

# $\delta^{13}\text{C}$ VALUES IN SIZE-SEGREGATED ATMOSPHERIC CARBONACEOUS AEROSOLS AT A RURAL SITE IN LITHUANIA

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The investigations of the carbon isotopic ratio in different size aerosol particles at the Rūgštelė Integrated Monitoring Station (IMS) at the rural site (Lithuania) during 10–24 July 2008 are presented. Total carbon (TC) concentrations ranged from 0.06 to 0.35  $\mu\text{g m}^{-3}$  and those of elemental carbon (EA) from 0.02 to 0.15  $\mu\text{g m}^{-3}$  in accumulation mode, as well as from 0.06 to 0.14  $\mu\text{g m}^{-3}$  and from 0.02 to 0.06  $\mu\text{g m}^{-3}$  in coarse mode of aerosol particles, respectively. Organic matter (OM) concentration ranged from 1.0 to 25.4  $\mu\text{g m}^{-3}$ , with the average value of 5.6  $\mu\text{g m}^{-3}$ . Carbon isotopic ratios relative to standard for TC differ significantly in accumulation and coarse modes ( $-28$  and  $-24\%$ , respectively) and indicate a different origin of size-segregated aerosol particles. Almost constant organic carbon isotopic ratio ( $-31.8\%$ ) indicates that OM originates from local vegetation. Organic matter concentrations and the carbon isotopic ratio measurements in the size-segregated aerosol particles allow identifying sources of aerosol particles.

**Keywords:** carbon isotopes, carbonaceous aerosols, size distribution of aerosol particles, composition,  $\delta^{13}\text{C}$ , source apportionment

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## 1. Introduction

Carbonaceous aerosols play an important role in radiative forcing and climate change of the Earth. They are liable for visibility impairment and have impact on human health [1, 2].

Particles, composed mainly of elemental (EC) and organic carbon (OC), are a major constituent of anthropogenic and natural aerosols. However, the relative contributions of different emission sources of carbonaceous aerosols, their formation mechanism, and their atmospheric transformation and transport are not well understood.

It is known yet that EC and OC can provide useful information about different aerosol particle sources, their formation, and transport. The organic fraction in aerosols consists of primary compounds of both biogenic and anthropogenic origin, mainly deriving from vegetation waxes, microbial activities, and petrogenic sources, but the overall understanding of the organic aerosol remains relatively unclear [3]. The carbon isotopic composition of EC fraction is expected to be unchangeable because of its inert nature, reflecting the original signature created under high temperature combustions. Compounds of different origin have a unique

carbon isotopic value. Stable carbon isotopes can give clearer insight into the origin and formation processes of primary and secondary carbonaceous particles.

In an environmental research various isotopes are used: for example, carbon and lead isotopes were used for aerosol particle source identification in Paris [4]; plutonium and americium isotopes were used for radioactive contamination source determination in lake sediments [5]. Isotopic analysis has been used previously attempting to identify the sources of organic carbon in the atmosphere [6, 7]. Kelly et al. [8] observed no clear difference of the carbon isotopic ratio between fine ( $<1 \mu\text{m}$  diameter) and coarse ( $>1 \mu\text{m}$  diameter) aerosol. The authors were not able to evaluate contribution of C3 plants, industrial combustion, and biomass burning to the measured  $^{13}\text{C}/^{12}\text{C}$  ratio. Martinelli et al. [9] used the carbon isotopic ratio for assessing organic carbon (from sugar cane, C4 plant) input to the aerosol.

Most of the investigations on the characterization of organic aerosols deal almost exclusively with urban samples. It is generally assumed that EC is only emitted by primary combustion sources. Particulate OC/EC ratios exceeding 2.0 can be used as indicators of secondary aerosol formation [10].

Chemical analysis of aerosol particles has been used to study transport and sources of size-segregated aerosols [11]. Chow et al. [12] used MOUDI concentrations to examine the factors responsible for the evolution of the aerosol size distribution in a rural Californian (US) environment.

The aim of this work is to estimate quantitatively the organic and elemental carbon concentrations in carbonaceous aerosol particles in the 0.056–18  $\mu\text{m}$  size range and to determine the relationship between the carbon amount variation in this size region and the stable carbon isotopic composition; to identify the origin of aerosol particles at a site far from anthropogenic pollution sources by combining the stable carbon isotopic composition and organic matter spectrometric measurement data.

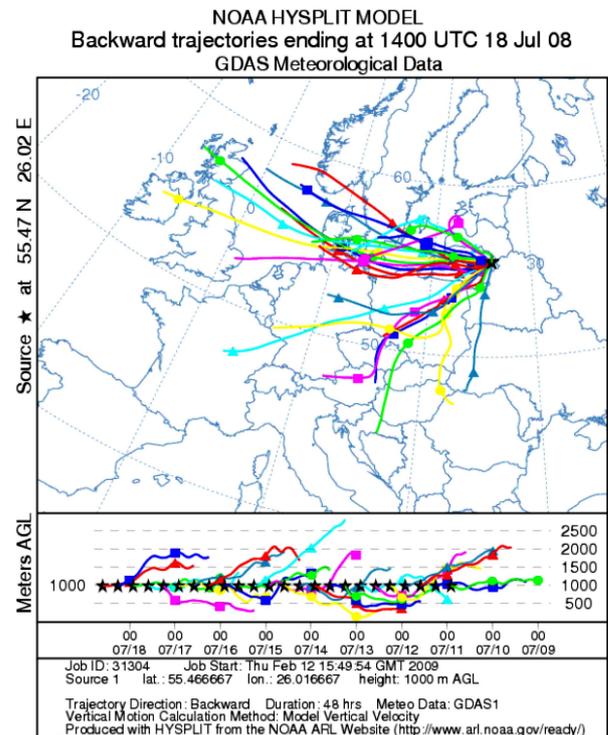
## 2. Experiment

### 2.1. Research site and sample collection

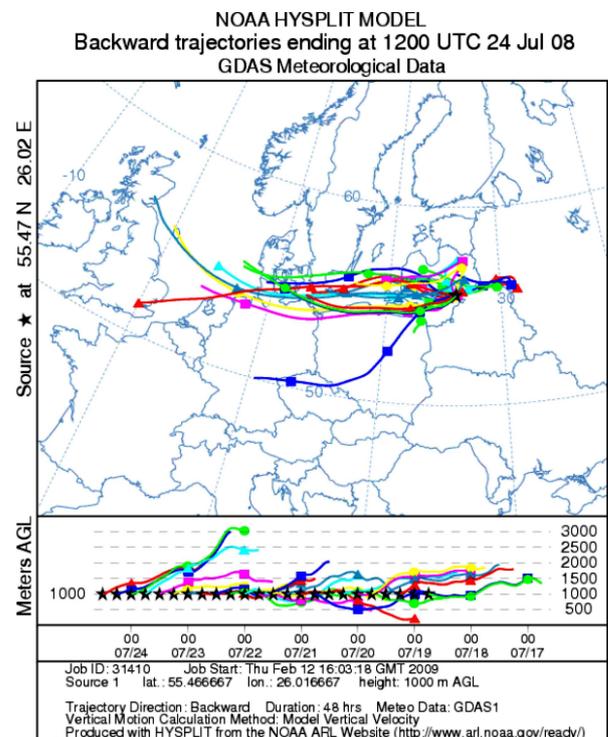
Investigations were carried out at the Rūgštelėškis Integrated Monitoring Station (IMS). This rural station is located in the eastern part of Lithuania (55°28' N, 26°01' E, 170 m above sea level) in a forested, hilly area about 300 km from the Baltic Sea. The nearest source of anthropogenic pollution, Utena town, is at a 25 km distance.

For the evaluation of organic matter (OM) concentration changes, air mass backward trajectories were calculated (Fig. 1) using HYSPLIT4 model and the meteorological data from National Oceanic Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) [13]. Three-day backward trajectories at 6-hour intervals (4 per day) were computed at the arrival height of 1000 m, for each of the two periods that lasted 8 and 6 days.

For carbon isotope analysis of the total carbon (TC) and EC in aerosol particles, atmospheric samples were collected with a Micro-Orifice Uniform Deposit Impactor (MOUDI, model No. 110, MSP, USA) during two periods, 10–18 July and 18–24 July 2008. The flow rate was 30 l  $\text{min}^{-1}$ , and the 50% aerodynamic cutoff diameters  $D_p$  of 11 stages were 18, 9.9, 6.2, 2.5, 1.8, 1.0, 0.56, 0.31, 0.18, 0.10, and 0.056  $\mu\text{m}$ . Aluminium foils were used as the impaction surface. These foils were baked before sampling at 600 °C for 3 hours to remove organic impurities. After sampling they were stored in a refrigerator. The foils were combusted with the elemental analyzer EA (*FlashEA 1112*) connected to



(a)



(b)

Fig. 1. Backward trajectories calculated by NOAA HYSPLIT4 model for air parcels ending at the Rūgštelėškis site: (a) period 1 (10–18 July 2008), (b) period 2 (18–24 July 2008).

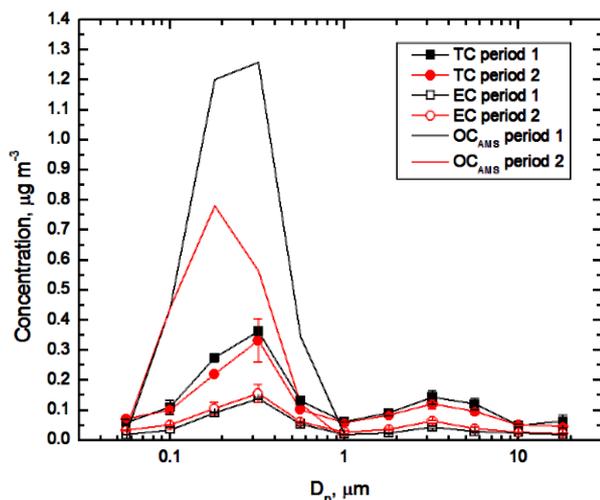


Fig. 2. TC and EC mass concentrations in aerosol particles of different size.

the stable isotope ratio mass spectrometer IRMS (*ThermoFinnigan Delta Plus Advantage*). For total carbon (TC) analysis, the 1/4th part of the foil was placed into the tin capsule and combusted in the oxidation furnace at the temperature of 1020 °C and with the oxygen excess. For more details see [14]. Elemental carbon (EC) analysis was performed as for TC, but before the analysis the foil was heated to 320 °C in the ambient atmosphere for the organic carbon removal.

All carbon isotope data are reported in the delta notation ( $\delta^{13}\text{C}$ ) relative to the Pee Dee Belemnite (PDB) standard as follows (in ‰):

$$\delta^{13}\text{C} = \left[ \frac{(^{13}\text{C}/^{12}\text{C})_{\text{sample}}}{(^{13}\text{C}/^{12}\text{C})_{\text{PDB}}} - 1 \right] \cdot 10^3, \text{‰}.$$

We used glucose ( $\delta^{13}\text{C} = -10.6 \pm 0.4\text{‰}$ ) as a working standard.

For measurement of the chemical composition and mass size distribution of organic matter present in/on ambient aerosol particles, at a temporal resolution of five minutes, quadrupole aerosol mass spectrometer (Q-AMS) developed at Aerodyne Research was used. A brief description of Q-AMS is given here, for more details see [15]. The Q-AMS consists of three major parts: a particle beam generation inlet system, an aerodynamic sizing chamber, and a particle composition detection section. The Q-AMS draws sampled air into a high vacuum through a critical orifice and focuses particles into a narrow beam with an aerodynamic lens. The inlet system allows 100% transmission efficiency for spherical particles with aerodynamic diameters between 60 and 600 nm. The vacuum aerodynamic diameter ( $D_{va}$ ) measured in the free molecule regime is calculated from the time-of-flight (TOF) measurement. In the composition detection section, non-refractory ma-

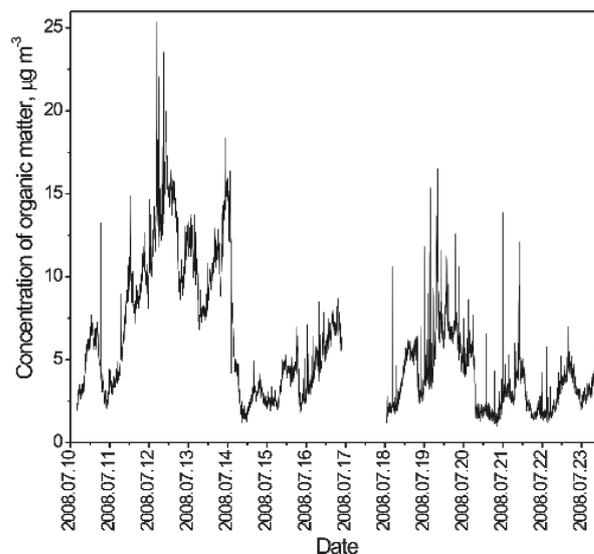


Fig. 3. Concentrations of organic matter in aerosol particles at Rūgštelėškis IMS during 10–24 July 2008.

terials in the particles are flash vaporized upon impact with a heated surface maintained at 600 °C, at a vacuum pressure of  $10^{-8}$  torr. The vapour molecules are ionized by electron impact (70 eV), and the positive ions formed are analysed by a quadrupole mass spectrometer with unit mass-to-charge ( $m/z$ ) resolution.

### 3. Results

#### 3.1. TC, EC, OC, and OM concentrations

The total carbon mass concentrations on different MOUDI stages are shown in Fig. 2. Considering that the mean TC concentration was  $1.47 \mu\text{g m}^{-3}$ , the ultrafine particle size range (stage 11,  $D_p = 0.056$ – $0.1 \mu\text{m}$ ) made up <5%. The accumulation mode particles (stages 10–6,  $D_p = 0.1$ – $1.8 \mu\text{m}$ ) including particles directly emitted and/or grown by coagulation and condensation processes amounted to about 70%, and the coarse size range (stages 5–1,  $D_p = 1.8$ – $18 \mu\text{m}$ ) including contributions of soil erosion, resuspension, and abrasion processes to about 30% of the mean TC concentration.

TC concentration is 18% in accumulation mode and 13% less in coarse mode in period 2 compared with period 1 (Fig. 2). At first sight there is no significant difference between results of two sampling periods, therefore air mass transport was analysed. During period 1 the dominating air mass transport was from western, northwestern directions, passing over the Baltic Sea. During period 2 air masses spread mainly over the continent from western direction (Fig. 1).

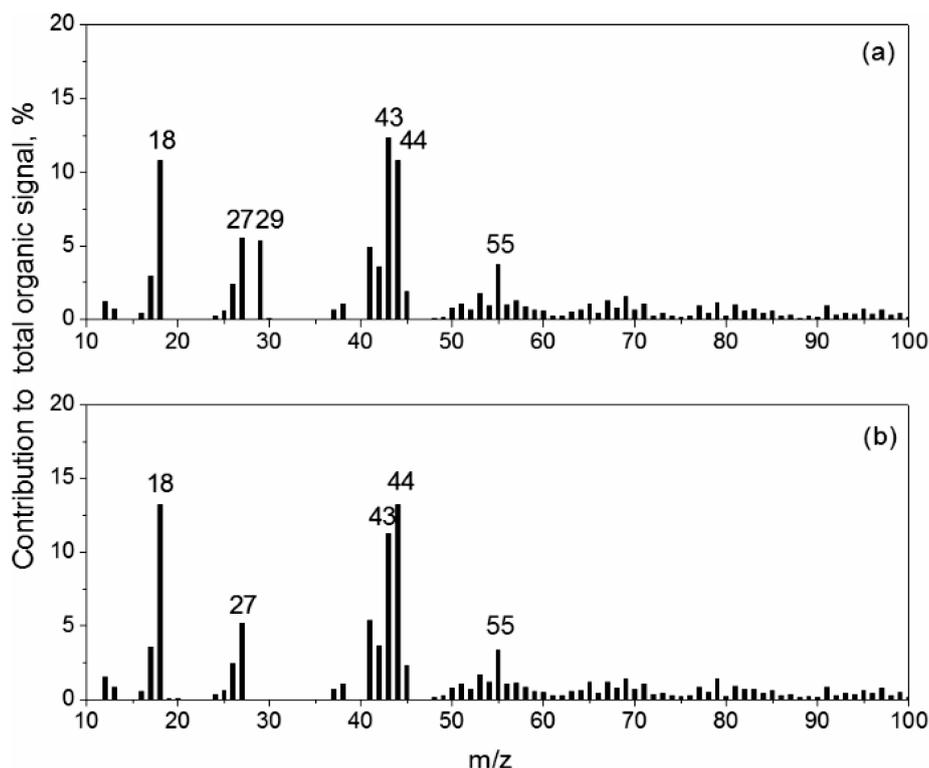


Fig. 4. Mass spectra of organic matter in aerosol particles in air mass transported from (a) Southern Europe, (b) the North Atlantic and the Baltic Sea.

Elemental carbon made up 10% of TC during period 1 and 18% during period 2.

The temporal variation of concentration for organic matter is shown in Fig. 3. Concentrations of organics were in the range of  $1.0\text{--}25.4\ \mu\text{g m}^{-3}$ , with an average of  $5.6\ \mu\text{g m}^{-3}$  and a standard deviation of  $3.7\ \mu\text{g m}^{-3}$ .

The changes in concentration of organic matter may be explained by the influence of long-range transport of aerosol particles. The long-range transport is related to the origin of air masses passing the Rūgštelėškis site.

Figure 3 shows that the highest concentrations of organic aerosol particles were measured during 13–14 July when the air masses were coming mainly from Southern Europe (Fig. 1). Those concentrations were noticeably higher than concentrations registered when air masses passed over Western Europe (19–20 July). The concentration considerably decreased (22 July) when air masses were coming from the North Atlantic, passing over the Baltic Sea. Generally, data revealed large variations in concentrations of organic species ranging within an order of magnitude depending on the air mass origin and history.

The derived mass spectra in comparatively clean air mass having passed over the North Atlantic and the Baltic Sea and in polluted air mass having arrived

from Southern Europe differ mainly in relative intensity rather than in the presence or absence of specific peaks.

The largest contributions to the spectrum (Fig. 4(a)) came from mass fragments characteristic of oxidized organic compounds such as  $m/z$  44 coupled with  $m/z$  18, signatures of di- and polycarboxylic acid, in addition to mass fragments 43 and 55 which could be associated with other types of oxidized organic compounds including aldehydes and ketones [16]. However,  $m/z$  44 is usually formed in anthropogenic and in biogenic processes and cannot be attributed solely to sources.

During the period when air mass have come from Southern Europe and have passed over polluted urban areas the relative abundance of peaks  $m/z$  43 is larger than in case of air mass coming from the Northern Atlantic. This mass spectrum has a significant intensity peak  $m/z$  29 attributed to the saturated alkanes. This may well indicate that rural oxidation products might be mixed together with oxidized urban aerosol.

Levoglucosan ( $m/z$  60) is a good indicator of biomass burning [17]. Absence of levoglucosan in the mass spectrum (Fig. 4) indicates that in submicron range the main source of aerosol particles has not been the biomass burning.

OC was evaluated dividing organic matter concentrations from Q-AMS by a factor of 2. Measurements have

Table 1. TC, EC, and  $OC_{(Q-AMS)}/EC$  in the aerosol particles at Rūgštelėškis site.

Stage No	$D_p$ , $\mu\text{m}$	Period 1 (10–18 July 2008)			Period 2 (18–24 July 2008)		
		TC, $\mu\text{g m}^{-3}$	EC, $\mu\text{g m}^{-3}$	$OC_{(Q-AMS)}/EC$	TC, $\mu\text{g m}^{-3}$	EC, $\mu\text{g m}^{-3}$	$OC_{(Q-AMS)}/EC$
1	18	0.06	0.02		0.05	0.02	
2	10	0.05	0.02		0.05	0.03	
3	5.6	0.12	0.03		0.10	0.04	
4	3.2	0.14	0.04		0.12	0.06	
5	1.8	0.09	0.02		0.08	0.04	
6	1.0	0.06	0.02	1.1	0.06	0.03	0.1
7	0.56	0.13	0.05	6.3	0.10	0.06	2.0
8	0.32	0.36	0.14	9.1	0.33	0.16	3.6
9	0.18	0.27	0.09	13.1	0.22	0.11	7.4
10	0.10	0.11	0.03	12.7	0.10	0.05	8.7
11	0.056	0.06	0.02	1.3	0.07	0.03	1.3

shown that using Q-AMS and MOUDI the OC ( $OC = TC - EC$ ) values differ from 2 to 9 times. Cabada et al. [18] have shown that more than 50% of the organic matter can be lost from the MOUDI aluminum foils during the summertime. OM losses during the summer are mainly due to volatilization of material caused by higher ambient temperatures and higher concentrations of secondary organic species. For the determination of the OC/EC ratio we used OC concentrations registered by Q-AMS, and those of EC collected by MOUDI and later measured by EA-IRMS. The size distribution measured with the Q-AMS and MOUDI has been recalculated to the geometric mean diameter. We have not transformed the vacuum aerodynamic diameter ( $D_{va}$ ) to the aerodynamic diameter ( $D_a$ ), because MOUDI resolution is lower than that of Q-AMS. It is not possible to determine the exact position of mode maximum.

Low OC/EC ratio was in the 0.56–1  $\mu\text{m}$  range (Table 1). Low OC/EC ratio values are indicators of traffic emissions, especially in the fine particle size range. High content of OC fractions could be originated from biogenic emissions (biogenic volatile organic compounds, plant abrasion) or anthropogenic sources (traffic, industry). OC from biogenic sources can be emitted as primary coarse particles (abrasion particles, fungi, bacteria, spores) or form secondary fine particles from gaseous precursors. As a result of tropospheric chemical reactions, volatile organic compounds of biogenic or anthropogenic origin form non-volatile products condensing as secondary organic aerosol (SOA) on all available particle surfaces.

The OC/EC values are found to be with a maximum of 13.1 on stage 9 ( $D_p = 0.18$ – $0.32 \mu\text{m}$ ) in the accumulation mode during period 1. OC/EC ratios of  $>10$  in accumulation particle mode are typical of background sites [19]. However, on stage 6 ( $D_p = 1$ – $1.8 \mu\text{m}$ ) the OC/EC ratio of 1.1 was observed (Table 1). This low

value can be caused by soot emissions of domestic fires and aged traffic emissions transported over longer distances. OC and EC concentrations in aerosol particles of accumulation mode are shown in Fig. 5. OC concentration was taken from Q-AMS, which has a detection range of  $<1 \mu\text{m}$  and OC could be underestimated, resulting in the low OC/EC ratio in aerosol particles of about 1  $\mu\text{m}$ .

Fresh traffic emissions from the near surroundings could not be considered as an important source as can be seen in the relatively low EC fraction of the particle mass on stages 10 and 11. Measurements at sites being strongly dominated by traffic resulted in an OC/EC ratio of about 0.3 [20]. In ultrafine particle size mode OC/EC ratios were about 1.3. This value shows that combustion processes significantly contribute to aerosols of this size mode.

### 3.2. Carbon isotopic ratio in size-segregated aerosol particles

The  $\delta^{13}\text{C}$  value variations of size-segregated aerosol particles collected with MOUDI are presented in Fig. 6. The averaged  $\delta^{13}\text{C}$  value at the Rūgštelėškis site was  $-27\text{‰}$  with clear transit from accumulation to coarse mode. The  $\delta^{13}\text{C}$  value of about  $-26.5\text{‰}$  for summer samples was reported in Zurich [21].

Measurement data show bimodal distribution of TC and EC in both investigation periods.

In accumulation mode,  $\delta^{13}\text{C}$  values ranged from  $-22.9$  to  $-26.3\text{‰}$  (average value  $-24.7 \pm 1\text{‰}$ ) for EC and from  $-26.7$  to  $-30.4\text{‰}$  for TC. Incorrect measurement probably caused unusual isotopic variation on stage 8 in period 2.

Comparable carbon isotopic ratios on all stages in accumulation mode show that aerosol particles in this mode have had the same source. The OC isotopic ratio

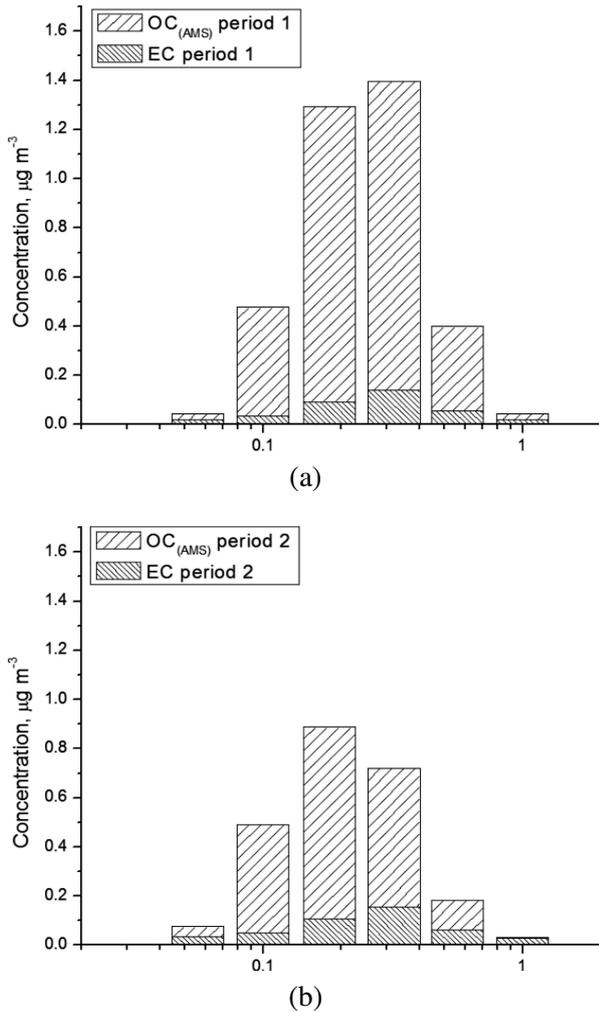


Fig. 5. OC and EC concentrations in aerosol particles of accumulation mode.

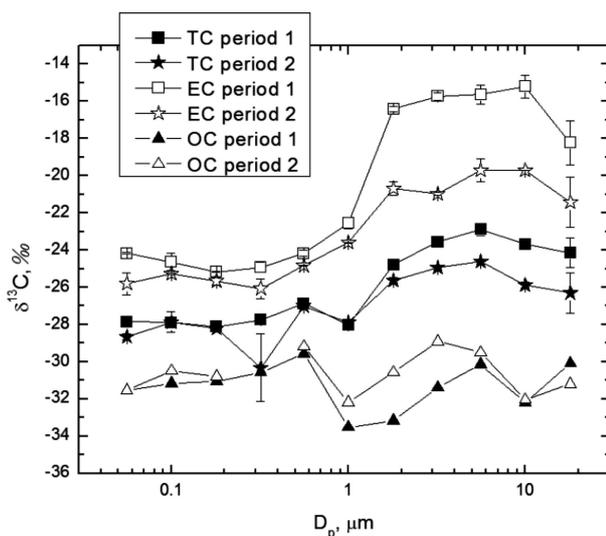


Fig. 6.  $\delta^{13}\text{C}$  variation in aerosol particles collected with MOUDI. Dashed line represents estimated OC isotopic ratios.

was derived from TC, EC isotopic ratios (Fig. 6). OC  $\delta^{13}\text{C}$  value of  $-31.1 \pm 1.4\text{‰}$  is typical of dominant local plants (coniferous wood). From presence of oxidized organics matter in AMS spectra it is not possible to tell that biogenic or anthropogenic source is dominant. By combining Q-AMS and isotopic ratio data it is evident that in accumulation mode aerosol particle precursors have had biogenic origin.

In coarse mode of aerosol particles the TC  $\delta^{13}\text{C}$  values ranged from  $-22.8$  to  $-26.3\text{‰}$  and those of EC from  $-15.2$  to  $-21.4\text{‰}$ . Such positive carbon isotopic values indicate that in coarse mode TC consists of a mixture of EC, OC, and unknown third constituent. Naturally, materials with higher  $\delta^{13}\text{C}$  values are carbonates, which could have been transferred with dust from surrounding areas at the Rūgšteliškis site due to elevated air temperatures with no precipitation, and they probably have determined such positive  $\delta^{13}\text{C}$  values in coarse mode aerosols.

#### 4. Conclusions

Organic matter concentrations and carbon isotope ratio measurements in size-segregated aerosol particles allow identifying sources of aerosol particles.

The  $\delta^{13}\text{C}$  values for the TC differ significantly in accumulation and coarse modes ( $-28$  and  $-24\text{‰}$ , respectively) and indicate a different origin of size-segregated aerosol particles. Almost constant organic carbon isotopic ratio (with  $\delta^{13}\text{C}$  value of  $-31.8\text{‰}$ ) in all aerosol particle size ranges indicates that the organic matter has originated from the local vegetation.

Carbon isotope ratio measurements on MOUDI stages have revealed that in accumulation mode aerosol particles have two sources: OC with  $\delta^{13}\text{C} = -31.1 \pm 1.4\text{‰}$  and EC with  $\delta^{13}\text{C} = -24.7 \pm 1\text{‰}$ . More positive  $^{13}\text{C}$  values in coarse mode indicate that TC consists of a mixture of EC, OC, and third constituent, probably carbonates.

Concentrations of organic matter were in the range of  $1.0$ – $25.4 \mu\text{g m}^{-3}$ , with an average of  $5.6 \mu\text{g m}^{-3}$  and a standard deviation of  $3.7 \mu\text{g m}^{-3}$ . The changes in concentration of OC may be explained by the influence of long-range transport of aerosol particles. The long-range transport is related to the origin of air masses passing the Rūgšteliškis site.

Presence of saturated alkanes in the registered mass spectrum indicates that rural oxidation products might be mixed with oxidized urban aerosol. Using only the Q-AMS data it is not possible to state that the biogenic or anthropogenic source is dominant.

Due to loss of organic material from MOUDI in summertime the OC amount for OC/EC determination from Q-AMS measurements was used. The highest OC/EC ratio was in the 0.056–0.18  $\mu\text{m}$  size range revealing a biogenic origin of aerosol precursors. Absence of levoglucosan in the Q-AMS mass spectrum indicated that during the measurement period there was no input from wood burning activities.

By combining carbon isotope data and Q-AMS measurements it is evident that in accumulation mode aerosol precursors have had a biogenic origin. Carbon isotope ratio measurements in size-resolved aerosols allow identifying aerosol sources more precisely. Combination of Q-AMS data with  $\delta^{13}\text{C}$  is a powerful tool for the source apportionment.

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**ANGLIES IZOTOPŲ SANTYKIAI ( $\delta^{13}\text{C}$ ) ĮVAIRIAUS DYDŽIO ANGLINGOSE AEROZOLIO DALELĖSE FONINĖJE LIETUVOS VIETOVĖJE**

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Pateikti anglies izotopinio santykio įvairaus dydžio aerozolio dalelėse Rūgšteliškio integruoto monitoringo stotyje 2008 liepos 10–24 d. tyrimų duomenys. Nustatyta, kad bendros anglies koncentracija kito nuo 0,06 iki 0,35  $\mu\text{g m}^{-3}$ , o elementinės anglies – nuo 0,02 iki 0,15  $\mu\text{g m}^{-3}$  aerozolio dalelių akumuliacinėje modoje bei atitinkamai nuo 0,06 iki 0,14  $\mu\text{g m}^{-3}$  ir nuo 0,02 iki 0,06  $\mu\text{g m}^{-3}$  stambiųjų aerozolio dalelių modoje. Tuo pačiu metu išmatuota organinių medžiagų koncentracija aerozolio dalelėse buvo 1,0–25,4  $\mu\text{g m}^{-3}$ , o vidutinė vertė – 5,6  $\mu\text{g m}^{-3}$ . Taikant HYSPLIT modelį oro masių pernašos atgalinių trajektorijų skaičiamui, nagrinėta aerozolių dalelių, atneštų oro masėmis į nutolusią

nuo antropogeninės taršos šaltinių vietovę, kilmę, siejant ją su išmatuotomis anglies izotopinių santykių vertėmis. Bendrosios anglies izotopiniai santykiai akumuliacinėje ir stambiųjų aerozolio dalelių modose žymiai skiriasi (–28 ir –24‰, atitinkamai). Tai rodo, jog įvairaus dydžio aerozolio dalelės yra skirtingos kilmės. Beveik pastovi organinės anglies izotopinio santykio vertė (–31,8‰) rodo, jog aerozolio dalelėse esantys organiniai junginiai emituoti vietinės augmenijos. Naudojant organinių junginių koncentracijos matavimus aerozolių masių spektrometrija ir stabilios anglies izotopų santykius įvairaus dydžio aerozolio dalelėse, atsiranda galimybė identifikuoti aerozolio dalelių kilmę ir vietinius bei globalius jų šaltinius.