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Effects of hydrocarbon contamination on the engineering geological properties of Neogene clays and Pleistocene glacial tills from Central Poland

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ABSTRACT:

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Petroleum products influence the engineering behaviour of the soil. Neogene clays and glacial tills from Central Poland were tested under laboratory conditions to evaluate the changes of selected physical and mechanical parameters: particle size distribution, particle density, swelling, shear strength and permeability. Four petroleum products were used in the experiments: diesel fuel, kerosene, jet fuel and mineral engine oil. The study revealed that even for the lowest degree of contamination the values of physical and mechanical properties of the soils changed significantly. Greater variation can be expected in soils contaminated with high-viscosity compounds. Also, higher relative changes were found for glacial tills than for Neogene clays. Consolidation tests revealed changes in soil permeability depending on the soil composition and the physical properties of the contaminant – considerable reduction of permeability was observed for glacial tills contaminated with light Jet fuel, while the reduction was lower for Neogene clays. The obtained results indicate the role of mesopores and the dimensionless pore pressure coefficient in changes of soil permeability. The methodological issues regarding testing and analysing the hydrocarbon-contaminated soils were also presented and discussed, which might be useful for researchers studying contaminated soils.

Key words: Petroleum products; Contamination; Clay soils; Permeability; Shear strength; Swelling; Particle size distribution.

INTRODUCTION

Oil and its derivatives are some of the most common soil pollutants in Poland, mainly because of leakages from fuel storage and pipeline systems, industrial premises, airports and petrol stations, or due to improper management of oil waste disposal. In Europe, the issue of soils contaminated with petroleum products and their remediation are among the most complex tasks in environmental protection with regard to the financial and organizational aspects (Streche *et al.* 2018). Oil contamination poses not only a significant threat to the environment, but also alters the physical and chemical properties of soils. It may also affect the mechanical behaviour of soils, therefore prediction of the engineering geological characteristics of contaminated soils is critical for geotechnical engineering (Cyrus *et al.* 2010; Karkush *et al.* 2013).

The key factor influencing the changes of the physical and mechanical properties of clay soils contaminated with liquid hydrocarbons includes physical-chemical processes at the boundary between the liquid and solid phases. Due to the low dielectric constant of non-polar hydrocarbons compared to water, the electrical double layer on the surface of contaminated clay particles (Birdi 2003) is reduced. In effect, there is significant reduction of electrostatic forces between the clay particles, and flocculation of clay particles is observed (Kaya and Fang 2005).

Processes taking place in the microstructure of contaminated soils are reflected in the macroscopic characteristics of the particle size distribution. Because of flocculation caused by contamination with oil-derived products, an increase of silt-size aggregates is observed, and thus there is a diminishing contribution of the clay fraction (Korzeniowska-Rejmer 2001; Izdebska-Mucha 2005; Gupta *et al.* 2009; Echeverri-Ramirez *et al.* 2015; Stajszczak 2019).

Adsorption of hydrocarbons on the surface of soil particles, and the low density of oil-derived products compared to water, result in lower values of bulk density and particle density of the soil as obtained in standard analyses; and in consequence, in the results of porosity calculations (Barański 2000; Chen *et al.* 2000; Ahmed *et al.* 2009, Rajabi and Sharifipour 2017; Stajszczak 2019; Hangshemo and Arabani 2022). The range of changes results from the physical properties of the contaminating phase and its content in the pore space of the soil.

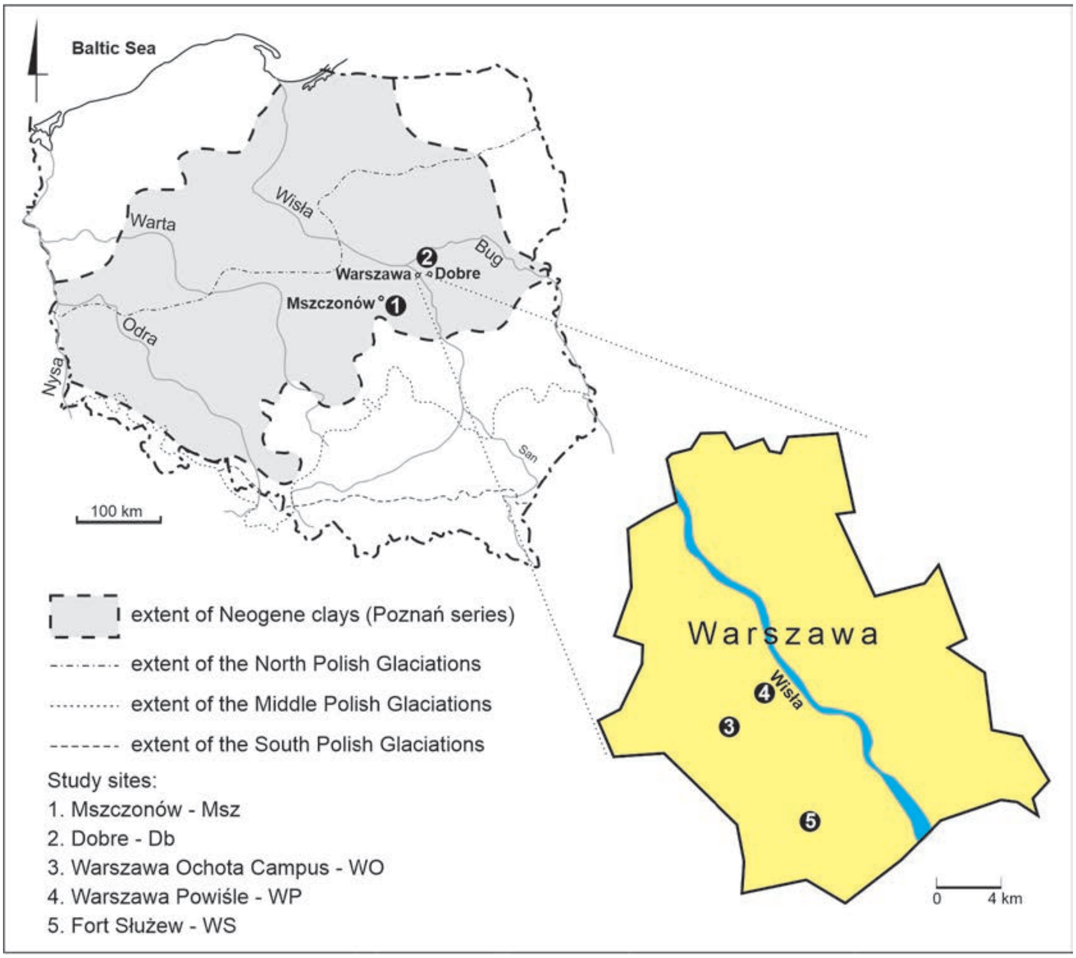
Barański (2000), Kaya and Fang (2000), and Izdebska-Mucha (2003) have shown that significant flocculation occurs in cohesive soils of various origin after full saturation with petroleum products. Contaminated soils lose cohesion, and thus determining their plastic limit becomes impossible. Izdebska-Mucha (2003), Khamehehiyan *et al.* (2007), Rahman *et al.* (2010), Nazir (2011), Oyegbile and Ayininuola (2013), Joseph and Hari (2015), and Raveendran and Poulose (2016) reported that hydrocarbon contamination results in reduction of the liquid limit from over ten to several percent. In turn, Srivastava and Pandey (1998), Barański (2000), Olchawa and Kumor (2007), Talukdar and Saikia (2010), Elisha (2012), Kermani and Ebadi (2012), Khosravi *et al.* (2013), Akinwumi *et al.* (2014), and Rasheed *et al.* (2014) observed increase of the liquid limit for cohesive soils after hydrocarbon pollution.

Hydrocarbon contamination modifies the expansiveness of cohesive soils. Low sorption of non-polar fluids by clay soils results from the fact that hydrocarbons cannot penetrate the interlayer space of clay minerals. Sorption of such compounds will thus depend on the external surface of the soil particles and the soil composition (Izdebska-Mucha 2005). Although the shrinkage properties of Neogene clays and glacial tills from Central Poland have been reported in the literature, there are few data on the swelling of contaminated soils, especially in Polish literature. Izdebska-Mucha and Wójcik (2021) showed significant influence of diesel oil contamination on the shrinkage properties of soils depending on the degree of pollution. Izdebska-Mucha and Korzeniowska-Rejmer (2010) reported changes of linear shrinkage and the shrinkage index of hydrocarbon contaminated soils and concluded that the presence of petroleum substances may significantly change the properties of soils used in the construction of mineral sealing barriers. In this light, this paper fills the gap and presents the influence of petroleum substances on selected parameters of swelling of Neogene clays and glacial tills from Poland.

Analysis of the shear strength of cohesive soils contaminated with hydrocarbons in most cases indicates that after contamination both the angle of internal friction and the cohesion attain lower values (Korzeniowska *et al.* 1995; Srivastava and Pandey 1998; Barański 2000; Siang *et al.* 2014; Nasehi *et al.* 2016). Due to their different density, viscosity and surface tension in comparison to water, hydrocarbons reduce the friction between soil particles, thus decreasing the angle of internal friction.

Changes of permeability in soils contaminated with petroleum products depend on the type and content of these substances, as well as the geological engineering properties of the contaminated soil (particle size distribution, porosity, microstructure, etc.). Increase of the coefficient of permeability due to hydrocarbon contamination (Mesri and Olson 1971; Barański 2000; Nazir 2011), coupled with decrease of permeability (Puri *et al.* in O'Shay and Hoddinott 1994; Silvestri *et al.* 1997; Khamehchiyan *et al.* 2007; Rahman *et al.* 2010; Huang and Lu 2014; Stajszczak 2021) were observed.

These variable trends of changes in physical, strength and permeability parameters require further investigations. Therefore, this study is focused on Neogene lacustrine clays and Quaternary glacial tills as representative and variable in terms of the genesis of geological environments in Central Poland. The impact of contamination with selected petroleum products on the physical and mechanical properties, crucial for subsoil behaviour, was tested. The study



Text-fig. 1. Location of the study sites with regard to the extent of Neogene clays and Pleistocene glacial tills.

was aimed at characterizing the often uncertain reaction of the soil to contamination, as well as the development of an analytical methodology necessary for this specific pore medium. Due to contamination of the analysed soils, the non-standard interpretation was applied and discussed.

GEOLOGICAL SETTING

The studies were performed on Neogene lacustrine clays and Pleistocene glacial tills from the Mazowsze region in Central Poland (Text-fig. 1).

Neogene clays

Clays of the Poznań Formation (Odrzywolska-Bieńkowa and Pożaryska 1981; Piwocki and Ziembińska-Tworzydło 1995) commonly occur in the Polish Lowlands (Text-fig. 1) as a thick succession of lacustrine deposits with traces of numerous marine ingressions. They represent the topmost part of the Neogene. Sedimentation of the Poznań Formation took place from the middle Miocene (c. 13 Ma) to the early Pliocene, and lasted for about 9 my. These deposits have often been referred to as: Mio-Pliocene clays, Pliocene clays, variegated clays, Tertiary clays, or as the Poznań series (Dyjor 1970) or, in the past, the Poznań clays (Berendt 1867; Jentzsch 1876).

The following horizons can be distinguished in the central part of the sedimentary basin: 1 – lower horizon – grey clays (marshlands, flooded peatland facies), 2 – middle horizon – green clays with glauconite (brackish facies), 3 – upper horizon – red clays (lacustrine facies). Clays of the Poznań Formation are usually c. 50–150 m thick (Dyjor 1970; Kozydra and Wyrwicki 1970). During the Pleistocene glaciations, the complex was repeatedly subject to the impact of the advancing ice-sheets, which had disturbed the primary setting of these deposits and caused signifi-

Text-fig. 2. Neogene brown-grey clay in the northern part of the Budy Mszczonowskie exposure; shovel for scale (photo by P. Stajszczak).

cant structural modifications. The cohesive, susceptible character of the Neogene clays favoured mechanical deformation and strain, and resulted in a rich spectrum of the effects of glacial activity, which have become typical glaciotectonic deformations in Poland. The observed changes are reflected as continuous or discontinuous plastic deformations in the topmost part of the clay complex. Folds with a diverse geometry, detachments, hidden and scarred surfaces of shears and overthrusts, lenses and xenoliths are commonly registered in the sub-glacial deposits. In effect, numerous exaration depressions and deformations with elevation differences often exceeding 100 m are commonly noted. Their rich geological history determining the engineering geological properties, coupled with their usage as a construction substrate is of interest to geologists and designers.

These deposits have been known for a very long time with regard to their economic usage (pottery, manufacture of bricks and tiles). They commonly occur in Warsaw and its vicinity. They are used as raw material for the production of construction ceramics. Their low permeability and very good sorption capacity and ion-exchange properties of the beidellite clays make them feasible as isolation material in waste dumps (Łuczak-Wilamowska 2013).

Samples of Neogene clays were collected from three sites: exposures in Mszczonów (cMsz), c. 45 km to the south-west of Warsaw, and Dobre village (cDb), c. 70 km to the east of Warsaw, and from cores of boreholes made in the Powiśle district in Warsaw (cWP).

Neogene clays occurring in Budy Mszczonowskie near Mszczonów have been exploited since 1963 as raw material for the production of expanded clay aggregate, and wall and roof elements. The clays occur in the form of a glaciotectonic floe, transported by the Scandinavian ice-sheet and squeezed into the deposits of the Odranian Glaciation (Wartanian Stadial) (Czarnecki and Czerny 1960; Czarnecki and Niedzielski 1961). They occur at various depths and are surrounded by glacial deposits. Clay and sandy sediments do not form regular beds but display large vertical and horizontal variability.

Macroscopically, the clays exploited in Budy Mszczonowskie represent various colour types (brown, brown-grey, black, blue, yellow, red), which result from their mineral composition, iron minerals in particular (Kozydra and Wyrwicki 1970). The red, rusty, and yellow-brown colour comes from the presence of hematite and goethite. Dispersed iron sulphides give grey-blue or greenish tints.

Samples collected as monoliths with an undisturbed structure and natural moisture content represent type II brown-grey clays (according to the subdivision by Łuczak-Wilamowska 2002), representing the most common clay type in the analysed exposure. Following norm PN-EN ISO 14688-1, soil represents stiff, grey, non-calceareous clay with numerous brown spots and smears, and low moisture content (Text-fig. 2).

The analyses were also performed on samples of Neogene clays from the clay deposit in the Wienerberger Ceramika Budowlana Sp. z o.o. brickyard, located c. 17 km to the north-east of Mińsk Mazowiecki and c. 1 km to the south of Dobre village. The exposure is situated at the junction of two roads linking Dobre with Rudzienko and Kamionka, respectively. The existence of a parallel structure composed of stacked Neogene deposits has been confirmed near Dobre (Piotrowska and Kamiński 2005). Between Dobre and Antonina the maximal elevation of their top occurs at 160 m a.s.l. (Nowak 1971a, b). The stacking extends for over 10 km in width and reaches to the Osownica valley to the east and to the Rządza valley to the west. To the south of the elevation, the top of the Neogene deposits rapidly drops to c. 80 m a.s.l.

Glacial tills

Glacial tills represent one of the most common Quaternary deposits in Poland. These soils were formed as a result of direct deposition of mineral material from the melting ice-sheet and could have inherited some features from the transport process (e.g., texture) (Lindner 1992). During the Quaternary, Scandinavian ice-sheets covered the area of Poland several times (e.g., Marks *et al.* 2016). Especially, glacial tills of the Middle Polish Glaciations (Odranian Glaciations) are subject to construction influence in the vicinity of Warsaw. Tills of the Odranian Glaciation occur commonly in Warsaw and its vicinity forming an almost continuous cover with a thickness from several to over a dozen metres in the western and south-western districts (Frankowski and Wysokiński 2000). In the sampling sites the tills occur in a c. 5–10 m thick horizon.

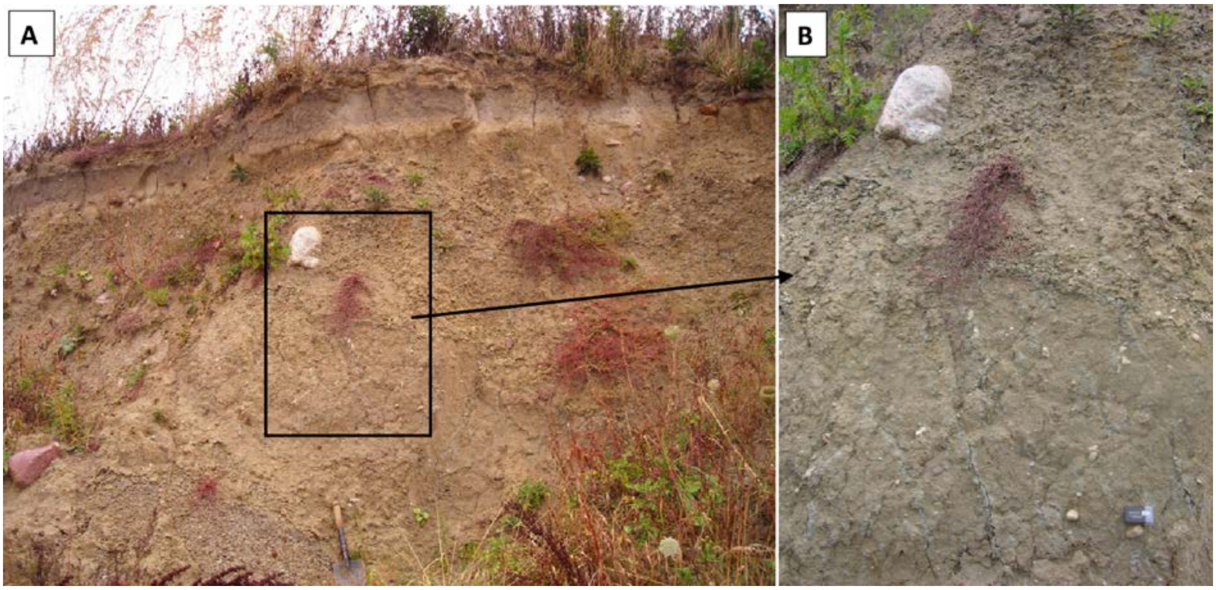
The choice of glacial tills for the laboratory tests resulted from the common occurrence of these deposits in Poland. The vast extent of glacial tills means that they often compose the substrate of foundations for objects that may be potential sources of contamination with oil-derived products (petrol stations, fuel depots, industrial plants, linear objects). Moreover, the different origin of glacial tills in comparison to the Neogene clays results in a different structure and particle size distribution than those for soils with an ice-dammed origin. This factor significantly influences their permeability as well as interactions between the soil solid phase and the oil-derived product contaminating the soil substrate.

Text-fig. 3. Two glacial till horizons (upper – brown and lower – grey) in a foundation excavation at the Ochota Campus of the University of Warsaw (photo by P. Zawrzykraj).

Text-fig. 4. Grey glacial tills in the excavation at the Ochota Campus of the University of Warsaw, depth c. 5 m (photo by P. Zawrzykraj).

The analysed samples of glacial till were collected from two localities in Warsaw and one in Mszczonów. The first locality was situated in the Ursynów district, close to the junction between Nowoursynowska and Dolina Służewiecka streets (tWS). Deep excavations for the foundations of the newly constructed ‘Fort Służew’ housing development allowed us to collect Odranian Glaciation till samples from c. 5–7 m below surface level.

The second locality was the area of the Ochota Campus of the University of Warsaw (tWO) on Banacha street, where an investment requiring deep foundation excavations allowed access to two glacial till horizons from the Odranian Glaciation (Text-figs 3 and 4). Brown tills occurred at c. 5 m and grey tills – at c. 10 m from the surface level in the excavation walls. The thickness of both horizons was similar,



Text-fig. 5. Glacial tills in the southern wall of the Mszczonów exposure (photo by M. Wilk).

at c. 5 m. The contact of the two horizons was highlighted by a several centimetre thick layer of sands and silts with an admixture of organic matter. In some places in this horizon, larger accumulations of plant fragments in form of peat were observed. At the present stage of studies, the upper till horizon is linked with the Wartanian Stadial of the Odranian Glaciation.

Samples of glacial till (tMsz) were also collected in the Przedsiębiorstwo Produkcji Kruszyw Lekkich 'Keramzyt' in Mszczonów (Text-fig. 5a). Glacial tills assigned to the Wartanian Stadial of the Middle Polish Glaciations (Lindner 1992) are exposed in the southern wall of the excavation pit (field studies were performed here in 2009). According to macroscopic analyses, the soil represents silty clay with sand (sasiCl), characterized by light-brown colour, stiff consistency, low moisture content, and $CaCO_3$ content of 3 to 5% (calcite veins are present; Text-fig. 5b). Erratic boulders, fractures, detachments, slip surfaces and fragments of sandy sediments can be observed in the exposure, which indicates that the analysed till is a lodgement till.

MATERIALS

Soils

The general characteristics of the analysed cohesive soils from Warsaw and its vicinity (Mazowsze) were based on 50 samples with an undisturbed structure, characterized by a variable cohesion and different origin (30 samples of Neogene lacustrine clays and 20 samples of glacial tills). Table 1 presents the physical parameters of the analysed soils depending on the locality and study area, as well as the typical values (max, min, average) of parameters, which decide on their geological engineering properties.

Neogene clays

The lithology of the Neogene clay deposits is restricted mainly to clays with an average content of the clay fraction at 37.0–67.0%, silt fraction at 29.0–41.0%, and sand fraction at 4.0–22.0% (Table 1). Clays from Mszczonów (cMsz) and Dobre (cDb) have a similar particle size distribution, whereas the sample from Warsaw (cWP) contains also silty clays.

The density state, reflected by natural bulk density, points to larger density of the Neogene clays from Warsaw (cWP) compared to clays from the other study sites. The total porosity of cohesive soils from Mszczonów (cMsz) and Dobre (cDb) varies in a similar range, whereas clays from Warsaw have lower values of this parameter.

The average value of the liquidity index I_L points to stiff and very stiff consistency. The plasticity index of the deposits varies in the following order: Ip cDb > Ip cMsz > Ip cWP. With regard to activity, the analysed clays may be classified as inactive and normal following the classification of Head (1992). The

	Neogene clays n = 30									Glacial tills n = 20								
	cMsz n = 13			cDb n = 12			cWP n = 5			tWS n = 8			tWO n = 7			tMsz n = 5		
	Mean Min–Max	M	s	Mean Min–Max	M	s	Mean Min–Max	M	s	Mean Min–Max	M	s	Mean Min–Max	M	s	Mean- Min–Max	M	s
Water content w_n [%]	28.9 21.6–34.1	28.7	3.8	28.6 21.8–35.0	28.4	4.5	19.8 15.3–26.9	18	3.9	17.4 15.3–18.9	18.1	1.3	13.0 11.8–13.9	13.0	0.7	14.2 12.8–15.6	14.6	1.1
Plastic limit w_P [%]	26.8 18.9–30.5	28.3	3.6	30.2 23.7–39.3	30.6	4.6	23.3 16.4–33.1	24.1	5.8	16.9 15.1–18	17.5	1.1	11.4 10.2–12.7	11.3	0.9	13.2 12.4–14.3	13	0.7
Liquid limit w_L [%]	73.4 52.8–88.0	75.2	11.1	81.3 60.1–111.9	79.7	14.8	51.6 38.8–82.2	45.2	15.6	43.8 40.3–46.2	45.0	2.2	28.8 22.4–35.1	29.6	4.4	24.0 23.3–25.8	23.6	0.9
Plasticity index I_P [%]	46.6 33.9–60.0	49.6	8.6	51.1 35.4–73.4	50.2	10.8	28.3 20.4–49.1	23.4	10.7	26.9 24.9–28.5	27.4	1.1	17.3 12.3–22.08	17.8	3.5	10.8 10.0–11.5	11.2	0.6
Liquidity index I_L [-]	0.04 -0.05–0.11	0.05	0.1	-0.03 -0.09–0.13	-0.06	0.1	-0.13 -0.29–0.11	-0.13	0.2	0.02 -0.06–0.11	0.03	0.0	0.10 0.00–0.23	0.08	0.1	0.1 0.04–0.16	0.11	0.1
Activity A [-]	0.71 0.47–0.91	0.77	0.2	0.79 0.47–0.93	0.84	0.1	0.76 0.63–0.92	0.75	0.1	0.72 0.63–0.89	0.71	0.1	0.87 0.76–0.92	0.89	0.1	0.44 0.41–0.48	0.43	0.0
Clay (<0.002 mm) f_i [%]	67 51–88	66	9.5	66 44–85	63	13.1	37 29–61	31	12.2	38 31–44	38	3.5	20 14–24	21	3.6	25 24–27	24	1.2
Silt (0.05–0.002 mm) f_π [%]	29 11–40	31	7.4	29 10–54	29	13.1	41 25–57	38	11.7	28 22–38	27	4.9	26 23–28	27	2.0	17 13–23	17	3.2
Sand (2–0.05 mm) f_p [%]	4 0–9	3	2.5	5 2–14	4	3.5	22 1–45	18	16.6	35 27–39	36	4.3	53 47–60	51	4.9	58 52–61	59	3.3
Gravel (>2 mm) $f_ż$ [%]	0 0–0	0	0.0	0 0–0	0	0.0	0 0–0	0	0.0	0 0–0	0	0.0	1 1–3	1	0.7	1 0–2	0	0.8
Specific density ρ_s [Mg/m^3]	2.73 2.67–2.81	2.7	0.1	2.69 2.66–2.73	2.69	0.0	2.68 2.65–2.74	2.67	0.0	2.67 2.66–2.67	2.67	0.0	2.68 2.67–2.69	2.68	0.0	2.70 2.68–2.71	2.70	0.0
Wet/bulk density ρ [Mg/m^3]	2.00 1.92–2.09	1.97	0.1	1.99 1.83–2.08	2.02	0.1	2.07 1.97–2.15	2.08	0.1	2.12 2.1–2.14	2.12	0.0	2.09 2.04–2.14	2.08	0.0	2.21 2.17–2.25	2.21	0.0
Dry density ρ_d [Mg/m^3]	1.55 1.43–1.74	1.51	0.1	1.55 1.38–1.71	1.58	0.1	1.73 1.58–1.82	1.8	0.1	1.81 1.77–1.86	1.80	0.0	1.85 1.80–1.90	1.84	0.0	1.94 1.88–1.99	1.93	0.0
Porosity n [%]	43 37–47	44	0.0	42 36–48	41	3.8	35 32–42	32	4.1	32 30–33	33	0.0	31 29–33	31	1.3	28 26–30	29	0.0
Void ratio e [-]	0.76 0.59–0.89	0.77	0.1	0.74 0.57–0.93	0.7	0.1	0.56 0.47–0.74	0.48	0.1	0.47 0.43–0.5	0.48	0.0	0.45 0.41–0.49	0.45	0.0	0.39 0.36–0.43	0.4	0.0

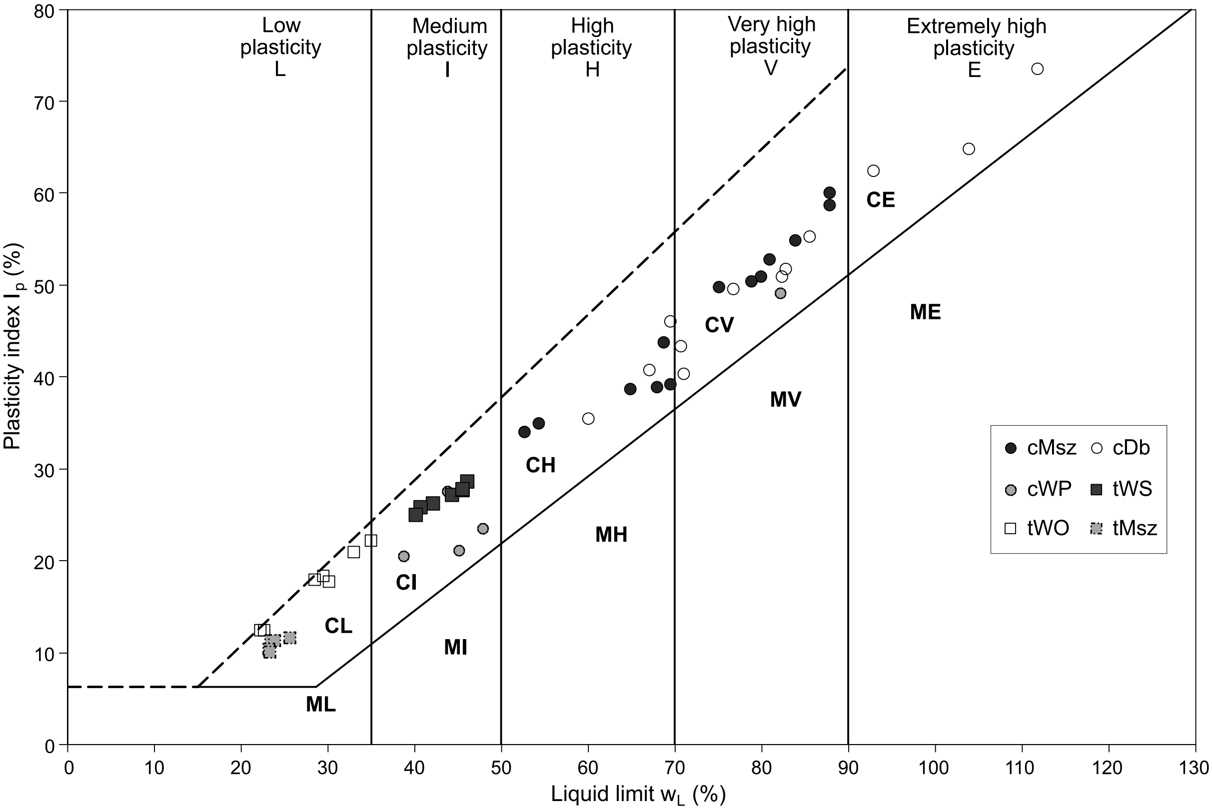
Table 1. Selected geological and engineering properties of Neogene clays and glacial tills. Explanations: c – clays; t – tills; Db – Dobre; WP – Warszawa Powiśle; WS – Fort Służew; WO – Ochota Campus; Msz – Mszczonów; n – number of samples, M – median, s – standard deviation.

Minerals	cMsz	cDb	cWP	tWS	tWO	tMsz
Clay minerals [%]	65.1	56.2	44.8	31.4	21.1	20.1
Beidellite [%]	54.7	41.4	36.7	17.6	12.4	8.4
Kaolinite [%]	10.4	14.8	8.1	3.2	3.4	6.8
Illite [%]	–	0.0	0.0	10.6	5.3	4.9
Goethite [%]	3.4	3.5	2.2	–	1.9	2.1
Calcite [%]	0.0	0.0	0.0	5.6	7.3	–
Thermally inactive minerals and quartz [%]	31.5	40.3	51.4	63.0	69.8	77.8

Table 2. Mineral composition of samples from selected localities based on derivatographic analysis.

soils have a variable plasticity. The Casagrande chart (Text-fig. 6) shows that the geological engineering parameters of the Neogene clays allow the assignation of them to several categories, i.e., clays with a medium to extremely high plasticity (CI, CH, CV, CE). Numerous authors (Kaczyński and Grabowska-Olszewska 1997; Izdebska-Mucha and Wójcik 2014, 2015; Stajszczak 2017) have characterized these clays as expansive.

The clays are characterized by a high content of clay minerals reaching 65%. The dominating mineral is beidellite with subordinate kaolinite (Table 2). These result are concordant with the mineral compo-



Text-fig. 6. Classification of Neogene clays and glacial tills used in the study according to the Casagrande chart (modified by Head 1992).

sition of the clay fraction from Budy Mszczonowskie (Łuczak-Wilamowska 2002) and Warsaw (Kaczyński 2003). The content of quartz and other thermally inactive minerals varies from 31 to almost 52%. Goethite reaches just 3.5% in the analysed clays.

Glacial tills

The particle size distribution of the glacial tills studied is extremely variable. The deposits are dominated by the sand fraction – the average content varies within 35–58%. The content of the silt fraction is within 17–28%, and the clay fraction at 20–38%. The content of the gravel fraction was observed in a few samples at levels reaching maximally 3% (Table 2). The lithology includes sandy clays and silty clays. Tills from the vicinity of Warsaw (tWS and tWO) are characterized by similar values of typical parameters such as specific density, bulk density, porosity, and porosity index compared to the tills from Mszczonów (tMsz). The mean value of the liquidity index I_L points to a stiff and very stiff consistency, and the plasticity index changes according to the following order: *Ip* tWS > *Ip* tWO > *Ip* tMsz. The glacial tills are represented by soils with a low and medium plasticity (CL, CI).

The mineral composition of the analysed soils is more variable. The main components of the tills include quartz and thermally inactive minerals. The clay mineral content in the analysed tills varies within 20–32%. The main clay mineral is beidellite with a significant content of kaolinite and illite (Table 2). Moreover, a high carbonate content (up to 7.3%) and goethite admixtures (up to 2.1%) have been noted in samples from two localities.

The soils are characterized by inhomogeneity of the basic physical parameters. Their variable properties are the effect of a complex geological history and acquire multifaceted characteristics on a regional scale. Samples selected for detailed studies represented physical properties typical of their formations, and the values of their physical parameters are in accordance with literature data (Kaczyński 2003, 2011, 2017).

Petroleum products

Four petroleum products were used in the experiments: diesel fuel (ON), kerosene (Ke), jet fuel (Jet

Parameter	Diesel fuel (ON)	Kerosene (Ke)	Jet fuel (Jet A1)	Mineral oil 15W40 (MO)	Water
Physical state at room temperature	liquid				
Initial boiling point	175–180ºC 95% vol. distills up to 360ºC	180–310ºC	130–290ºC	no data	100ºC
Density at 15 ℃ [g/cm^3]	0.82–0.845	0.75–0.84	0.775 – 0.840	0.882	1.00
Water solubility	insoluble and immiscible				–
Kinematic viscosity [mm^2/s]	2.0–4.5 (at 40ºC)	< 3 (at 40ºC	1.25–1.75 (at 20–40ºC)	13.5–16.3 (at 100ºC)	0.30 (at 20ºC)
Dielectric constant	~2	1.8	~2	~2	~80
Hydrocarbon composition	C_9–C_{25}	C_9–C_{16}	C_9–C_{16}	C_{20} – C_{24}	–

Table 3. Physical and chemical properties of petroleum products and water (Surygała 2000; Safety data sheets 1998, 2002, 2005, 2006).

A1) and mineral engine oil 15W40 (MO). Their crucial physical and chemical properties compared to water are listed in Table 3. The products represent Light Nonaqueous Phase Liquids (LNAPLs), which are hydrocarbons lighter than water. Petroleum products used in this study are composed of hydrocarbon compounds ranging from C_9 to C_{25}, which are insoluble in water. Physical properties of petroleum products vary according to their composition. However, in comparison to water, all products are characterized by lower density, higher viscosity, extremely low dielectric constant, and higher range of boiling temperature. In our study we will point out how these differences influence the soil geotechnical properties.

METHODS

Comparative laboratory analyses of clean soils and soils contaminated with oil-derived products were performed on soil pastes. Pastes, made out of natural samples collected from exposures, have a natural particle-size and mineral composition, and, contrary to natural samples, a homogenized structure in which natural interparticle bonds were destroyed. The homogeneity of soil pastes and the controlled contamination of the samples studied assure repeatability of the initial conditions in the analyses and comparability of the obtained results.

Particle density

Particle density (ρ_s) was determined for the <2 mm soil fraction in a helium gas pycnometer (AccuPyc 1330, Micromeritics). Prior to the measurements, the soil samples were dried at 105ºC.

ON-contaminated samples of Neogene clays (cMsz) and glacial tills (tMsz) from Mszczonów were used in the particle density, particle size distribution and swelling tests. Natural soil samples were air-dried, pulverised to pass the 2 mm sieve, then oven-dried at 105ºC and mixed with diesel fuel (ON) in the amount of 0 (reference sample), 4, 8, 12, 16% by dry weight of the soil. The mixtures were kept in closed containers at room temperature for 3 months.

Particle size distribution

Particle size distribution (PSD) was determined by the microaggregate method following Kaczynski's procedure (after Myślińska 2016). The soil samples were wet sieved on a 0.063 mm sieve and further measurements were carried out using hydrometer (cMsz ON, tMsz ON) or pipette methods (cMsz Jet A1, MO; tWS Jet A1, MO). The coarse fraction was determined by sieve analysis. Microaggregate analysis better reflects the natural particle size composition of a soil, as the procedure does not include the addition of a dispersing agent in the sample preparation. Such approach is also in accordance with the European standard PKN-CEN ISO/TS 17892-4:2009, which does not recommend using a dispersing agent unless coagulation of a soil suspension is observed.

Swelling

The comparative assessment of soil swelling for natural samples of Neogene clays from Warsaw (cWP) and Dobre (cDb) in deionized water and kerosene (Ke) were performed based on the modified free swell index (MFSI) proposed by Sridharan *et al.* (1985) according to the formula:

$$\mathrm{MFSI} = \frac{\mathrm{V_d}}{10}$$

where: MFSI is the modified free swell index (cm^3/g), $\mathrm{V_d}$ is the volume of 10 g of soil after sedimentation (cm^3), and 10 represents the dry mass of the tested material (g).

Soil samples with a mass of 10 g, sieved through a 425 µm mesh were placed in 100 cm^3 graduated cylinders and deionized water or kerosene was added.

Swelling properties of ON-contaminated Neogene clays (cMsz) and glacial tills (tMsz) from Mszczonów were analysed based on free swelling (FS) and swelling pressure (σ_{sp}). Free swelling was determined following ASTM D 4546-90 (method A). Swelling pressure was determined following ASTM D 4546-90 using the automatic swelling pressure apparatus h-200 A (Geonor, Norway). The constant volume method (method C) was employed. The initial moisture content of the contaminated soils was related only to the contaminant content, and the control soil samples (uncontaminated) were air-dry. All the samples were tested with deionized water. Prior to free swelling and swelling pressure tests, the samples were consolidated to obtain a similar dry density. Discussion of the swelling properties was completed with results of methylene blue capacity (MBC) measurements. The MBC was determined following PN-B-04481 (1988).

Direct shear tests

The shear strength of glacial tills from Warsaw Ochota Campus (tWO) was tested in a standard direct shear test apparatus following ASTM D3080-04. The tests were performed on remoulded samples contaminated with diesel fuel (ON) in laboratory conditions. The procedure of sample preparation allowed us to obtain homogeneous soil mixtures with bulk density and moisture content as similar as possible to the undisturbed samples. Firstly, the natural soil samples were air-dried, pulverized to pass the 2 mm sieve and oven-dried at 105ºC. Then, the sample was divided in three equal portions – one represented the reference sample (0% ON), whereas the remaining two were mixed with ON in the amount of 4% and 8% by dry weight of the soil. All the samples were kept in closed containers at room temperature for 1 month to allow ageing and possible reactions between the contaminant and soil. Afterwards, the equilibrated samples were mixed with deionized water in the amount of 15% corresponding to the natural moisture content of the tested glacial tills. The mixtures were again kept at room temperature for one month to reach equilibrium. Then, the contaminated samples were placed in layers up to 1 cm in thickness in a mould (dimensions 62 × 62 × 120 mm). Each layer was slightly tamped. Finally, soil in the mould was consolidated under a constant pressure of 50 kPa. Bulk density values of all samples were similar (0% ON – 2.15 Mg/m^3, 4% ON – 2.11 Mg/m^3, 8% ON – 2.09 Mg/m^3), however they slightly decreased in the contaminated samples due to the additional presence of ON as pore fluid.

Permeability

The coefficients of permeability for the glacial tills from Warsaw Służew (tWS) and the Neogene clays from Mszczonów (cMsz) were determined for uncontaminated soil pastes and pastes contaminated with two types of fuel differing in physical properties. Values of coefficient of permeability were assessed by interpretation of the results of consolidometer tests CRL (constant rate of loading) based on solutions of the consolidation theory (Dobak 1999) adapted to these conditions. Consolidation tests were conducted in a Barden-Rowe-type consolidometer specially adapted (Stajszczak 2019) to studies of contaminated soils. The procedure of soil preparation allowed for a controlled and uniform distribution of water in the pore space with an assumed contaminant content and for obtaining full soil saturation with water.

Preparation of contaminated soil pastes included soil pulverization, sieving on a 400 µm mesh and homogenization with application of deionized water, followed by drying. Air-dry samples were optionally mixed with Jet fuel A1 or mineral engine oil 15W40 in amounts corresponding to 0 (reference sample), 2, 5, 10 and 20% of the soil mass. After several weeks of homogenization in tight vessels, the contaminated samples were mixed with deionized water to obtain moisture content corresponding to 1.0−1.1w_L of the uncontaminated soil. Samples prepared according to this procedure were carefully mixed after 24 h and then inserted in specially constructed chambers for initial consolidation. The soil samples were gradually loaded for 4−5 weeks to reach the stress level 20 kPa, so that the soft plastic consistency was obtained. Samples for the consolidometer rings were cut out from such homogenous paste; on such samples, tests of particle density, particle size distribution and microstructural analysis were performed.

Microstructure

Microstructural analysis was performed with application of a scanning electron microscope (SEM), model JSM 6380LA, JEOL, and software for quantitative image analysis STIMAN – STructure IMage ANalysis (Sokolov *et al.* 2002; Trzciński 2004).

RESULTS AND DISCUSSION

Particle density

The particle density of hydrocarbon-contaminated soils decreased with increasing contaminant content. Text-fig. 7 shows the results for glacial tills (tMsz) and Neogene clays (cMsz) from Mszczonów, glacial tills from Warsaw Służew (tWS), and selected literature data. Reduction of ρ_s significantly depends on the soil composition and contaminant type. Comparison of equations (Text-fig. 7) shows that for a given petroleum product (i.e., diesel/jet fuel or mineral oil), the rate of change is higher for glacial tills than for Neogene clays. Evidently, a higher reduction was revealed for samples contaminated with mineral engine oil compared to soil contaminated with fuels (compare tWS MO, cMsz MO *vs.* tWS Jet A1, cMsz Jet A1 in Text-fig. 7).

Reduction of particle density is probably due to the presence of oil hydrocarbons $>C_{12}$ adsorbed on the soil mineral skeleton. These compounds have a lower density than minerals and do not completely evaporate at 105ºC (following standard sample preparation). A higher viscosity of the hydrocarbons results in a higher adsorption on soil particles, and thus in the linear reduction of ρ_s with increasing hydrocarbon content. Measurements of ρ_s according to standard procedures do not show alteration of the mineral phase, but are indicative of the presence of oil hydrocarbons in the soil. According to these findings we propose to describe the measured parameter as apparent particle density.

Particle size distribution

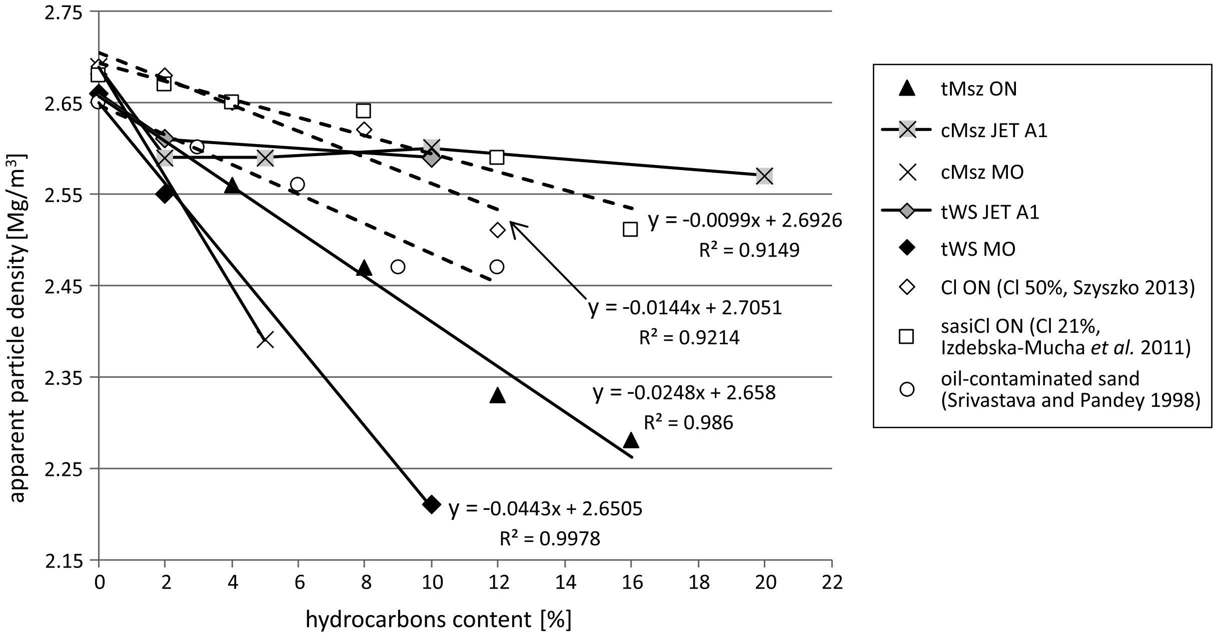
Particle size distribution (PSD) was analysed for the following soils and contaminants:

Neogene clays from Mszczonów (cMsz) contaminated with diesel fuel (ON), jet fuel (Jet A1) and mineral engine oil (MO);

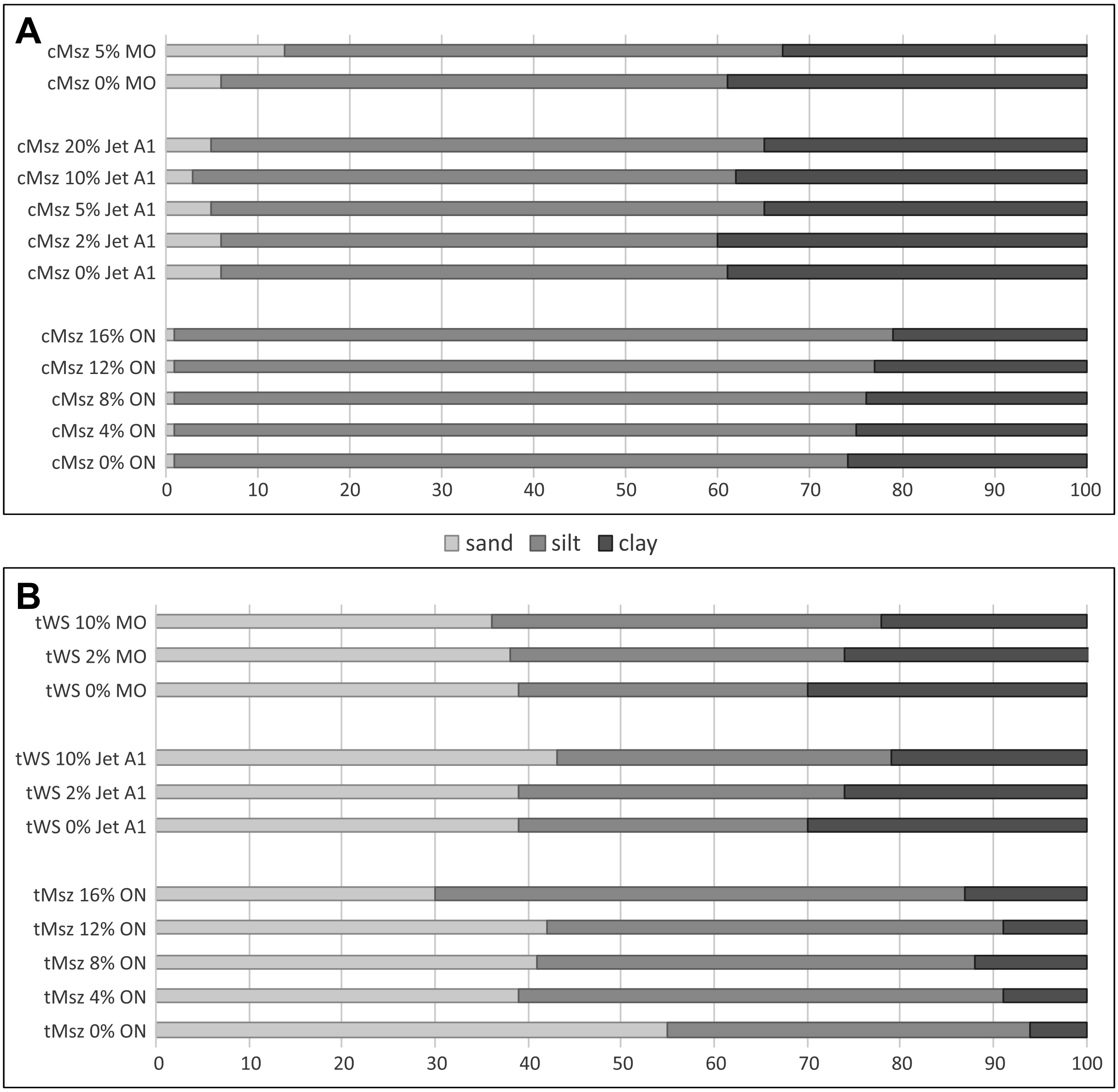
- glacial tills from Mszczonów (tMsz) contaminated with diesel fuel (ON);
- glacial tills from Warsaw Służew (tWS) contaminated with jet fuel (Jet A1) and mineral engine oil (MO).

Results of the combined sieve-microaggregate method for Neogene clays and glacial tills are presented in Text-fig. 8A, B. The results have revealed that for all contaminated mixtures of Neogene clays with increasing hydrocarbon content the clay content decreased (by 4–6% points) and the silt content increased (by 5% points). The sand content increased by 7% points for the MO-contaminated sample, whereas an opposite effect was noted for samples contaminated with jet fuel (reduction by 1–4% points). Despite the higher degree of contamination, jet fuel and diesel fuel caused similar changes in the soil composition as mineral oil.

Contaminated glacial tills also showed increase in the silt content (by 5–18% points), but the trend of changes in sand and clay contents is not the same for



Text-fig. 7. Particle density of soils contaminated with petroleum products.



Text-fig. 8. Particle size distribution of samples contaminated with diesel fuel (ON), jet fuel (Jet A1) and mineral engine oil 15W40 (OM). A – Neogene clays from Mszczonów (cMsz); B – glacial tills from Mszczonów (tMsz) and Warsaw Służew (tWS).

all mixtures (Text-fig. 8B). Glacial tills from Warsaw Służew (tWS) contaminated with Jet A1 and MO showed reduction of the clay content by 9 and 8% points, respectively, whereas in the tMsz ON samples the clay content increased by 7% points. The sand content generally tends to decrease with the increasing content of ON and MO. Higher values of changes in the PSD of glacial tills compared to Neogene clays may suggest that glacial tills are more sensitive to the transformation of their particle size (textural) composition due to hydrocarbon contamination.

The obtained results can be attributed to the physical-chemical interactions between the hydrocarbons and the soil particles. Hydrocarbons, as non-polar compounds with a low dielectric constant, cause reduction of the electric double layer thickness as well as interparticle forces (Kaya and Fang 2000, 2005). As a result, clay particles tend to flocculate and form larger (silty) assemblages, which explains the increase of silt content in the contaminated samples. This effect was observed in a simple sedimentation test of fine fractions in water and diesel fuel (Text-fig. 9).

Text-fig. 9. Sedimentation test of fine fractions (<0.063 mm) in distilled water (left cylinder) and diesel fuel (right cylinder), after 6 days.

The test showed that in diesel fuel, fine particles coagulated and behaved like fine sand. Moreover, hydrocarbons with a viscosity higher than water cause ‘gluing’ of the soil particles, and formation of larger structural elements. This effect is responsible for the relatively significant reduction of the clay content and increase of silt and/or sand content in soils contaminated with mineral oil. On the other hand, long-term interactions between soil and hydrocarbons lead to disintegration of the primary structural elements and their rearrangement (Izdebska-Mucha and Trzciński 2021). As a result, increase of fine fractions may be observed. A similar effect was reported by Jia *et al.* (2010, 2011). Changes of PSD in the contaminated soils are the combined result of these processes.

Our findings show general trends and allow for a preliminary estimation of the range of changes that can be expected in the tested soils after contamination with different petroleum products. The obtained results are generally in line with published reports on the cohesive soils from Poland contaminated with petroleum products (Korzeniowska-Rejmer and Izdebska-Mucha 2006; Bobrowska 2008; Izdebska-Mucha and Korzeniowska-Rejmer 2010; Izdebska-Mucha and Trzciński 2011; Izdebska-Mucha *et al.* 2021).

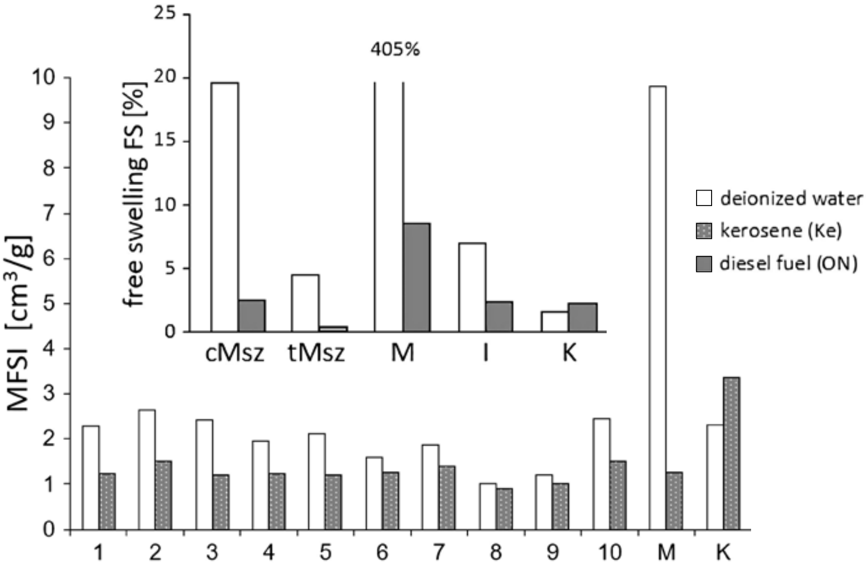
Swelling

There are several methodological and interpretational models for assessing soil swelling. In this work we tested: 1) samples of natural (uncontaminated) soils in water and different non-polar organic fluids (Ke, ON); and 2) samples of soils contaminated with diesel fuel (ON) in water.

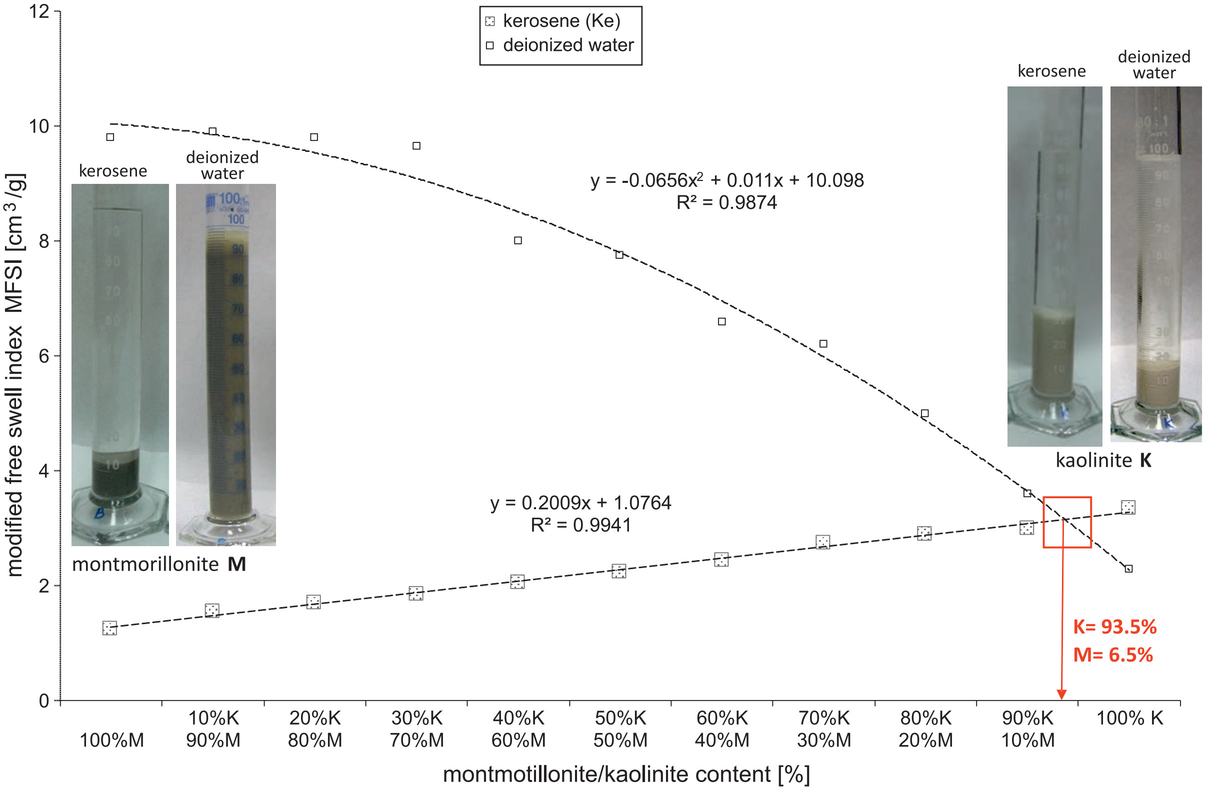
Analysis of free swelling in 10 samples of natural soils has indicated that the studied clays are characterized by 0.1 to 1.22 cm^3/g larger swelling in water than in kerosene (Text-fig. 10), with larger differences observed for the clay samples from Dobre (cDb) compared to those from Warsaw (cWP). Text-fig. 10 presents also the results of free swelling of monomineral clays – kaolinite from Sedlec (K) and bentonite from Wyoming (M). As our study indicates, MFSI values for clean montmorillonite show a similar trend as in natural soils, and depending on the chemistry of the pore fluid, vary in a wide range from 9.8 cm^3/g in deionized water to 1.25 cm^3/g in kerosene. Kaolinite shows an opposite trend, where the differences in swelling are smaller, but the mineral swells more intensely in non-polar organic fluids. An analogous relationship was observed in the study of free swelling in water and diesel fuel (ON). Montmorillonite swelled in water about 50 times more than in diesel fuel, whereas kaolinite showed an opposite trend (Text-fig. 10).

In the case of natural soils, i.e., Neogene clays and Pleistocene glacial tills from Mszczonów (cMsz and tMsz), free swelling of Neogene clays in water and diesel fuel is respectively 4–5 times larger than that of glacial tills (Text-fig. 10).

Observations of swelling of soil mixtures with a different percentage contribution of model minerals: Na-montmorillonite (M) and kaolinite (K) in deionized water and in kerosene were performed by Izdebska-Mucha and Wójcik (2016). The observed regularities indicate the behaviour of cohesive soils with different pore fluids, including contamination with non-polar organic fluids. Beside significant differences in the swelling time (24 h for kerosene and 5×24 h for deionized water), other functional relationships with a high correlation coefficient were noted for MFSI values and the percentage content of montmorillonite and kaolinite (Text-fig. 11). Soil swelling in aqueous solutions increases with rising montmorillonite content, whereas in kerosene the relation is opposite – higher swelling values were obtained for samples with the largest kaolinite content. The conclusion is that even a small content of montmorillonite at the level of 6.5% may signifi-

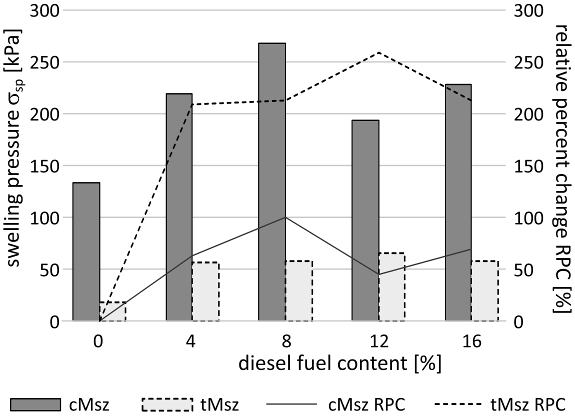


Text-fig. 10. Comparative swelling tests of uncontaminated monomineral clays and cohesive soils in water and oil-derived fluids. Abbreviations: M – montmorillonite, I – illite, K – kaolinite, 1–5 – cDb, 6–10 – cWP.

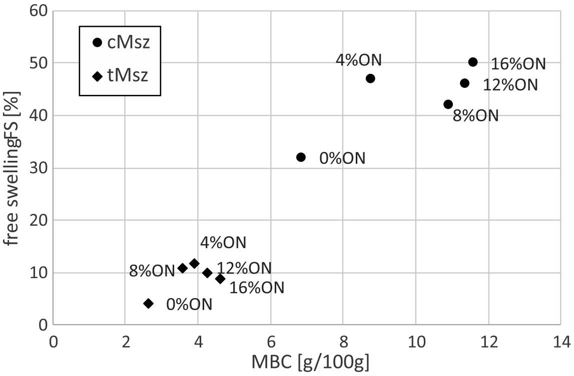


Text-fig. 11. Experimental swelling tests of monomineral clay mixtures in water and kerosene.

cantly influence the swelling in soil, and thus have impact on its expansiveness, whereas the content of kaolinite to about 30% does not have significant impact on processes of volume change. The character of this process is reflected in the photographs of cylinders with pure model minerals swelling in water and kerosene. The magnitude of kaolinite swelling in kerosene at higher values than montmorillonite finds its explanation in the mechanism of sorption of non-polar organic fluids by clay minerals, as evidenced by Sridharan *et al.* (1985) and Sridharan and Prakash (1999). When considering the magnitude of soil swelling the percentage content of the clay fraction should be taken into account.



Text-fig. 12. Effect of diesel fuel contamination on the swelling pressure (σ_{sp}) of Neogene clays (cMsz) and glacial tills (tMsz) from Mszczonów.



Text-fig. 13. Effect of diesel fuel contamination on free swelling (FS) and methylene blue capacity (MBC) of Neogene clays (cMsz) and glacial tills (tMsz) from Mszczonów.

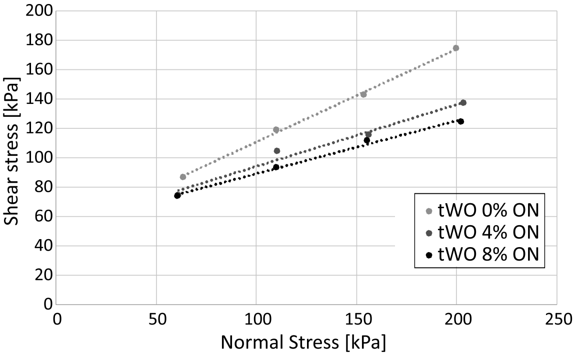
Tests of swelling properties for ON-contaminated Neogene clays (cMsz) and glacial tills (tMsz) from Mszczonów were performed along with the analysis of their sorption properties expressed by methylene blue capacity (MBC) and final moisture content after swelling. Analyses have shown that both for Neogene clays and glacial tills, the values of free swelling and swelling pressure are higher for samples contaminated with ON than for 0% ON samples (Text-figs 12 and 13; Table 4). The highest value increase can be observed at the lowest soil contamination of 4% ON. With contamination increase, the values of each parameter do not reveal a clear change trend and are attained at a relatively similar level in each soil type. Values of the percent difference related to clean samples (RPC) are much higher for glacial tills than for clays (Text-fig. 12 and Table 4), however the values of soil swelling parameters are very low and their variability range is quite narrow for samples 4–16% ON. Measurements of soil moisture content before and after free swelling tests (Table 4) show the content of water absorbed during the swelling process (Δw). These values are significantly higher in the contaminated samples of Neogene clay (84–82%) than in clean samples (72%), which is in accordance with FS values. For glacial tills the values of Δw clearly decrease at highest soil contamination 12–16% ON. Results of MBC measurements confirm the trends of changes in FS and σ_{sp} values. Text-fig. 13 shows that contamination increase results in rising sorption properties of the soil. It is worth noting that the largest increase of MBC values is again observed for the lowest contamination levels – 4% ON, and 4% and 8% ON for glacial tills and Neogene clays, respectively.

Reports on swelling properties of hydrocarbon-contaminated soils are very limited. According to

Soil	Diesel fuel content [%]	Free swelling FS [%]	Relative percent difference of FS RPC [%]	Initial moisture content w_0 [%]	Moisture content after FS w_f [%]	Adsorbed water Δw [%]
Neogene clays from Mszczonów (cMsz)	0	32	–	6.02	77.9	71.9
	4	47	47	4.11	88.5	84.4
	8	42	31	7.64	90.0	82.4
	12	46	44	9.39	90.6	81.2
	16	50	56	10.9	92.8	81.9
Glacial tills from Mszczonów (tMsz)	0	4.12	–	2.17	39.3	37.1
	4	11.8	186	4.01	40.9	36.9
	8	10.9	164	7.60	44.8	37.2
	12	9.9	140	10.65	44.3	33.7
	16	8.8	114	13.69	43.5	29.8

Table 4. Results of free swelling (FS) of Neogene clays (cMsz) and glacial tills (tMsz) from Mszczonów contaminated with diesel fuel (ON).

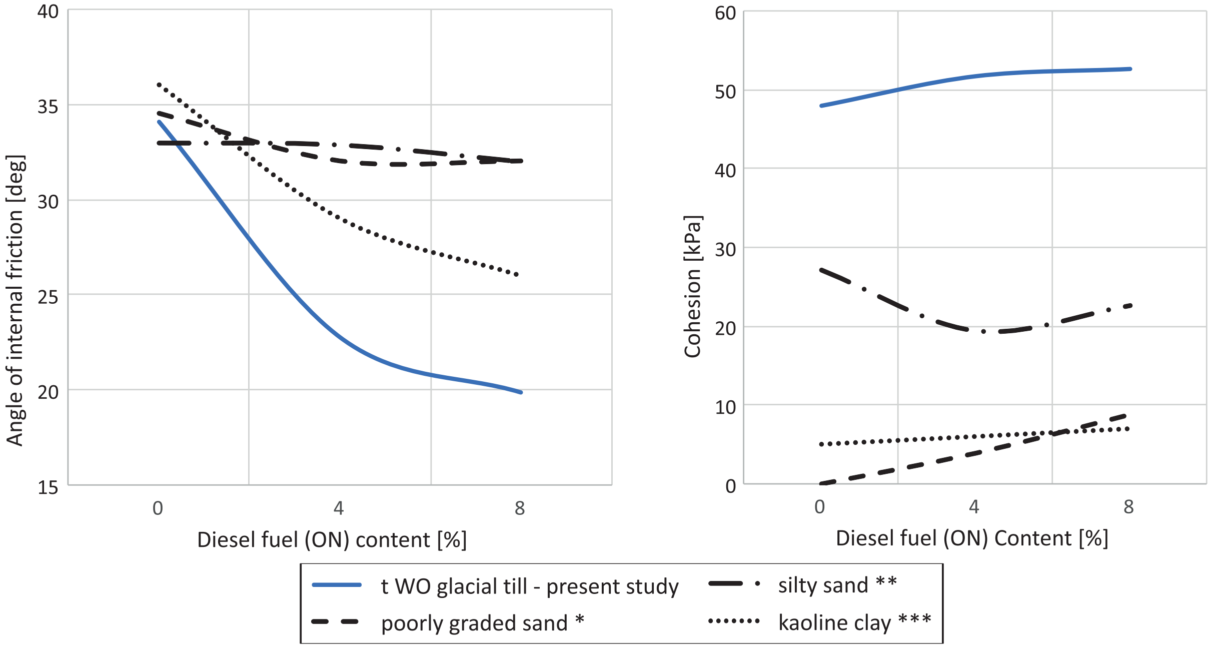
Mosleh and Al-Obaidy (2021), only about 10% of the studies on the behaviour of oil-contaminated soils focus on the behaviour of expansive soils. The reported data is ambiguous and does not deliver an insight to explain the observed changes in the swelling of contaminated soils. For example, Majeed and Majeed (2017) concluded that petroleum products reduce the free swelling index and swelling pressure of expansive soils, whereas Harsh *et al.* (2016) reported the increase of the free swelling index of hydrocarbon-contaminated clay soils. Salimnezhad *et al.* (2021) presented the increase of free swelling and decrease of swelling pressure for clayey soil contaminated with crude oil up to 12%. The reason for these differences is not clear. Further comparative investigations of undisturbed soils contaminated *in situ* may shed more light on this matter.



Text-fig. 14. Shear stress *vs.* normal stress for glacial tills from Warsaw Ochota Campus (tWO).

Shear strength

Results of studies on mechanical properties provide evidence that contamination with oil-derived substances has a significant impact on the soil shear strength (Text-fig. 14). The lubricant role of the substance facilitates sliding of the soil particles, which is reflected in the significant decrease of the angle of internal friction. In the analysed soils, contamination admixture at the level of 4% (first level) results in significant decrease of the angle of internal friction up to 30%. Rise of the contaminant content by another 4% results in decrease of the angle of internal friction by an additional 8%. An identical trend of changes is confirmed by the results obtained by other researchers. In studies focused on non-cohesive soils (sand and silty sand), the decrease of the angle of internal friction is less distinct and does not exceed 7% at increase of diesel fuel content to 8% (Khamehchiyan *et al.* 2007; Hafshejani and Hajiannia 2016). Studies performed on clays confirm the decrease of the angle of internal friction by several degrees with rising diesel fuel content (Khosravi *et al.* 2013). Changes of the angle of internal friction with rising contamination content (diesel fuel) are presented in Text-fig. 15. They show that increase of soil cohesion is reflected



Text-fig. 15. Friction angle and cohesion of soils contaminated with diesel fuel (ON); * – ON-contaminated poorly graded sand (Khamehchiyan *et al.* 2007), ** – ON-contaminated silty sand (Hafshejani and Hajiannia 2016), *** – ON-contaminated kaolinite clay (Khosravi *et al.* 2013).

in the higher susceptibility of the soil to contamination with oil-derived substances.

In turn, assessment of the influence of diesel fuel contamination on the changes of soil cohesion are not that explicit. In all analysed samples, cohesion determined in simple shear tests increased insignificantly or was at a similar level (for tWO 0%, c = 48 kPa; for tWO 4%, c = 52 kPa; for tWO 8%, c = 53 kPa). Such insignificant increase of cohesion for cohesive soils caused by ON-contamination is confirmed e.g., by the studies of Khosravi *et al.* (2013). Contrary to other researchers dealing with soils contaminated with hydrocarbons, these authors have observed increase of cohesion after introducing diesel fuel into the pore space of the kaolinite samples. The documented trend of changes of the strength parameters is apparently caused by the reduction of the values of repulsive forces between the particles of clay minerals in a non-polar fluid with a low value of the dielectric constant (Khosravi *et al.* 2013). In the case of studies performed on clays, cohesion increased by several kPa with increase of contaminant content not more than 8% (Khosravi *et al.* 2013). In studies on non-cohesive soils whose initial cohesion was zero or soils with a low apparent cohesion, addition of diesel fuel caused only slight increase of cohesion at the level of 8 kPa (Khamehchiyan *et al.* 2007). It is difficult to address the physical sense of such changes in cohesion of contaminated soils, especially as both our research and the literature data were based on reconstituted samples. Such samples should be characterised by minimal or no cohesion.

Permeability

The optimal conditions for assessing the quantitative effect of contamination on permeability can be obtained by testing soil pastes. Compared to stiff plastic soils with a natural structure, firm and soft plastic soil pastes were characterized by greater homogeneity and uniform conditions for the propagation of pollutants. The comparison between the physical parameters of the natural (undisturbed) soil samples and the model soil pastes are presented in Table 5.

The presence of two fluid phases (water and/or oil-derived substance) of diverse physical-chemical properties in the soils may lead to significant changes of their permeability. The process of seepage depends on the contaminant type and content in the fluid phase, as well as the geological engineering properties of the soil (particle size composition, porosity, microstructure, etc.).

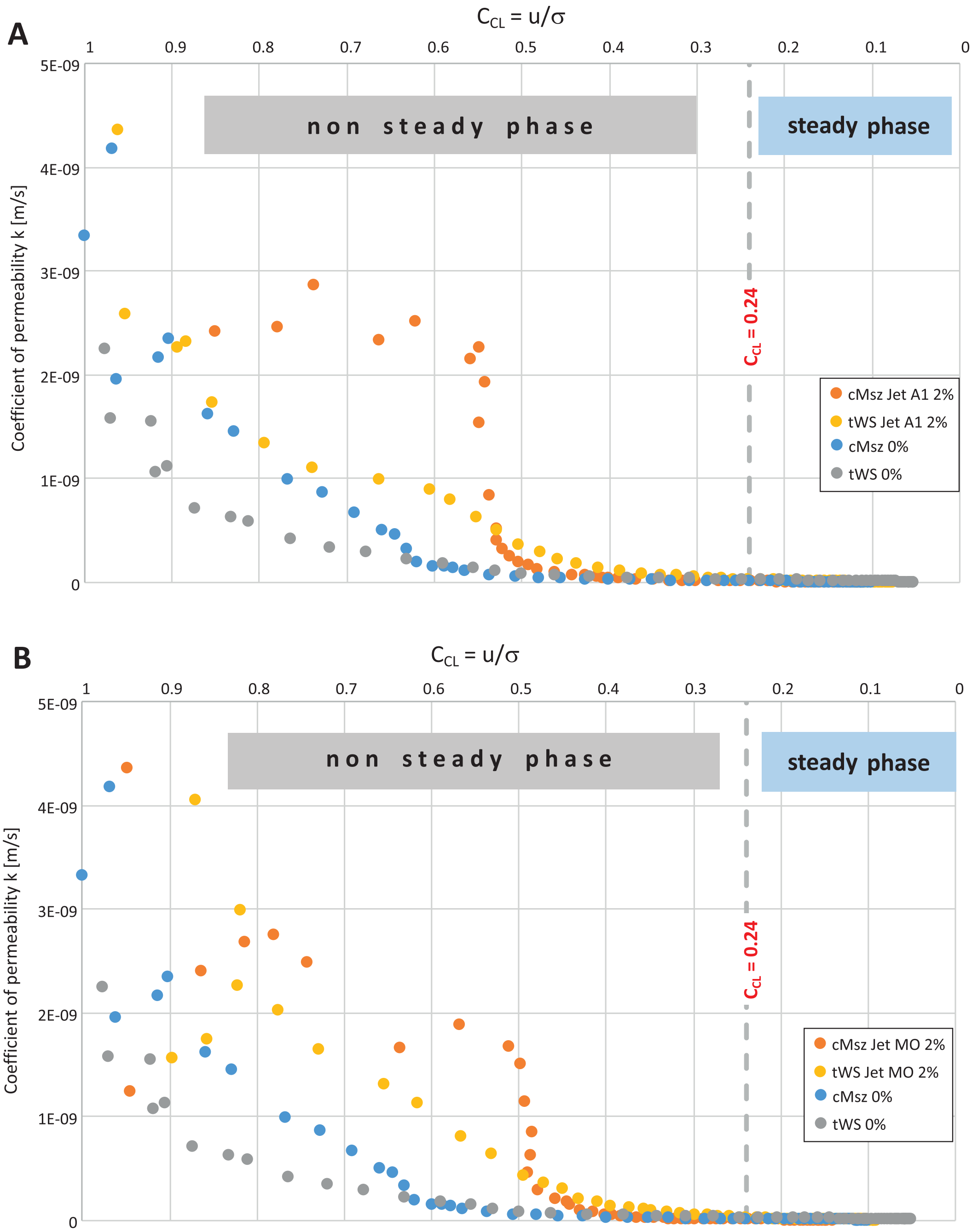
Directions of changes in the permeability in the soil after contamination with oil-derived products presented in the literature are ambiguous. Investigations show results pointing both to increase of permeability in soils contaminated with hydrocarbons as well as decrease of permeability in soils contaminated with oil-derived fuels. In cohesive soils these effects may be affected by porosity changes caused by flocculation and aggregation of the clay matrix. By not mixing with water, oil-derived products may, due to their viscosity, exclude part of the pores from the seepage process, thus decreasing the permeability of the contaminated soil.

In reference to the analysed soil pastes with a particle size distribution of the Neogene clays (cMsz) and Pleistocene glacial tills (tWS), the assessment of permeability changes is presented on the basis of parameters of pore water pressure and strain obtained during investigations of uniaxial consolidation at a constant rate of loading. Theoretical models in CRL studies (Dobak 1999, 2008) show that reliable values of the soil compressibility modulus M_0, as well as consolidation c_v and permeability coefficients k are obtained in the steady phase, when the dimensionless parameter of pore water pressure $C_{CL} = u/\sigma$ is below 0.24. Studies performed in uncontaminated soils and soils contaminated with oil-derived products generally confirm the character of theoretical relations C_{CL}-k, but enrich them with some detailed effects (Text-fig. 16).

In the non-steady phase the presence of contaminants results in variable anomalies of k values, with locally significant increments with regard to standard characteristics observed in non-contaminated material. The registered effects indicate that in soil under load, momentary local restriction of conductivity takes place in conditions of fluid phase flow with the contaminant. The most diverse anomalies were observed in contaminated pastes of Neogene clays

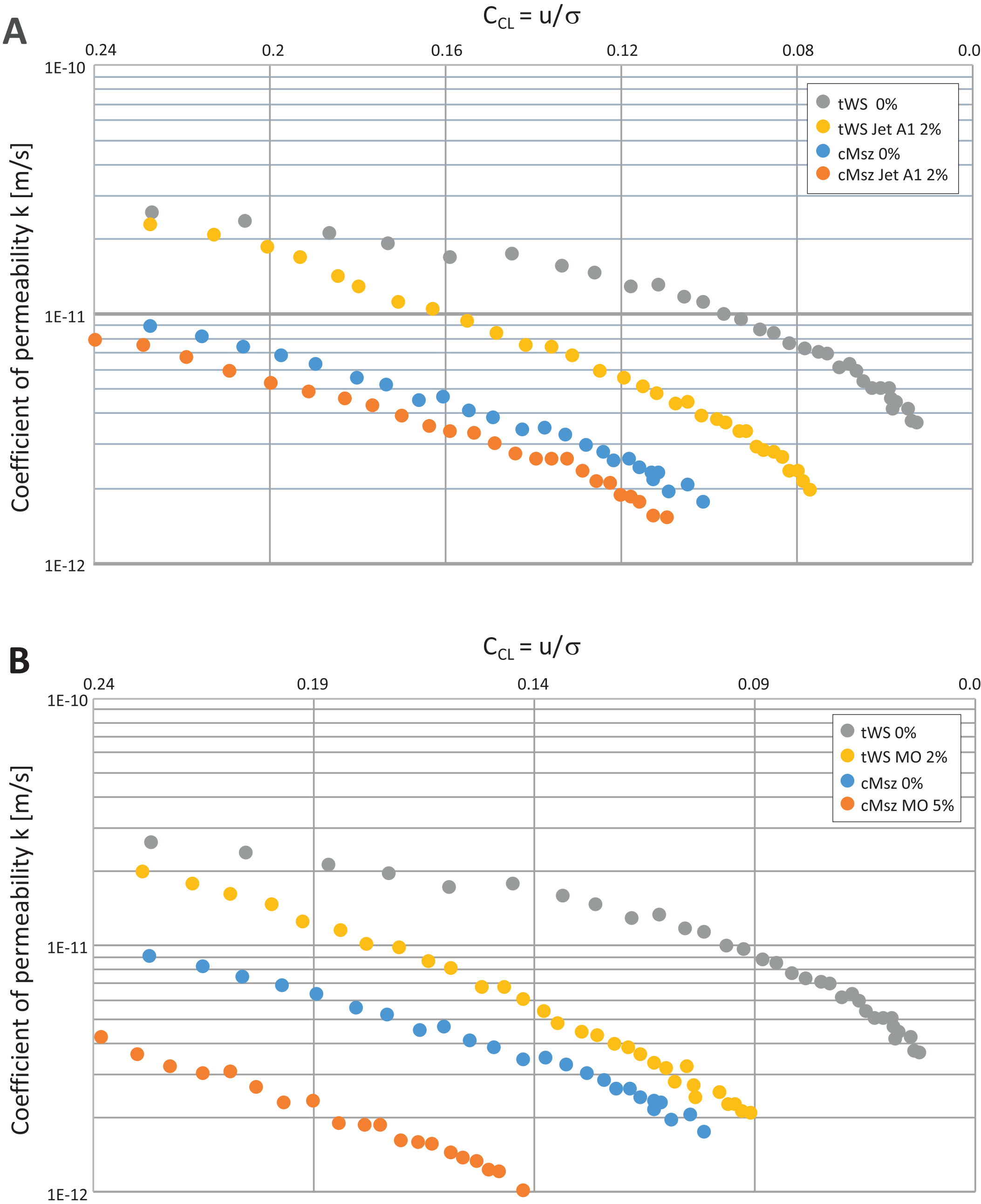
Soil type	Sample type	Water content w [%]	Particle density ρ_s [Mg/m^3]	Bulk density ρ [Mg/m^3]	Liquidity index I_L [-]	Void ratio e [-]
Neogene clay (cMsz)	undisturbed sample	26.8–34.1	2.69	1.92–2.06	0.01–0.11	0.64–0.89
	soil paste	54.3–55.4		1.68–1.69	0.52–0.54	1.46–1.49
Glacial till (tWS)	undisturbed sample	15.3–18.9	2.67	2.10–2.14	-0.06–0.11	0.43–0.50
	soil paste	38.4–39.7		1.83–1.86	0.74–0.85	0.96–1.04

Table 5. Physical properties of the tested soil pastes and undisturbed clay soils (natural conditions).



Text-fig. 16. Coefficient of permeability with regard to phases of the CRL analysis, soil material and contaminant type; A – contamination with jet fuel (Jet A1); B – contamination with mineral engine oil 15W40 (MO).

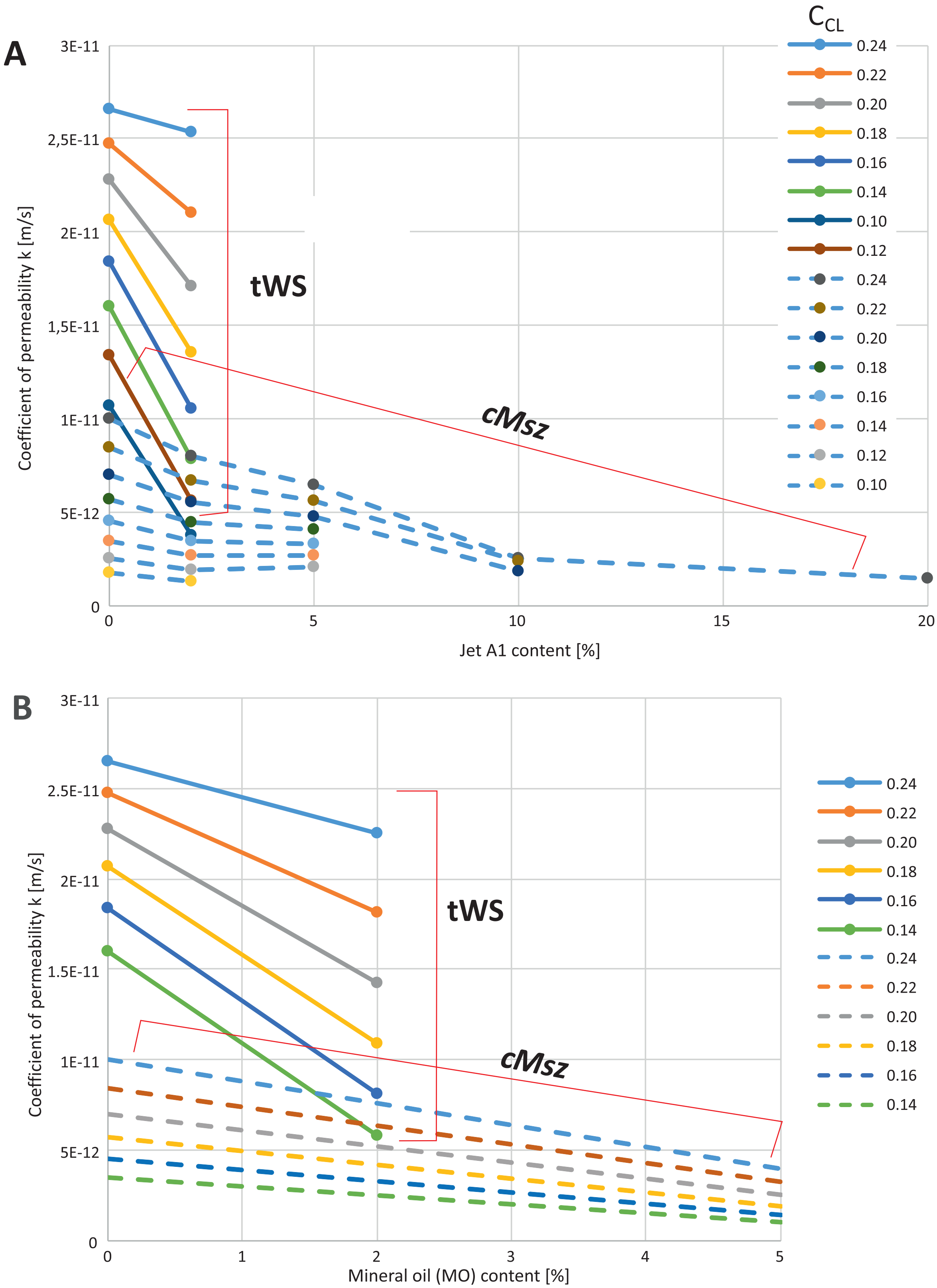
(cMsz), and smaller in contaminated pastes of glacial tills (tWS). The effects were more strongly marked in soil material contaminated with heavier, viscous fractions of mineral oil (Text-fig. 16A), although jet fuel in the pore space of the soil also causes significant anomalies in the non-steady phase (Text-fig. 16B). With proceeding consolidation CRL, when the contribution of pore water pressure in load transfer decreases (C_{CL} <0.4), the asymptotic approach of the C_{CL}– k charts and smaller changes of the coefficient of permeability values can be observed. Charts C_{CL} – log k have a small convex-upward curve, which points to a less intense decrease of permeability in the function of load increase and pore water pressure



Text-fig. 17. Changes of the coefficient of permeability values for pastes made of glacial tills and clays; A – contamination with jet fuel (Jet A1); B – contamination with mineral engine oil 15W40 (MO).

in comparison to rectilinear logarithmic relations. The obtained results indicate that the magnitude of permeability changes depends on the particle size distribution of the analysed soil pastes and the type of contaminant (Text-fig. 17).

Contamination with mineral oil causes significant decrease of permeability both in the poorly graded and coarser material from glacial tills and in the finer Neogene clays. In turn, the presence of light jet fuel to a smaller degree results in permeability changes in clays than in tills (Text-fig. 17A). The observed decrease of the coefficient of permeability k during consolidometer tests was compared in reference to selected values of the dimensionless pore water pressure parameter C_{CL} from 0.24 to 0.10 (with an interval of 0.02). In the Neogene clays con-



Text-fig. 18. Changes of permeability depending on the type of soil and contaminant at comparable C_{CL} values; A – contamination with jet fuel (Jet A1); B – contamination with mineral engine oil 15W40 (MO).

taminated with light jet fuel from 2 to 20%, decrease of the coefficient of permeability can be observed almost proportionally to fuel concentration. At 20% contamination, the reduction of permeability is much less intense (Text-fig. 18A).

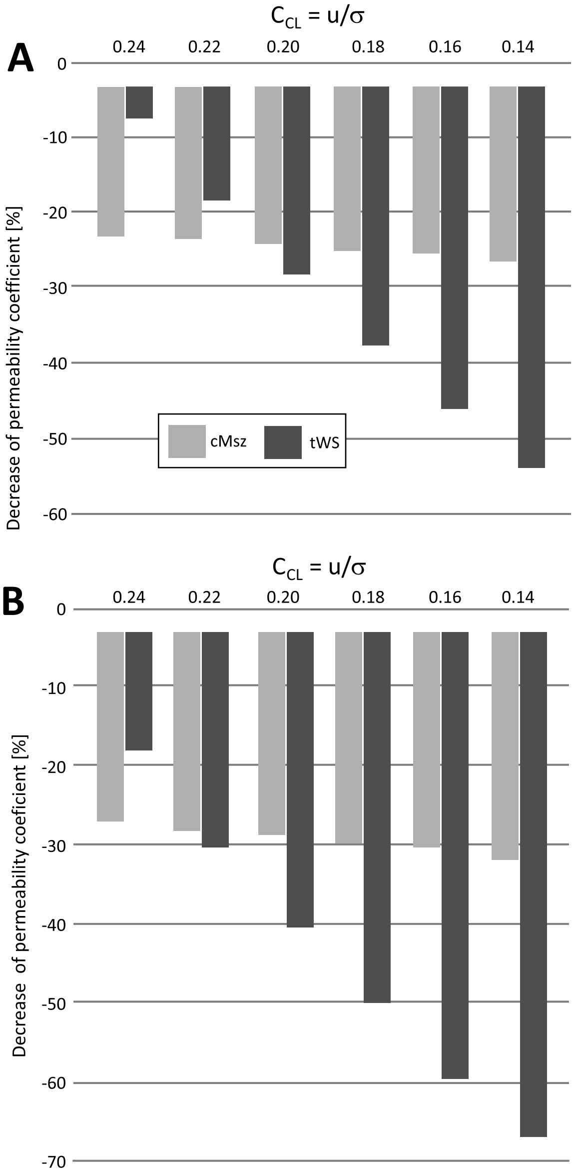
The characteristics presented in Text-fig. 18 indicate that the values of the coefficient of permeability for uncontaminated glacial tills were almost 3 times higher than those for Neogene clays. This relation was analysed for comparable C_{CL} values also in tills at 2% contamination with light jet fuel (Text-fig. 18A) and in clays at 5% contamination with mineral oil (Text-fig. 18B). Higher k values in pastes of glacial tills decrease more intensely than in Neogene clays. In this case, values k obtained at lowest C_{CL} values become similar to the characteristics of Neogene clays at the limit of the non-steady phase. These comparisons thus document sealing of the soil material due to contamination leading to unifying the characteristics of permeability for soils with a variable and coarser particle size distribution (contaminated pastes of glacial tills) and soils with finer particle size distribution characterising clays of lacustrine origin. The quantitative characteristics of permeability changes referred to 2% contamination are presented in the diagrams (Text-fig. 19).

In Neogene clay pastes the reduction of permeability caused by contamination with light fuel (Jet A1) does not show significant changes, lying within -20% to -25% at C_{CL} from 0.24 to 0.14 (Text-fig. 19A). In the case of contamination with mineral oil, the reduction is only slightly higher, reaching -30% (Text-fig. 19B). In turn, in the samples of glacial tills characterised by variable particle size distribution, the percentage reduction of permeability caused by contamination with Jet fuel A1 changes significantly from -5% to over 50% at C_{CL} from 0.24 to 0.14 (Text-fig. 19A). A similar but slightly higher reduction of permeability (from over 10 to over 60%) was obtained for samples of glacial tills contaminated with viscous and heavier particles of mineral engine oil.

In general, it can be concluded that the key role in shaping the coefficient of permeability is played by structural factors, and next by distinctiveness resulting from the physical properties of the contaminant (viscosity, density).

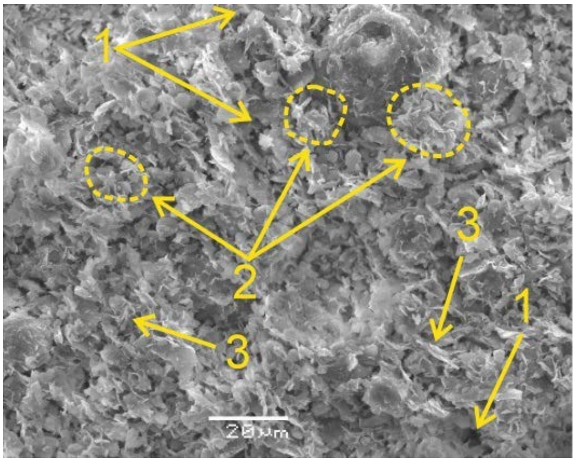
Microstructure

Reference to structural features requires assessment of microscopic images. Uncontaminated soil pastes prepared from Neogene clays and glacial tills have shown a matrix microstructure in SEM images. The chaotic distribution of silt and sand fraction grains in a homogenous loosely packed clay matrix, is characteristic for this microstructure. The contacts of clay particles in individual microaggregates may be described as face-to-face F-F (Text-fig. 20) and edge-to-face E-F (Text-fig. 21). Anisometric, rarely isometric pores without distinct orientation dominate between the aggregates.

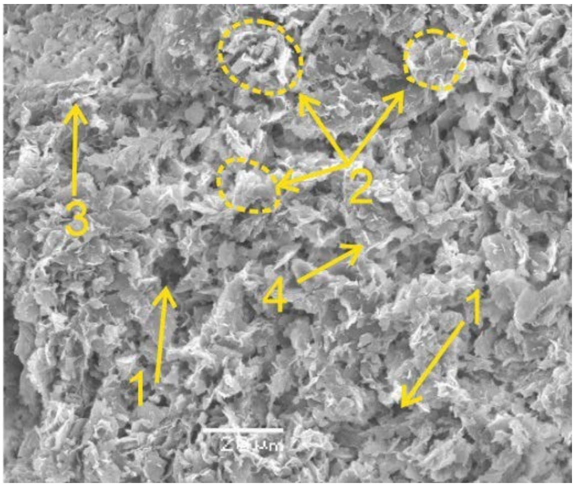


Text-fig. 19. Quantitative changes of the coefficient of permeability depending on the type of contaminant and soil material; A – contamination with jet fuel (Jet A1); B – contamination with mineral engine oil 15W40 (MO).

After contamination with jet fuel and mineral oil, the soil pastes made of Neogene clays and glacial tills



Text-fig. 20. Microstructure of uncontaminated soil paste made of Neogene clays from Budy Mszczonowskie (magnification ×800). Explanations: 1 – pores, 2 – microaggregates, 3 – F-F contact.



Text-fig. 21. Microstructure of uncontaminated soil paste made of glacial till of the Odranian Glaciation from the vicinity of Warsaw (magnification ×800). Explanations: 1 – pores, 2 – microaggregates, 3 – F-F contact, 4 – E-F contact.

have still shown a matrix structure without distinct orientation of the structural elements, with loose distribution of the clay mass, and F-F and E-F contacts of the clay mineral particles. However, aggregation of the clay mass and quantitative changes of parameters characterising the geometry of the pore space (Table 6) are visible in the microstructural images after contamination.

Mesopores still dominate in the microstructure of contaminated soil pastes. Increase of contamination results in increased contribution of anisometric pores and reduction of isometric pores.

After contamination of cohesive soils with oil-derived fuels, change in the pore number and dimensions are observed; for example, the Neogene clays show increased maximal values of the pore surface S_{max}, diameter D_{max} and perimeter P_{max}. In general, however, the parameters determined by STIMAN point to a significant structural variability, which may explain local flow anomalies in the soil medium registered in consolidometer tests during permeability determination.

CONCLUSIONS

1. The particle density decreased with the increasing content of hydrocarbons. These are apparent changes related to the presence of contaminant residues on soil particles and not to a change in the mineral composition. Knowledge on this apparent effect becomes an additional tool indicating the presence of petroleum substances when testing contaminated soils.

2. Similarly, the influence of oil-derived substances on particle size distribution results from the interactions between contaminated soil particles, and not the actual changes in the size of soil particles. Therefore, the main apparent effect of shifting the fraction content from finer to coarser ones is observed at an increasing contaminant content. It should be emphasised that the effect may diminish in the long term.

3. Mineral composition and clay content are key factors controlling soil swelling. Free swelling tests of kaolinite-montmorillonite mixtures in water and kerosene showed that hydrocarbons reverse the trends of soil swelling compared to behaviour in water. The increase of swelling pressure was observed for Neogene clays and Pleistocene glacial tills from Mszczonów contaminated with diesel fuel. Even the lowest applied content of 4% ON resulted in changes in swelling behaviour of the soil. Results of MBC measurements are in line with these observations, pointing to the increasing sorption potential of the contaminated soils.

4. A reduction of shear strength was observed in glacial tills contaminated with diesel fuel. Contamination at the level of 4% ON caused a 30% decrease of shear strength, which is a fundamental change of the soil bearing capacity. The soil cohesion increased slightly or was at a similar level with increasing contaminant content. As in the discussion of swelling and particle size distribution, this may suggest a complex role of the interactions between hydrocarbons and mineral particles, structural changes, aggregation of clay particles, and not only interface friction between soil particles and grains.

Sample		tWS				cMsz		
Type and concentration of the soil contaminant		JET A1 0%	JET A1 2%	JET A1 10%	MO 2%	JET A1 0%	JET A1 5%	JET A1 20%
Porosity [%]		17.4	19.1	22.8	19.7	23.3	32.1	21.8
Number of pores		31162	39254	25922	47074	32642	39027	76674
Pore diameter	Maximal value D_{max} [μm]	120,1	105.2	204.4	134.5	104.8	147.5	170.3
	Minimal value D_{min} [μm]	0.313	0.302	0.302	0.340	0.306	0.306	0.298
	Average value D_{av} [μm]	2.2	1.9	2.1	1.8	2.5	2.7	1.5
Total pore surface $S_t \times 10^3$ [μm^2]		402	447	542	458	543	739	499
Pore surface	Maximal value S_{max} [μm^2]	11331	8685	32817	14219	8630	17098	22775
	Minimal value S_{min} [μm^2]	0.077	0.072	0.072	0.091	0.073	0.073	0.070
	Average value S_{av} [μm^2]	12.9	11.4	20.9	9.7	16.6	18.9	6.5
Total pore perimeter $P_t \times 10^3$ [μm]		570.7	549.4	435.5	629.8	707.8	742.3	950.2
Pore perimeter	Maximal value P_{max} [μm]	3214.7	3661.9	6370.3	2205.0	4750.3	5951.3	6750.0
	Minimal value P_{min} [μm]	3.5	1.5	2.2	2.0	2.9	1.8	2.3
	Average value P_{av} [μm]	18.3	14.0	16.8	13.4	21.7	19.0	12.4
Form index of pores	Maximal value Kf_{max} [-]	0.95	0.95	0.90	0.94	0.93	0.98	0.97
	Minimal value Kf_{min} [-]	0.11	0.05	0.09	0.08	0.10	0.02	0.04
	Average value Kf_{av} [-]	0.56	0.50	0.47	0.53	0.52	0.49	0.54
Mesopores $10 < d < 10\,000$ μm [%]		58	64	80	61	64	68	50
Micropores $0.1 < d < 10$ μm [%]		42	36	20	39	36	32	50
Fissure pores $a/b > 10$ [%]		0.00	0.09	0.03	0.06	0.00	0.03	0.02
Anisometric pores $1.5 < a/b < 10$ [%]		68.6	80.5	83.3	76.4	75.2	83.5	73.1
Isometric pores $a/b < 1.5$ [%]		31.4	19.5	16.7	23.6	24.8	16.4	26.8

Table 6. Quantitative microstructural parameters for the tested soils.

5. Permeability of the analysed soils points to changes depending on the particle size distribution as well as contaminant viscosity and density. Obtaining reliable characteristics of the coefficient of permeability is possible only in the steady phase of the consolidometer analysis (our recommended tool for indirect determination of k), when the dimensionless parameter of pore water pressure $C_{CL} < 0.24$.

6. Changes of the coefficient of permeability may be reliably compared at the same values of parameter C_{CL}. The decreasing values of coefficient k, reflected in the quasi-logarithmic relation C_{CL} – log k, result from fluid phase flow aggravation caused by decreasing compressibility of the successively loaded soil. The trends of permeability changes of soils contaminated with liquid hydrocarbons, as well as anomalies of the experimental characteristics depend *inter alia* on the resistance of filtration at random distribution of particles in the clay matrix and shifting of contaminant particles in the mesopore space.

7. Contaminated soil pastes of Neogene clays and glacial tills showed a matrix microstructure with increased contribution of anisometric pores. Neogene clays showed increased maximal values of the pore surface Smax, diameter Dmax and perimeter Pmax. The analysis revealed a significant structural variability of the tested soils, which may explain local flow anomalies during permeability determination.

8. The study has revealed that in laboratory testing contaminated soils show different behaviour and characteristics compared to typical soils. This is related to the different physical properties of the contaminants, as well as change in interparticle friction and soil cohesion; and flow conditions of the fluid phase. In investigation of contaminated soils, comparison of uncontaminated and variably contaminated soils with analogous structures are essential. These data are also crucial in the quantitative assessment of anthropogenic pressure on the geological engineering environment.

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