mutual relations between both processes, their influence on the aerosol size distribution and relation to the Ångström coefficient values.

2. THE AEROSOL DATA SETS

2.1. Baltic Sea Experiment

Aerosol optical thickness and aerosol chemical composition were measured during two cruises at the Baltic Sea, in July 1997 and March 1998. The measurements and data analysis procedures were extensively described in Kusmierczyk-Michulec et al.[14]. Relevant for the present paper are the Ångström coefficient values obtained from the spectral aerosol optical thickness measurements at eight channels (412, 443, 490, 510, 555, 670, 765 and 865 nm, bandwidth 10 nm).

The meteorological conditions during both experiments were completely different. Summer wind speeds were usually quite low with a maximum of 12 m/s, whereas in winter high wind speed conditions dominated with a maximum of 17 m/s. The time spent by the air masses over the sea (TOS) was estimated on the basis of 96-hourly backward trajectories,

by the application of the method of Petterssen[18], using wind fields from the high-resolution limited-area model (HIRLAM) of the Finnish Meteorological Institute[19] as input.

2.2. RED and Inisheer Experiments

The experimental procedure to record aerosol data was similar during both experiments. Aerosol size distributions were measured simultaneously by two classical scattering probes (Particle Measuring Systems, CSASP-200 and CSASP-100HV), providing a combined range of diameters between 0.21 and 42.5 μm. The internal data integration time was 1 second, and raw data was stored on disk after an accumulation time of 1 minute. Next, the raw data was averaged over 10 minute intervals. Prior to the experiments, the probes had been calibrated with particles of known sizes. The measured size distributions were used to calculate the extinction and the Ångström coefficient values.

The Rough Evaporation Duct (RED) experiment took place off Oahu, Hawaii from 26 August to 15 September 2001. During the experiment the aerosol counters and meteorological sensor were installed on the FLIP, an ONR-sponsored platform maintained by Scripps Institute of Oceanography (SIO). The aerosol probes at FLIP were mounted at a height of 20 m and during the measurements the probes were facing into the wind. The RED data set is representative for the maritime aerosol type.

The Inisheer experiment took place on the Irish Island –Inisheer, during 2 campaigns. The first one took place from 31 August to 17 September 1994, and the second one from 19 August to 20 September 1995. During the experiment the aerosol counters and meteorological sensors were placed on the top of a light house, at a height of 30m. PMS probes were pointed into the open sea direction. The Ireland data set represents both maritime and continental aerosol types.

The wind speeds during both experiments were similar. During the RED experiment the mean wind speed was about 6 m/s and the maximum value did not exceed 12 m/s. During the Inisheer experiment the mean wind speed was about 5m/s and the maximum value reached 14 m/s. For both data sets fetch lengths (distance from the coast) and wind speed values were used to estimate the time air masses spent over the sea (TOS).

3. ÅNGSTRÖM COEFFICIENT

The relation between the aerosol size distribution and the extinction coefficient c(λ) [1/km] can be described by the relation

$$c(\lambda) = \pi \int_{r_1}^{r_2} r^2 Q_{ex} n(r) dr \tag{1}$$

where λ is the wavelength , r is radius and Q_{ex} is the extinction efficiency factor, being a function of the complex index of refraction[20]. The extinction integrated over the whole column of atmosphere is a non-dimensionless parameter called the aerosol optical thickness:

$$\tau_a(\lambda) = \int_{H_{\min}}^{H_{\max}} c(\lambda, h) dh \approx \int_{H_{\min}}^{H_{\max}} c(\lambda) f(h) dh \tag{2}$$

where f(h) represents the vertical distribution of aerosols, h is the altitude in km, H_{min} and H_{max} are the lower and the upper altitude, respectively, at which a given aerosol type can be found.

The atmosphere can be modelled as superposition of three layers: boundary layer (BL), free troposphere and stratosphere. Mc Clatchey et al.[21] and Hess et al.[10] assume that whithin each layer the aerosol composition is constant with height. As a rough estimate, the BL is often assumed to have a depth of 2 km.

As mentioned in the introduction, the variation of the extinction coefficient or the aerosol optical thickness with wavelength can be presented in the form of a power law function1:

$$c(\lambda) = \gamma_c \lambda^{-\alpha} \tag{3}$$

or

$$\tau_a(\lambda) = \gamma_\tau \lambda^{-\alpha} \tag{4}$$

where γ_c and γ_τ are constant and α is the Ångström coefficient (also known as Ångström exponent or Ångström parameter). Usually, this parameter is determined in the spectral range from 440 nm to 870 nm.

4. RESULTS

4.1. Ångström coefficient vs aerosol size distribution

Deposition of continental particles can be demonstrated in a simple way by analyzing the aerosol size distribution measurements. Two different experimental data sets collected during Inisheer (Fig 1A) and RED (Fig.1B) experiments were used for that purpose. Figure 1A presents the measured volume aerosol size distributions representing the following Ångström coefficient values:-0.14, 0.13, 0.53, 0.70, 1.08 and 1.45. It is easily seen that the smaller is the Ångström coefficient the less significant is the fine mode representing small particles. Since the coarse mode for all distributions is unchanged, this observation suggests that deposition process was the main process influencing the aerosol optical properties. In view of the coastal nature of the dataset, and in view of the relatively smaller sizes associated with particles of continental origin as compared to sea-spray particles, we interpret this as deposition of continental particles.

While the measurements taken at the Irish island were under a significant continental influence, the measurements obtained near Oahu represent more maritime conditions. Hence, the ratio of the smaller continental and the larger maritime aerosols is smaller and therefore the Ångström coefficient values are much lower. The maximum value of the Ångström coefficient found for this data set was 0.6, nearly three times lower than for the Inisheer data set. These single higher values were observed during a day when a storm event took place. During that day, as the air mass backward trajectories indicated, the air mass originated over the Pacific Ocean and have passed over part of the Hawaiian archipelago. Knowing all these details the presence of small continental particles in this “open ocean” data set is understood. Figure 1B presents four volume aerosol size distributions showing that the fine mode decreases as the Ångström coefficient decreases, suggesting deposition of continental particles.

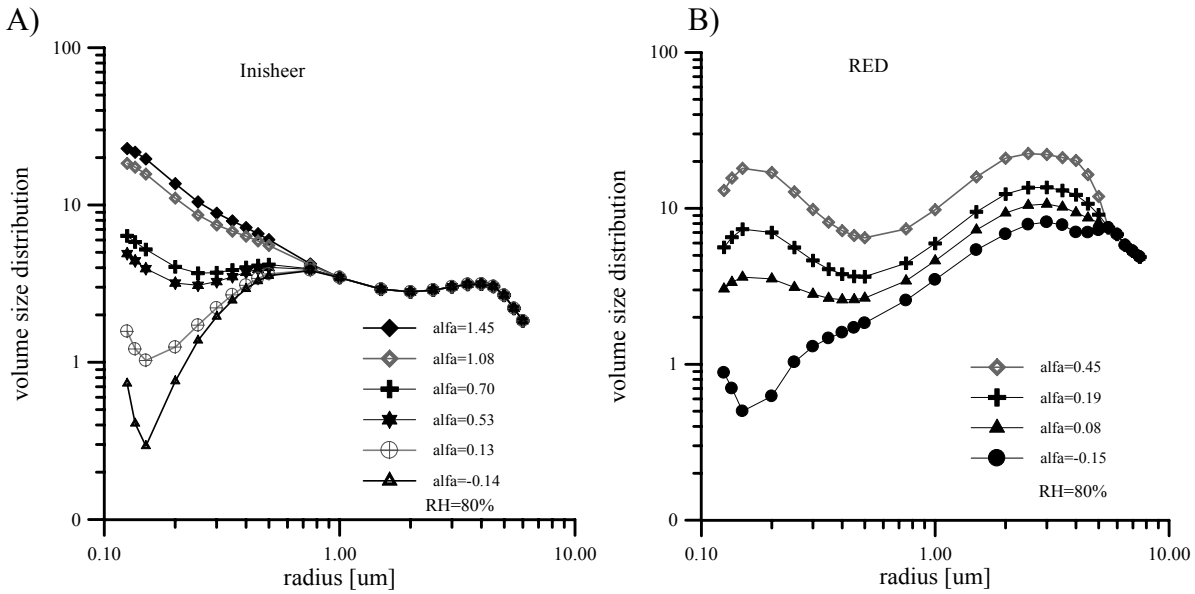

Fig.1. Examples of the aerosol volume size distributions corresponding to different values of the Ångström coefficient values (in plot indicated by "alfa") for two data sets: A) Inisheer and B) RED.

The relation between the Ångström coefficient and the concentration of smaller particles shown in figure 1 can also be presented differently. Figure 2 shows, for particles of 0.25 μm in diameter, a scatterplot of Angstrom coefficient and aerosol concentration for all records in the Inisheer and RED datasets. While there is some scatter, it is evident that the concentration increases with increasing Angstrom coefficient.

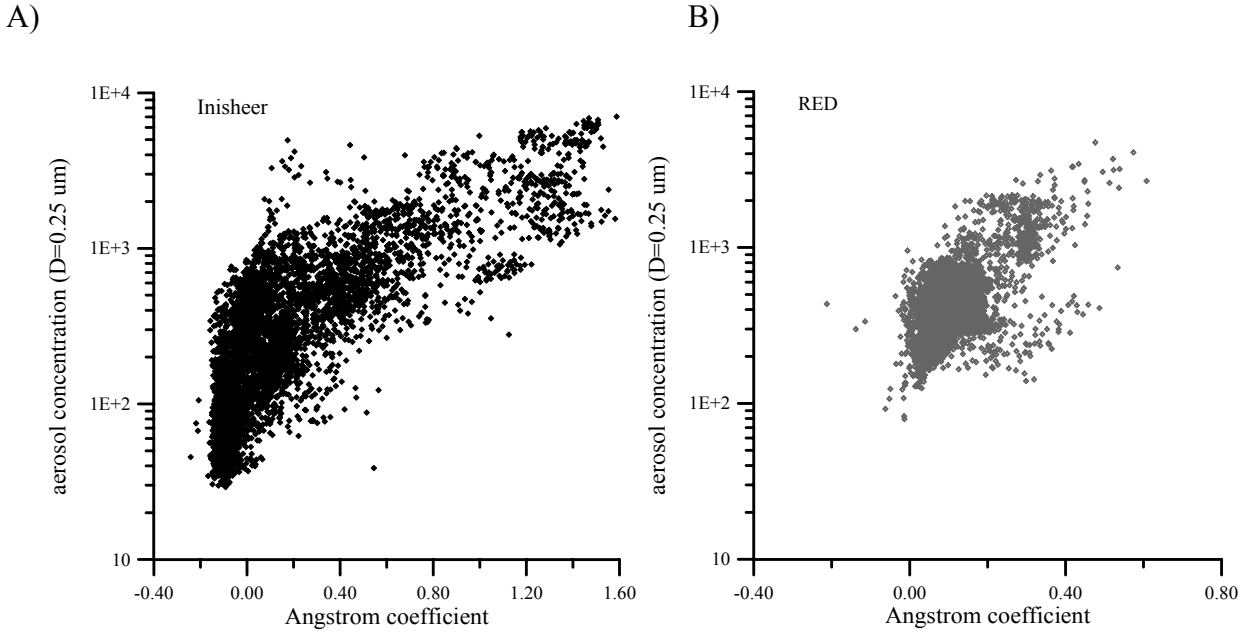

Fig.2. A-B Ångström coefficient vs aerosol concentration at 0.25 μm

4.2. Ångström coefficients vs TOS

When aerosol measurements are made at open sea it can be expected that they will represent the pure maritime aerosol type. However, in coastal areas the situation becomes more complicated. Usually the measured aerosols are a mixture which composition depends on the aerosol sources and the air mass history. The air masses residing longer over sea reflect the characteristics of maritime aerosols better than air masses that have been over sea for a shorter time. Kusmierczyk-Michulec et al.[12] demonstrated, by analyzing the aerosol optical thickness measurements in terms of the travel time of air masses over sea surface, that they could distinguish between continental, maritime and mixed aerosol type.

The authors showed that for the low wind speed conditions when deposition of continental particles dominates over production of maritime particles, a minimum TOS of 33 hours was required for an air mass to become predominantly maritime. The more complicated situation occurs when the wind speed is higher then 6 m/s, because in addition to the slow processes of deposition of continental aerosols also marine aerosol production takes place. In such a case the minimum TOS value to observe maritime aerosol characteristics (as indicated by low values of the Ångström coefficients) is much lower. This is nicely illustrated in Figure 3. Data collected during the Baltic experiment in July 1997 (European summer) reflect changes of the Ångström coefficient values caused by deposition of continental particles (relatively low wind speeds). Thus, we observe a slow decrease of the Angstrom coefficient with time over sea. Contrary to this, the data collected in March 1998 (winter and high wind conditions) represent the situation when the decrease in the Ångström coefficient is the result of a superposition of two processes: deposition of fine particles and production of coarse particles. In this case, the transition from a continental aerosol type to a maritime one takes place much faster (within a few hours), and Ångström values representative for maritime conditions are already observed in the first TOS-bin.

There is another reason that we observe this difference between summer and winter data and it is related to the BL (boundary layer) height. If we write a simplified equation describing changes in the aerosol concentration caused by only dry deposition[5]:

$$C_a = C_{a0} \exp(-\frac{V_d * X}{BL * U}) \tag{5}$$

where C_{a0} is the aerosol concentration [mol/m^3] at the beginning of the air mass trajectory and C_a -at the end (i.e., after traveling a distance X over water), V_d is the (size-dependent) deposition velocity in [m/s], BL is the height of the boundary layer [m] and U is the wind speed in [m/s]. Introducing the parameter TOS=X/U in [h], we can rewrite the above equation:

$$C_a = C_{a0} \exp(-\frac{V_d * TOS * 3600}{BL}) \tag{6}$$

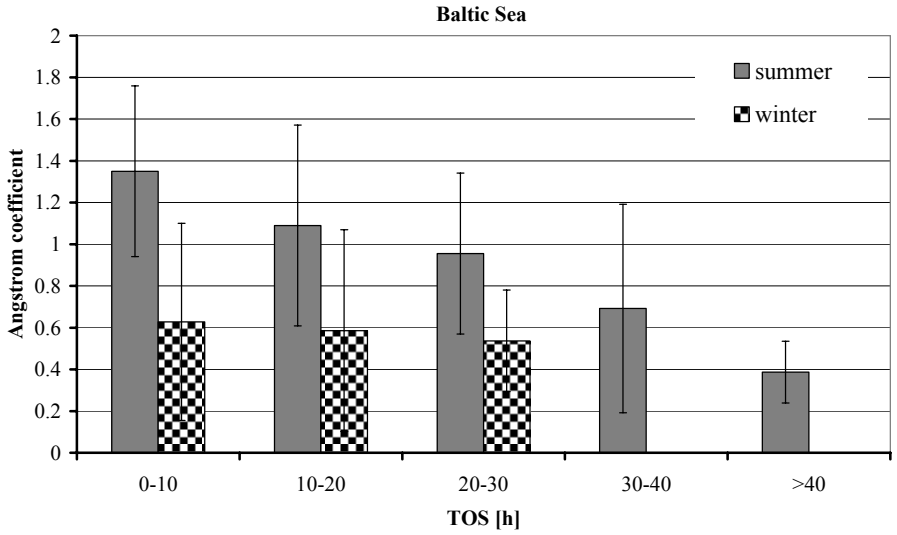

Fig.3. Decrease in the Ångström coefficient values with TOS. The vertical bars represent the standard deviation from the mean value of the Ångström coefficient.

During both experiments on the Baltic Sea radiosondes were launched on the research vessel Alexander van Humboldt four times per day. Based on these measurements the height of BL was estimated to be 1 km in summer and 0.5 km in winter. Using these values and equation 6, we can estimate what TOS is needed to observe significant changes in the aerosol concentration. The evaluation of equation 6 also requires the dry deposition velocity, which is subject to some debate. Therefore, two wind deposition velocities representative for the fine mode were considered: 0.001 m/s and 0.006 m/s. The first value was given by Slinn and Slinn[22] and the second one by De Leeuw et al.[23]. Figure 4 demonstrates the effect of different BL depths and dry deposition velocities on the decrease of the aerosol concentrations. The smaller BL depth in winter results in a more pronounced decrease, in agreement with the results shown in Figure 3.

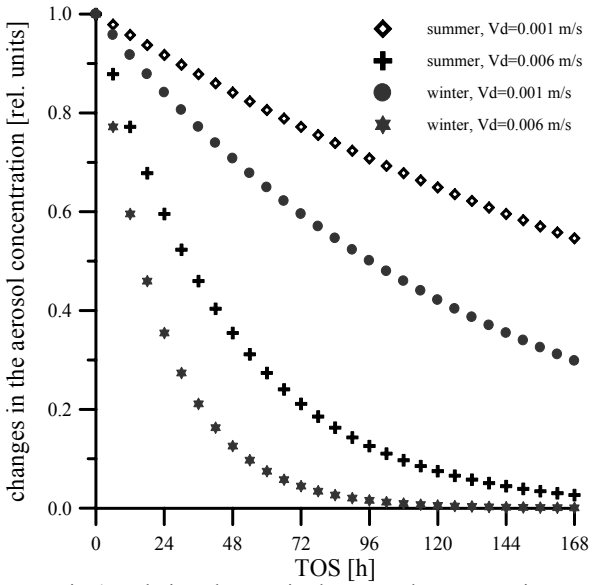

Fig.4. Relative changes in the aerosol concentration caused only by a deposition process that take place during 7 days of travelling over the sea surface. The simulation is done for two BL values representing summer (1km) and winter (0.5 km), and two deposition values for small particles: V_d=0.001 and 0.006 m/s.

4.3. Ångström coefficients vs U24

During the RED experiment most of the aerosol measurements correspond to almost unlimited fetch and TOS. Hence, there is very little continental contribution and therefore the deposition process of smaller particles was less visible. Figure 5 shows changes in the Ångström coefficient as function of the wind speed averaged over 24 hours (U24). Since the aerosol optical properties reflect the history of the air masses rather than instantaneous changes U24 seems to be a better parameter than the instantaneous wind speed U.

The left panel of Figure 5 shows that the Ångström coefficient decreases with increasing U24. Since deposition of continental particles played a minor role during the RED experiment, we must attribute this decrease to an increase of larger particles, or a more efficient production of marine aerosols. To better illustrate this effect the Ångström coefficient values were plotted against $U24^3$ (Figure 5B) due to the fact that production of marine aerosols is related to the third power of the wind speed values[17]. It is easily seen that starting at wind speed of 7-8m/s the Ångström coefficient values become stabilized.

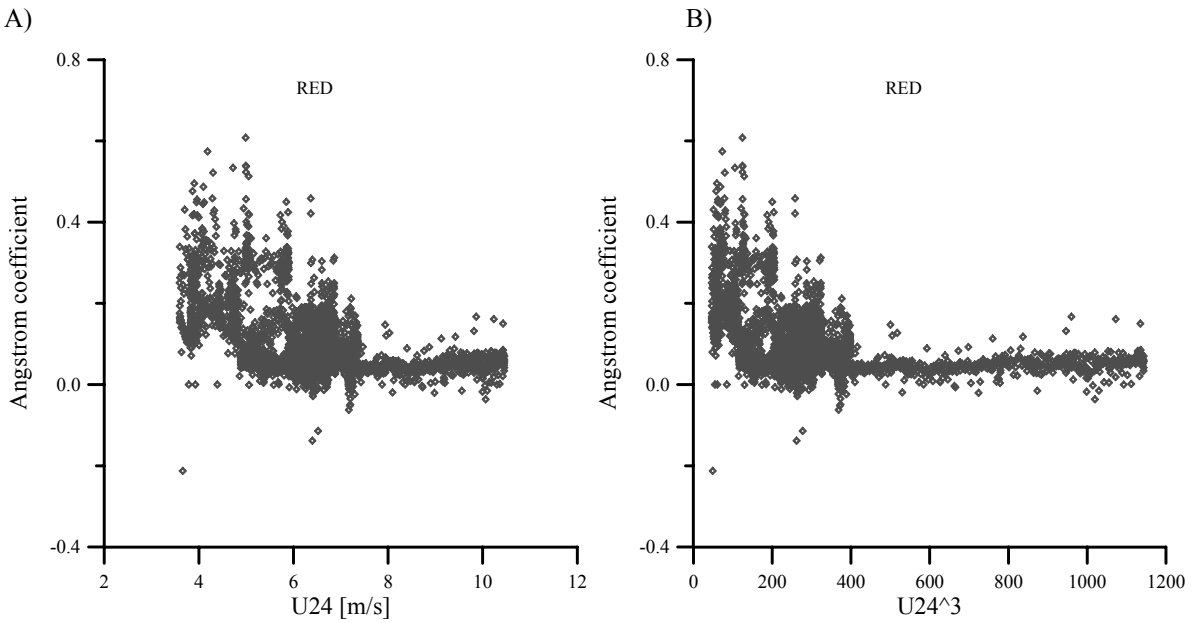

Fig. 5. The Ångström coefficient values vs wind speed averaged over 24 hours (U24) (plot on the left, A) and $U24^3$ representing the sea-salt aerosol production.

5. CONCLUSIONS

The Ångström coefficient values include information about the proportion between the fine and coarse modes. The fine mode usually can be attributed to the continental/industrial aerosols while the coarse mode represents sea-salts (unless there is an evidence that dust particles can be present). The relations between the Ångström coefficient values and the meteorological parameters like TOS and U24 will be further analysed for other data sets.

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