

Review

The geological significance of novel anthropogenic materials: Deposits of industrial waste and by-products



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ABSTRACT

Industrial wastes and by-products are increasingly (re-)used as filling material in constructions. To enhance awareness among the geological community of the growing and widespread occurrence of deposits that contain these “novel anthropogenic materials”, this paper reviews three volumetrically important materials: (1) ash remaining from the incineration of household waste (Municipal Solid Waste Incineration bottom ash), (2) slag from steel production, and (3) the stony aggregate fraction of construction and demolition waste. We review their origin, main geochemical weathering reactions, and influence on the natural geogenic environment. These materials have properties that set them apart from the geogenic materials that they replace and overlie. They are formed under high-temperature conditions, are thermodynamically unstable, and may exhibit physical and chemical changes on relatively short timescales, an aspect relatively new for mapping geologists. Because knowledge of deposits in the shallow urban subsurface is increasingly important for urban planning, more articulation and detail in the classification of novel anthropogenic materials is necessary.

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1. Introduction

Humans have become a geological factor (Price et al., 2011). Direct or indirect human interventions affect processes that shape Earth's surface on a large scale, e.g., sedimentation, erosion and soil formation (Zalasiewicz et al., 2011). In the Netherlands, which includes some of the World's most artificialized terrains,

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excavation, dredging, quarrying, filling, rainbowing, coastal nourishing, embanking, trenching, shipping and trucking have become the dominant depositional and erosional processes (Van der Meulen et al., 2007a,b). About 4800 km² (14 %) of the Dutch land surface is now built up and most built-up areas have an anthropogenic deposit (“made ground”) up to several meters in thickness (CBS, 2018, Fig. 1). Regionally sourced sand comprises the bulk of these deposits (Van der Meulen et al., 2007a). The properties of these deposits generally match those of the local superficial deposits.

A growing share of the raw materials used as filling material in constructions is “secondary”, and consists of recycled stony or earthy waste materials and industrial by-products. Examples include slags from the steel industry (coastal protection, roads, foundations for parking lots), ashes from municipal solid waste incineration (road construction, noise barriers) and construction and demolition waste (foundations, road construction). These materials are increasingly becoming part of the shallow urban subsurface, but they have properties that set them apart from the primary geological raw materials they replace and overlie. Peat set aside, Dutch superficial Quaternary deposits are mineralogically mature and geochemically stable. In contrast, many secondary materials form under high-temperature conditions and may exhibit physical and chemical changes on relatively short time-scales of years to decades. These characteristics may be relatively

new for mapping geologists. Hence, this paper refers to these materials hereafter as “novel anthropogenic materials”.

Around the turn of this century, systematic subsurface modelling replaced traditional geological mapping at the Geological Survey of the Netherlands. Four of their reference models cover three depth ranges (for detail, see Van der Meulen et al., 2013). The upper tens of meters are modelled as a voxel grid (GeoTOP) with stratigraphic and lithological attributes (Stafleu et al., 2011). Past geological mapping has traditionally avoided urbanized areas (Van der Meulen et al., 2013). In particular, anthropogenic deposits are perceived as unpredictably heterogeneous because of the occurrence of novel anthropogenic materials, among other reasons (Van der Meulen et al., 2013). Accordingly, anthropogenic deposits are presently still treated as a single lithological class in GeoTOP (Stafleu et al., 2011).

A growing awareness exists of the need to better resolve the geometry and properties of deposits in the shallow urban subsurface. Improved understanding would help to avoid subsurface-related risks as well as to make use of its opportunities and functions (Schokker et al., 2015; Van der Meulen et al., 2016; Mielby et al., 2017; Koster et al., 2018; Le Guern et al., 2018). Meeting this challenge requires more articulation and detail in classifying anthropogenic deposits, in particular how novel anthropogenic materials behave geologically and geochemically over longer timescales.

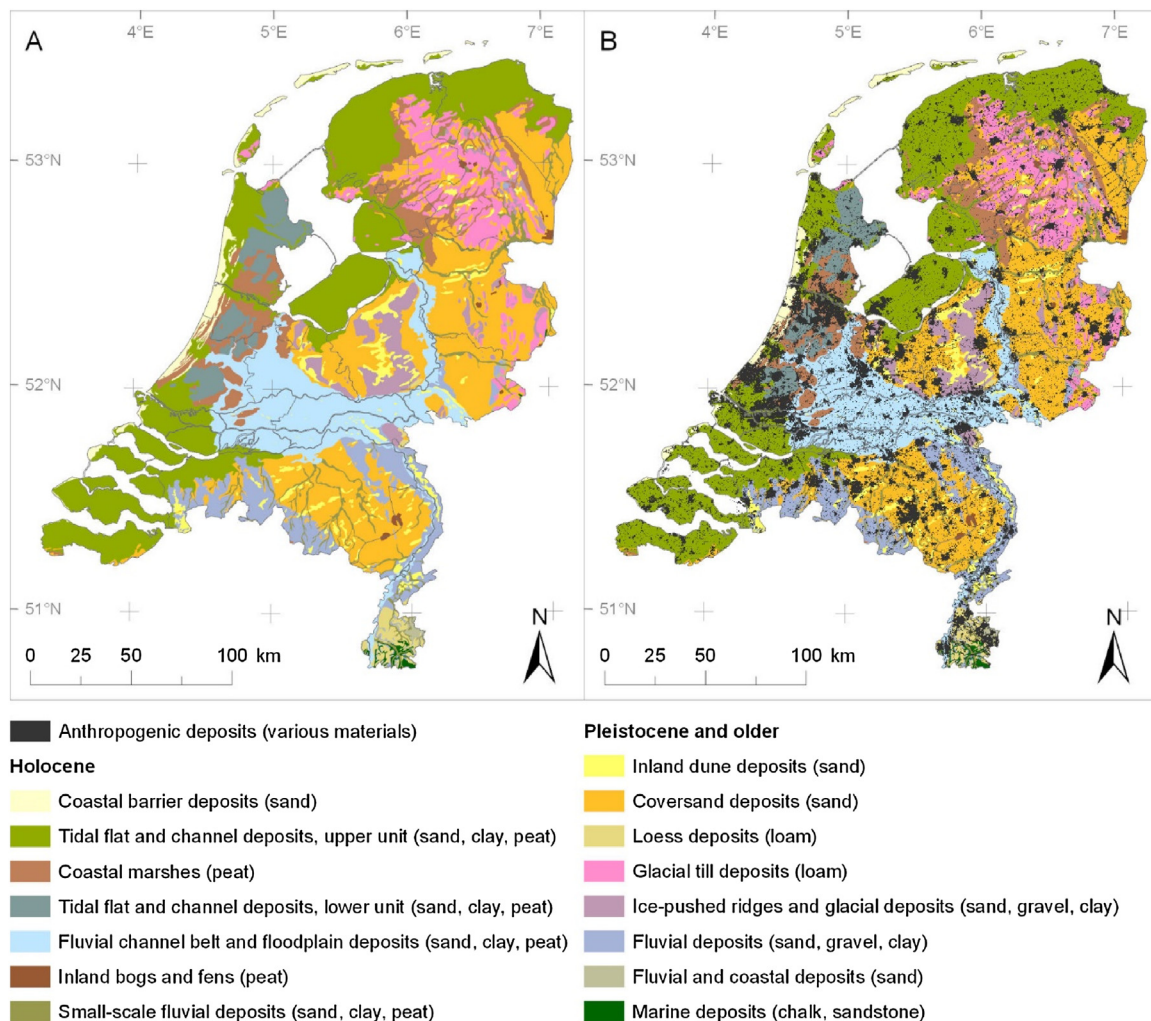


Fig. 1. Surficial natural sediments and rocks in The Netherlands (A) are becoming increasingly overlain by anthropogenic deposits (B) that encompass both reworked natural deposits and novel anthropogenic materials of various nature.

To this end, this paper reviews three volumetrically important and geochemically relevant novel anthropogenic materials: (1) the major ash fraction remaining from the incineration of household waste, Municipal Solid Waste Incineration (MSWI) bottom ash, (2) slag from steel production, also known as Linz-Donawitz (LD-) steel slag, and (3) the stony aggregate that forms the major fraction of construction and demolition (C&D) waste. In the context of their widespread use in The Netherlands as filling material in constructions, this paper focuses on their origin, main geochemical weathering reactions, and influence on the natural geogenic environment.

2. Origin and geochemical properties of three major types of novel anthropogenic materials

2.1. Use of novel anthropogenic materials in constructions in the Netherlands

The use of wastes and by-products in construction is becoming common practice in many European countries and elsewhere (e.g., USA, Japan). However, the Netherlands might be unique with respect to its very high recycling rates of wastes and by-products in unbound surficial deposits. Aiming to stimulate recycling, the Dutch national government promoted the use of secondary materials in construction since the early 1990's with a combination of regulatory and financial instruments (taxation/subsidies), and by using secondary materials in national infrastructure development. Legislation and standards were developed, putting a strong emphasis on constructional and environmental safety (e.g., [Netherlands Ministry of Housing Spatial Planning and the Environment, 2007](#), see Supporting Information for detail). All construction materials, including re-used wastes and by-products, need to pass strict leaching limit values before application, and different protective modes of applications are distinguished (open to the atmosphere or closed and sealed; see Supporting Information).

Virtually the entire production of bottom ash (1.2 Mt/a), steel slag (0.7 Mt/a) and stony aggregate from construction and demolition waste (18 Mt/a) is currently applied as unbound filling material in constructions. Together these three categories of waste material add to ~ 20 Mt/a ($\sim 13.3 \times 10^6 \text{ m}^3/\text{a}$), or about half the lower estimate of the average net Holocene sediment deposition ($25\text{--}50 \times 10^6 \text{ m}^3/\text{a}$) ([Van der Meulen et al., 2007b](#)) and currently replace a considerable share of filling sand (33.1 Mt/a or $22 \times 10^6 \text{ m}^3/\text{a}$) ([CLO, 2018](#), average 2010–2016, excluding non-regular use for land reclamation and coastal defense projects). With ambitious circular-economy targets in Europe ([European Commission, 2019](#)) and in The Netherlands ([Dutch Ministry of Infrastructure and Environment, 2017](#)), the use of recycled wastes and industrial (by)products in the shallow subsurface is expected to show a further growth.

2.2. MSWI bottom ash

In the Netherlands and other northern European countries, incineration is by far the dominant treatment for municipal solid waste that is not recycled or composted (Confederation of European Waste-to-Energy Plants ([CEWEP, 2019](#))). Modern incinerators are equipped with effective flue gas cleaning systems able to comply with stringent air emission criteria ([Astrup et al., 2016](#)). The bulk of the slag residue that remains after the incineration of municipal solid waste is generally referred to as “bottom ash”. After full processing (e.g., metal recovery, washing) the material comprises a greyish coloured, fine grained, glassy material ([Fig. 2](#)).



Fig. 2. Upper: MSWI bottom ash, applied as filling material under the provincial road N242 near Alkmaar, The Netherlands (picture: JJD); Middle: LD-steel slag waiting to find an application in construction (picture JJD); Lower: Typical application of Recycled concrete aggregate (RCA), here as sub-base of the national road E6 in Norway (picture kindly provided by C. J. Engelsens, SINTEF).

Incineration reduces the volume and the weight of the parent material by about 90 and 60 %, respectively ([Chandler et al., 1997](#)). The temperature in modern MSW Incinerators ranges between $700\text{--}1100^\circ\text{C}$, depending on the incineration technology and caloric value of the waste ([Chandler et al., 1997](#)). The residence time of waste in the combustion chamber is 45–90 min ([Meima, 1997](#)). The heavier ash residue that is collected from the combustion chamber is called bottom ash. Other waste streams are electrostatic precipitator fly ash (ash particles trapped in the flue gas stream), and air pollution control residues (residues after chemical treatment of the flue gas). In most incinerators, the hot bottom ash is “quenched” immediately after incineration in a water tank in order to cool the material, to prevent tertiary air entering the combustion chamber ([Astrup et al., 2016](#)) and to prevent dust generation.

In the Netherlands, bottom ash is further processed mainly to (1) recover valuable metals and (2) to lower the contaminant leaching potential to below regulatory limit values. Techniques for the separation of ferro- and non-ferro metals from the MSWI

bottom ash include e.g., sieving, magnetic separation, eddy current separation, optical separation, magnetic density separation and X-ray classification (Rem et al., 2004; e.g., Berkhout et al., 2011). Currently, the recovery mostly focuses on iron, aluminium, copper and zinc. Recovery is applied both on dry and on wet MSWI bottom ash. Wet techniques are in the Netherlands often part of the processing sequence in order to wash out soluble salts. Additional treatments to lower contaminant leaching include accelerated ageing, during which the moist and alkaline bottom ash is allowed to react with enhanced levels of carbon dioxide, leading to a chemically more stable product (see also below). For an overview of treatment techniques, see Astrup et al. (2016).

Table 1 shows the bulk chemical composition of typical MSWI bottom ash as used in Dutch infrastructure, based on the average of three well-studied samples (Meima and Comans, 1997; Dijkstra et al., 2006b, 2008). Lithogenic elements (Si, Ca, Fe and Al) dominate its composition, but the material also contains substantial amounts of soluble salts, mainly chlorides and sulphates (S in bottom ash is mainly present in the oxidized form, i.e. sulphate). Internationally, the variation in bulk chemical composition of MSWI bottom ash is surprisingly limited (for literature ranges see Table S1 and Figure S1 in the Supporting Information), and generally falls within an order of magnitude despite the obvious variation in waste composition, combustion temperatures and residence times (Kirby and Rimstidt, 1993; Astrup et al., 2016). The relatively high concentrations of trace metals, rare earth elements, platinum group elements and other precious metals are another interesting feature of MSWI bottom ash (Table 1 and Fig. 4). However, as concentrations are still much lower than in ores, there is not yet a business case for their recovery (Allegrini et al., 2014).

Freshly quenched MSWI Bottom ash is a thermodynamically unstable, highly reactive material because it consists predominantly (>70 wt%) of X-ray amorphous, glassy constituents (Kirby and Rimstidt, 1993; Zevenbergen et al., 1994b; Meima, 1997; Dykstra Eusden Jr et al., 1999) and because it has a high surface area and internal porosity. The quenching procedure causes a rapid cooling down of the partly molten material, and therefore “arrests” high thermal reactions before completion, leading to the presence of a series of intermediate reaction products in the bottom ash

(Zevenbergen, 1994). Roughly the following petrographical classification can be made of bottom ash particles (Meima, 1997; Dykstra Eusden Jr et al., 1999; Astrup et al., 2016):

- 1 Non-combustible material (15–45 %): waste glass, soil minerals (pyroxenes, quartz, feldspars), metals, metal alloys;
- 2 Newly formed material: Melt products (55–85 %): glasses (isotropic glass with schlieren, opaque glass), crystalline complex silicate phases (e.g., melilite group phases (gehlenite, akermanite) rich in Ca and Fe and depleted in Al, scapolite-like phases rich in Ca and Na and depleted in Fe) and crystalline complex oxides (e.g., lime (CaO), portlandite (Ca(OH)₂), iron oxides (e.g., hematite, magnetite), spinel-group phases), sulphates (e.g., gypsum (CaSO₄·2H₂O), anhydrite (CaSO₄), ettringite (Ca₆Al₂(SO₄)₃(OH)₁₂·26H₂O), carbonates (calcite (CaCO₃) and vaterite, a polymorph), hydroxides (e.g., goethite (FeOOH), gibbsite (Al(OH)₃).

The sequence of reactions taking place during combustion compares to a melt of melilite-bearing igneous rock (Dykstra Eusden Jr et al., 1999). Equilibrium of the reactions is not obtained, because of the interruption by the quenching process, and because of inequality of the temperature in the combustion chamber (“hot spots” and “cold spots”, Dykstra Eusden et al. (1999)). Truly cold spots exist in the combustion chamber, reflected by the sometimes recognizable fragments of combustible materials such as paper, plastic and textile in the ash. Another consequence of cold spots are small amounts (1–5 wt%) of unburnt natural organic matter in the ash, acting as a complexing agent causing enhanced metal leaching (Meima et al., 1999; Van Zomeren and Comans, 2004, 2009; Van Zomeren et al., 2009).

For a literature overview of identified mineral phases see Astrup et al. (2016). Due to the presence of alkaline phases such as portlandite and ettringite, fresh MSWI bottom ash in contact with water imposes a pH value of 11–12 (Meima and Comans, 1997; Dijkstra et al., 2006b).

2.3. LD-steel slag

Steel slag is a by-product of steelmaking. Modern converter steel plants, among which the single steel plant in The Netherlands, produce about 90–100 kg of steel slag per tonne of steel (World Steel, 2010, cited in Van Zomeren et al., 2011). Different slags are produced in different steps of the steel production process. In a blast furnace, iron ore (iron oxides such as hematite, magnetite) is transformed to a liquid metallic phase (crude molten iron), a liquid oxidic phase (blast furnace slag, BFS) and blast furnace gas. In The Netherlands, the BFS slags are mainly used as a supplemental cementitious material in concrete. In a next step, the liquid crude iron is converted to steel (and steel slag) by addition of limestone and/or dolomite, scrap iron and oxygen. This process is carried out in a basic oxygen furnace (also referred to as converter), and is known as the Linz-Donawitz process (abbreviated with “LD”). During this process, contaminants in the crude iron, such as Si, P, V and Mn, are bound in the oxidic (slag) phase. Scrap iron and cold slag are added as cooling agents to maintain a temperature of 1600–1640 °C (Yildirim and Prezzi, 2011). The process is controlled based on the quality of the steel and slag composition, mainly the ratio SiO₂/CaO. The molten slag is poured in slag beds, and is cooled down in a controlled manner using air or water. After cooling, a stony material remains that resembles basalt (Van Zomeren et al., 2011). In general, the more rapid the cooling, the less time there is for crystallization, and the more glassy-amorphous the structure of the slag (Tossavainen et al., 2007).

Table 1 shows the typical composition of Dutch LD-steel slag, derived from the average of two representative samples (Van

Table 1

Typical elemental composition of Municipal Solid Waste Incineration (MSWI) bottom ash, Linz-Donawitz (LD) steel slag and stony aggregate from Construction and Demolition (C&D) waste as used as filling material in Dutch infrastructure (for literature sources see main text, for composition ranges in literature see Supporting Information). * = only available for sample of Dijkstra et al. (2006b, 2008); n.a. = not available.

	MSWI Bottom ash	LD-steel slag	Stony aggregate from C&D waste
	g/kg	g/kg	g/kg
Si	216	63	267
Ca	87	307	76
Fe	92	183	22
Al	40	10	52
Na	21	< 1	15
Mg	13	45	7
K	9	< 1	20
S	8	< 1	4
Zn	4	< 1	< 1
Cl	4	< 1	< 1
Ti	6*	7	n.a.
P	3	6	< 1
Cu	3	< 1	< 1
Pb	3	< 1	< 1
V	<1*	6	< 1
Cr	<1*	1	< 1
Mn	1	35	< 1

Zomerén et al., 2011). LD-steel slag contains a considerable amount of residual iron (Table 1; literature ranges see Supporting Information). The high Ca and Mg contents result from the addition of limestone and dolomite in the process. Other major constituents of the slag are Si, Mn, Al, and Ti, followed by minor amounts of Cr, V and P.

The mineralogical composition of Dutch steel slag is dominated by magnesio wuestite ((Fe, Mg, Mn)O, 23–37 %), dicalcium silicate ($2\text{CaO} \cdot (\text{Si}, \text{P}, \text{V})\text{O}_4$, 36–49 %), dicalcium ferrite ($2\text{CaO} \cdot (\text{Fe}, \text{Ti}, \text{Al}, \text{V})_2\text{O}_3$, 13–26 %), free lime (CaO, 0–6 %), portlandite ($\text{Ca}(\text{OH})_2$, 0–3 %) and magnetite ($\text{FeO} \cdot \text{Fe}_2\text{O}_3$, 0–0.7 %) (Van Zomerén et al., 2011). This composition is similar to LD-steel slag produced worldwide (Shi, 2004; Yildirim and Prezzi, 2011; Piatak et al., 2015). Due to the presence of free lime, LD-steel slag exerts initially a high pH of 12–13 to its surroundings when in contact with water (Huijgen and Comans, 2006).

2.4. Recycled concrete aggregate (RCA)

The stony fraction of construction and demolition (C&D) waste that is re-used as unbound filling material is predominantly composed of debris originating from concrete and masonry (Hendriks, 1999). Although this material may also contain remnants of bricks, tiles, asphalt etcetera, this material generally has a strong “concrete”-fingerprint (Butera et al., 2014). In our discussion we focus on concrete debris being the major fraction, commonly referred to as Recycled Concrete Aggregate (RCA). Hence, the genesis and composition of this material is mainly determined by the original material, i.e., concrete.

Concrete is made using the basic ingredients of natural aggregate (sand, gravel), water, and a cementitious binder such as Portland cement. Portland cement is made by heating a mixture of natural limestone and clay, or other materials of similar bulk composition and sufficient reactivity, in a cement kiln ultimately to a temperature of 1450 °C (Taylor, 1997). The resulting material, “clinker”, is finely ground and mixed with a few percent of calcium sulfate. During the heating (“calcination”) calcium carbonates transform to lime (mainly as an intermediate product), and calcium silicates form such as alite ($\text{Ca}_3\text{O} \cdot \text{SiO}_4$), and belite (Ca_2SiO_4). Other important phases in Portland cement include aluminate ($\text{Ca}_3\text{Al}_2\text{O}_6$) and ferrite ($\text{Ca}_4(\text{Al}_x\text{Fe}_{1-x})_4\text{O}_{10}$) (Taylor, 1997).

When cement is mixed with water in certain proportions (these proportions are referred to as “cement paste”), hydration reactions result in hardening. These reactions are extremely complex (Taylor, 1997; Hewlett, 2010). Main reaction products from alite and belite hydration are portlandite, and calcium silicate hydrate, denoted as CSH in cement chemistry (Taylor, 1997). CSH is a generic name for amorphous or poorly crystalline calcium silicate hydrate, more highly ordered CSH phases include e.g., tobermorite and jennite. Another well-known cement mineral that forms upon cement hydration is ettringite.

Concrete consists predominantly (~80 wt%) of fine and coarse natural aggregate (Hewlett, 2010). The main mineral phases detected in RCA using X-ray diffraction are calcite and quartz. The abundance of these is so high that it masks other phases (Pacheco-Torgal et al., 2013). Phases detected at lower abundances include the reaction products of cement hydration, i.e. ettringite and portlandite, but also dolomite, plagioclase, feldspar, muscovite and gypsum (Pacheco-Torgal et al., 2013). The latter minerals, together with quartz, may originate from natural aggregates added to the concrete mix. Mineralogical analysis of C&D waste by Bianchini et al. (2005) has also revealed these minerals, but also minor amounts of Ca-Al-Fe hydroxides and hydrous silicates (typical for cementitious materials) and gehlenite and wollastonite (typical for bricks and terracotta). Clear patterns of CSH phases are absent in these studies, probably

because these phases are amorphous (Taylor, 1997) and hence are hard to detect using X-ray diffraction.

In the absence of data on major element composition of Dutch C&D waste, Table 1 shows the average bulk chemical composition of Danish and Norwegian C&D waste (predominantly consisting of concrete) as reported by Butera (2014) and Engelsen (2009) (composition ranges see Supporting Information). Si dominates the bulk chemistry, followed by Al and Fe, strongly matching that of natural aggregates of which concrete is predominantly composed. Many concretes are based on composite cements, which consists of Portland cement to which one or more inorganic materials are added that take part in the hydration reactions. These materials are commonly referred to as supplementary cementing materials, of which the most important examples are coal fly ash (byproduct from power plants), ground BFS (byproduct from steel production, see above), silica fume (byproduct from silicon production) and natural “pozzolanas” which are traditionally of volcanic origin (pumices, ashes and tuffs), but also sedimentary (e.g., diatomaceous earth) (Taylor, 1997; Snellings et al., 2012).

Due to the presence of alkaline phases, e.g., portlandite and CSH, fresh concrete initially exerts an alkaline pH value (pH ~12) to its surroundings when in contact with water (e.g., Van der Sloot et al., 2007; Hartwich and Vollpracht, 2017).

3. Differences and similarities with natural materials

Fig. 3 illustrates the position of the example materials discussed in this paper in a ternary diagram of the system SiO_2 - Al_2O_3 -CaO, together with the approximate fields of common industrial (by-) products and natural materials. Such diagrams have previously been used to characterize slags and ashes for their use as supplemental cementitious material in the ceramic and concrete industry (Adamiec et al., 2008; Snellings et al., 2012; Joseph et al., 2018), and to characterize glassy phases in bottom ash relative to natural materials (Zevenbergen, 1994; Dykstra Eusden Jr et al., 1999). MSWI Bottom ash (1) and (2) in Fig. 3 are fresh samples from Meima and Comans (1997) and Dijkstra et al. (2006b, 2008). The

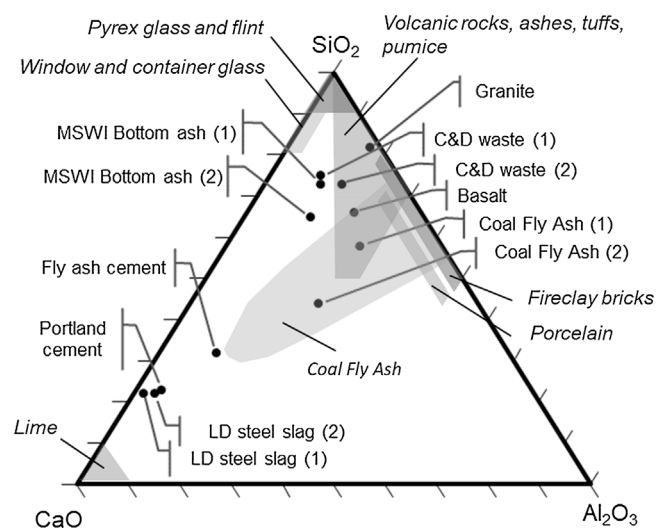


Fig. 3. Ternary diagram showing typical data for total Si, Al and Ca (wt%, expressed as their oxides) of MSWI bottom ash, construction and demolition (C&D) waste (predominantly consisting of concrete) and LD-steel slag (literature sources see main text). For illustration purposes, representative samples of ordinary Portland cement (Taylor, 1997) and fly ash cement (van der Sloot et al., 2011) are included; Coal Fly Ash (1) and (2) represent “Class 3” ashes from lignite and sub-bituminous coal burning (Taylor, 1997). Typical regions are indicated for volcanic ashes (Snellings et al., 2012), basalt and granite (Best, 2003), and regions of frequently used construction products (Zevenbergen, 1994).

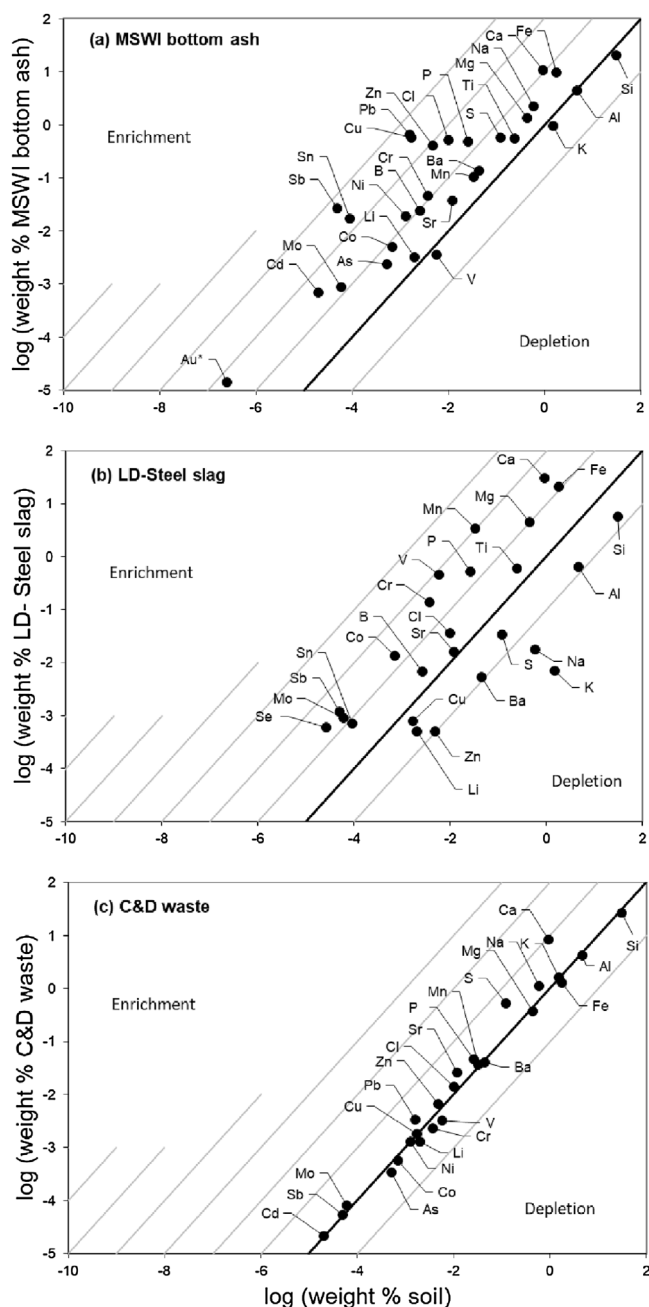


Fig. 4. Inspired by Kirby and Rimstidt (1993), this figure is constructed to show a comparison between the average weight percent of elements in topsoils (Shacklette and Boerngen, 1984; Sposito, 2008) and (a) MSWI bottom ash after ferro / non-ferro separation (data from Dijkstra et al. (2008)); (b) LD- steel slag (data from van Zomeren et al. (2011)); (3) Construction and demolition (C&D) waste (data from Butera et al. (2014)). Au* in panel (a) is the average from Allegrini et al. (2014) plotted against the Au crustal abundance (Wedepohl, 1995).

representative Dutch LD-steel slag sample (1) is adopted from Van Zomeren et al. (2011); sample (2) is from the USA (Yildirim and Prezzi, 2011). C&D waste (1) represents the average composition of Danish construction and demolition waste mostly consisting from cementitious materials (Butera et al., 2014); C&D waste (2) is Norwegian recycled concrete aggregate (Engelsen et al., 2009). For MSWI bottom ash and C&D waste in Fig. 3, Si, Al and Ca oxides constitute the vast majority of the total composition (65–80 %). The LD-steel slag is shown for comparison, even though these fractions account for 55 % of the total composition (85 % when corrected for

the residual iron). A $\text{SiO}_2\text{-FeO-CaO}$ diagram for steel slag is presented by Piatak et al. (2015).

The glassy nature of MSWI bottom ash becomes apparent due to its position in the upper corner of the diagram, and falls partially in the field of (pozzolanic) volcanic ashes used in the concrete industry. Despite the position of ordinary Portland cements (OPC) in the lower left corner, C&D waste falls into the glassy field. The latter is the result of the addition of about 80 wt% of natural (siliceous) aggregates such as sand and gravel during the production of concrete. LD-steel slag is relatively rich in Ca due to the addition of calcite and dolomite during the LD-steelmaking process, and contains relatively smaller amounts of Si and Al. This composition positions LD-steel slags close to that of Portland cement, but contrary to Blast Furnace Slag (BFS), the application of LD-steel slag as supplemental cementitious material is not common due to its expansive properties.

Fig. 4 shows the elemental composition of the three example materials compared to the average composition of topsoil. The soil concentrations are adopted from Sposito (Sposito, 2008) and refer to the average soil composition approximately 0.2 m beneath the land surface from uncontaminated mineral soils in the United States (Shacklette and Boerngen, 1984). These concentration data are representative for soils sampled worldwide (Sposito, 2008).

In the upper panel of Fig. 4, Dutch MSWI bottom ash (after separation of ferro- and non-ferro fractions and soluble salts) is compared to these values. The contents of Si and Al are quite similar to those in natural soil, while Ca and Fe are relatively enriched up to one order of magnitude. Interestingly, almost all other elements for which the content is known in MSWI bottom ash are relatively enriched by up to one order of magnitude (Na, Mg, Ti, S, Ba, Mn, Mn, Sr, Li, Co, As), two orders of magnitude (P, Cl, Zn, Cr, Ni, Mo, Cd) or even three orders of magnitude (Pb, Zn, Sn, Sb). MSWI bottom ash sampled in the USA in the early 1990's show similar enrichments (Kirby and Rimstidt, 1993), and given the uniformity of MSWI bottom ash sampled worldwide (see Supporting Information), this figure is expected to be not very different for bottom ash data from other sources. For Au, no data was available for the particular sample, but an enrichment of two orders of magnitude is found for Danish MSWI bottom ash (Allegrini et al., 2014). The relative enrichments are the result of the very diverse composition of the parent material, already being concentrated with these elements (see e.g., Hyks and Astrup, 2009), and the volume reduction of about 90 % during incineration. As the term “trace elements” is generally used for elements with a concentration below 100 mg/kg or 0.01 % (Sposito, 2008), many trace elements in soil would be termed “major elements” in MSWI bottom ash (Fig. 4; Ni, Cu, Zn, Pb, Cr, Sn, Sb, B).

Dutch LD-steel slag is relatively enriched in Fe, Ca, Mg and Mn, whereas Si and Al are relatively depleted (middle panel in Fig. 4). The high Fe content reflects the residual iron present in the slag, whereas the high contents of Ca and Mg originate from the added fluxes lime and dolomite. Other enrichments include Ti, Cr and V, of which the latter two are of interest from an environmental perspective (Huijgen and Comans, 2006; Van Zomeren et al., 2011). The slag is also relatively enriched with respect to Co, Sn, Sb, Mo and Se, but in absolute terms, concentrations are very low (0.01–0.001 %). Quite similar enrichments compared to average soils were also found for USA steel slags (Proctor et al., 2000).

Butera et al. (2014) presents the bulk chemical composition of a variety of Danish C&D wastes ($n = 33$, from 11 recycling facilities) consisting of (clean, unmixed) concrete, mixed masonry and concrete, asphalt and freshly cast concrete cores. The cement types included both fly ash and non-fly ash cements, but were mostly unknown. The average bulk chemical composition of these varying C&D wastes compares adequately to that of natural topsoils (lower panel in Fig. 4), which follows from the high contents of natural

aggregate in concrete (~80 %). It appears that C&D waste, and probably the recycled concrete aggregate in particular, is predominantly enriched in Ca and S which may originate from cement ingredients.

4. Geochemical weathering

Upon contact with water and air, weathering processes alter the material's character, including hydrolysis, oxidation, carbonation (i.e., the reaction of alkaline phases with carbon dioxide, see below) and mineral neoformation. Some processes are more manifest for one material than for the other. Which weathering processes dominate, depends strongly on the conditions under which the material is applied: open application (open to the atmosphere, exposure to rain and air), closed application (covered by an asphalt top layer or sealed by clay liners), or partially waterlogged (e.g., coastal defense). Furthermore, the rate of these processes depend on the grain size distribution of the material. A higher proportion of fine grains represents a relatively large reactive surface area with a corresponding much faster progression of weathering reactions.

4.1. Weathering of MSWI bottom ash

Weathering reactions of bottom ash proceed immediately upon contact with water, oxygen and carbon dioxide, and basically mimic the weathering processes documented for soils of volcanic origin (Zevenbergen et al., 1996, 1998; Astrup et al., 2016). Although it is well established that natural weathering leads to more stable secondary phases in the bottom ash, mineralogical transformations in bottom ash may continue for at least hundreds of years (Zevenbergen, 1994). One could strongly simplify the weathering sequence of bottom ash as follows. Initially, the most important mineralogical changes concern the carbonate chemistry: reaction of alkaline calcium-bearing phases with dissolved carbonate. On the medium term the neoformation of phases such as iron-(hydr)oxides occurs, while the long term mineralogical changes are dominated by the weathering of the glassy constituents (Zevenbergen et al., 1996, 1998; Meima and Comans, 1997; Piantone et al., 2004; Saffarzadeh et al., 2011).

Several weathering “stages” of bottom ash can be distinguished, of which the pH value of the material in contact with water is a useful indicator (Meima and Comans, 1997; Saffarzadeh et al., 2011). Unquenched bottom ash contains primary oxides that are formed during incineration, amongst which an important phase is lime. During first contact with water, i.e. in the quench tank, these oxides will hydrate. Consequently, freshly quenched bottom ash is characterized by a pH around 12.5 imposed by the portlandite solubility equilibrium. This stage is referred to as “stage A” (Meima and Comans, 1997). In particular in freshly quenched ash, interesting reactions take place that illustrate its highly unstable and reactive character, such as the corrosion of metallic aluminum present in the ash leading to considerable hydrogen formation (Miyake et al., 2018).

When all portlandite present in the ash is dissolved, the pH decreases to ~10.5 due to the formation of a mineral assemblage that temporarily stabilizes the pH, namely ettringite, gibbsite and gypsum. This stage is referred to as “stage B” and is generally reached after about 6 weeks of stockpiling open to the atmosphere (Meima and Comans, 1997). In several countries, including the Netherlands, regulations prescribe that quenched MSWI bottom ash is stockpiled before it is allowed to be used in construction (Astrup et al., 2016). During stockpiling, the ash is sufficiently “stabilized” in order to prevent excessive gas formation and swelling (e.g., due to hydration reactions) of the material during the intended use. Temperatures inside heaps may reach over 90 °C

as a result of exothermic reactions such as hydration and oxidation (Nørgaard et al., 2019) possibly speeding up otherwise slow reactions (Piantone et al., 2004; Astrup et al., 2016).

The reaction of alkaline phases in the bottom ash (such as portlandite and ettringite) with carbon dioxide leads to the precipitation of calcite. This process is referred to as “carbonation”, a commonly used term in the concrete sector (Taylor, 1997; Lagerblad, 2005; Galan et al., 2010). Carbonation is an aqueous reaction, i.e., CO₂ has to dissolve in water first in order to react with alkaline phases. This implies that under dry conditions, carbon dioxide can penetrate deeply in a deposit, but the absence of water prevents the carbonation reaction to take place. Vice versa, under water-saturated conditions the reaction rate is limited by the movement of dissolved carbonate ions, causing carbonation to proceed very slowly (cf. Lagerblad, 2005). The optimum conditions for carbonation are, therefore, partially water saturated conditions and a high relative humidity.

During continued carbonation, which uses both atmospheric CO₂ and CO₂ generated by microbial degradation of residual organic matter (Rendek et al., 2006), ettringite and other alkaline Ca-bearing phases dissolve, leading to a further pH drop to a more or less “final” pH value of around 8.3. This stage is referred to as “stage C” and is, in case of well-drained heaps of several meters high, generally reached after a few years (Meima and Comans, 1997; Piantone et al., 2004). The pH of around 8.3 is characteristic for a system consisting of calcite in equilibrium with CO₂ at atmospheric partial pressure (Meima and Comans, 1997).

It is estimated that the calcite buffer in MSWI bottom ash is large enough to stabilize the pH for thousands of years upon exposure to the atmosphere (Crawford, 1999). From an environmental perspective, this stage is preferred over the more fresh stage B (the typical stage at which MSWI bottom ash is applied in construction works in The Netherlands) and stage A. An advantage of C-stage bottom ash is that the potential leaching of alkalinity into soil has stopped (Van der Sloot et al., 2007). Secondly, most potentially toxic “heavy” metals exhibit a minimum solubility at this pH value, leading to lower leached concentrations. Third, prolonged weathering leads to the formation of secondary iron-(hydr)oxides from primary iron phases (unburnt iron pieces, spinel group phases, metallic inclusions). These amorphous and crystalline iron-(hydr)oxides have a high sorption capacity and act as an important sink for heavy metals and oxyanions (Meima and Comans, 1998; Dijkstra et al., 2006b; Wei et al., 2011). For these reasons, fresh (A or B-type) bottom ash is sometimes treated by a process referred to as “accelerated ageing”, carried out in large indoor facilities using moist air enriched with carbon dioxide (Dijkstra et al., 2006b; Astrup et al., 2016).

The glass fraction comprises about 65–75 wt% of the bottom ash (Kirby and Rimstidt, 1993; Zevenbergen et al., 1994b). Because glasses are metastable in natural weathering environments and alter to clays (Zevenbergen et al., 1994a, and references therein), weathering of the glass phase is likely to represent the dominant mineralogical change on the longer term. Zevenbergen et al. (1996, 1998) show that the weathering sequence of glasses in bottom ash follows a similar progression as in volcanic ashes and scoriae. In ashes weathered for 4 years open to the atmosphere, amorphous aluminium silicates were found (allophane), whereas in 12 year old ash deposits, direct evidence was found for neoformation of well-ordered primitive clay (illite). Such a rapid formation of well-ordered clay from glasses was not previously observed in nature; clay mineral formation in nature involves at least thousands of years of weathering (Zevenbergen et al., 1998, and references therein). The relatively rapid clay formation was attributed to the high specific surface area of the porous material in combination with the composition of the alkaline pore water, and was probably accelerated by alternating wetting and drying. In open ash

disposals, clay formation was found to be widespread, and shown to result both from in-situ transformation of glass into clay as well as glass dissolution and precipitation of clay (Zevenbergen et al., 1994a).

Interestingly, similar processes as observed in bog iron formation in peatlands may be observed in MSWI bottom ash deposits that have undergone prolonged weathering. Several studies on open ash disposals have revealed the formation of large and impermeable “iron pans” at places with alternating redox conditions, such as around the water table, at the underlying soil surface (Zevenbergen, 1994; Meima et al., 1997) or along preferential flow paths in the ash heap (Piantone et al., 2004).

In situations where MSWI bottom ash is applied as road subbase under well-drained conditions, ash sampled directly under an asphalt pavement (i.e. under more confined conditions) remains relatively fresh compared to ash near the sides (i.e. more exposed to water and air), as observed in field studies after 6–10 years of exposure (Dabo et al., 2009; Arm et al., 2011; De Windt et al., 2011). Limited entrance of water and air seems to conserve the initial properties of the ash for decades or longer (De Windt et al., 2011). However, when “field-moist” ash is sealed with impermeable liners, quite different mineralogical and physical transformations may occur. Even though such closed applications are highly common (if not standard) in The Netherlands (“IBC toepassing”, see Supporting Information), thorough investigations on the development of the ash properties are rare, as it would require breaking up a road. A well-documented case is the reconstruction of national highway (“Rijksweg”) 15 (Steketee, 2002). When its embankment was dismantled only ten years after construction, high pH values indicated incomplete carbonation (pH ~10). The embankment was also largely anoxic, confirming that exchange with the atmosphere was limited. Only near the sides and top, minor oxidation occurred, as inferred from visual evidence of iron(hydr)oxide precipitation. However, a more striking feature was the large-scale solidification of the ash deposit (Steketee, 2002). The degree of solidification varied spatially, but was observed virtually everywhere, except where the embankment was water-saturated (Steketee, 2002). Due to a construction error, settling caused the lower part of the construction to end up below the groundwater table. Cone penetration tests in the deposit revealed resistances up to 70 MPa, which is higher than highly compacted Pleistocene sands at large depths (~50 MPa; Van der Kolk, 2002). Examination of the binding material between the bottom ash particles using scanning electron microscopy - energy-dispersed analysis of X-rays (SEM-EDAX) revealed the presence of cement phases, most notably Ca-Al-Si phases (not further specified), calcite, iron oxides and to a lesser extent ettringite (Van der Kolk, 2002). Other studies have also found solidification of MSWI bottom ash, generally under poorly drained conditions (Zevenbergen et al., 1994a; Meima et al., 1997; Piantone et al., 2004). The formation of cement phases such as CSH and Calcium Silicate Aluminium Hydrates (CASH) in MSWI bottom ash has been further confirmed using spectroscopic techniques by e.g., Speiser et al. (2000); Bayuseno et al. (2010) and Saffarzadeh (2011).

4.2. Weathering of steel slag

The main weathering reactions that occur in steel slag - at least on the short term - are hydration, oxidation, and carbonation. In contact with water, primary reactive phases in steel slag, such as lime and dicalcium silicate, hydrate with portlandite and Calcium Silicate Hydrates (CSH) as reaction product (Van Zomeren et al., 2011; Hobson et al., 2017). Additionally, spinels and (hydr)oxide phases may form, and when areated calcite is formed because of carbonation (Hobson et al., 2017). The presence of residual iron in

steel slag in reduced form causes fresh steel slag to have reducing (oxygen-consuming) properties (Van Zomeren et al., 2011; Van der Sloot et al., 2007).

Steel slags produced in the Netherlands usually contain between 0–6 % free lime (Van Zomeren et al., 2011). Mainly due to the hydration of free lime to portlandite, but also due to oxidation of residual iron, steel slags may show volumetric instability when applied in construction works (Hendriks, 1999; Yildirim and Prezzi, 2011). In the Netherlands, steel slag needs to pass limit values on volume integrity before it is allowed to be applied in construction (Intron, 2016).

Under an asphalt pavement, Electric Arc Furnace steel slag, a slag type with similar mineralogy as LD-steel slag (Yildirim and Prezzi, 2011), is shown to remain virtually fresh in the middle of the road 10 years after construction (Arm et al., 2011). Small amounts of calcium silicate precipitates (not further specified) are found on the particle surfaces, in particular on particles close to the road sides (Arm et al., 2011). The limited weathering is attributed to the relatively dry conditions under the asphalt pavement (Arm et al., 2011).

In addition to humidity, the efficiency of steel slag carbonation depends on the surface area available for reaction, i.e. how finely ground the material is, and as a consequence the proportion of alkaline phases that can be liberated for reaction (Bacocchi et al., 2009). In accelerated carbonation experiments under a moist, CO₂-rich atmosphere (T = 90 °C/pCO₂ = 0.2 bar), using LD-steel slag that was ground and sieved to 2–3 mm, it is found that about 4 % of total Ca present in the material can be converted to calcite within 100–200 hours (Van Zomeren et al., 2011). Even under these forced conditions, pH of the reacted slag remains alkaline (pH 10–12.5 depending on the slag type), indicating that the carbonation is not complete. Carbonation experiments at high partial CO₂ pressure (T = 100 °C/pCO₂ = 19 bar) using finely ground (<38 µm) LD-steel slag leads to much higher efficiencies of about 74 % of total Ca within 30 min (Huijgen and Comans, 2005). These authors show that on an individual particle scale, carbonation requires the leaching of calcium from the steel slag particles into the solution first, followed by precipitation of calcite on the surface of these particles. They conclude that the reaction products formed on the outside of the particles preserve the alkalinity and reducing properties in the interior of the particles (Huijgen and Comans, 2005).

Coarse (45–180 mm) fractions of LD-steel slag are in The Netherlands predominantly used in water works as coastal protection (Table 2, Van der Sloot et al., 2007). In-situ measurements in a coastal protection in the Biesbosch, The Netherlands, show the formation of a discontinuity between the core of the (submerged) steel slag deposit that remains reduced and highly alkaline, and a thin zone close to the edge of the deposit, where neutral and oxidic conditions prevail (Comans et al., 1995). In the weathered zone, calcite precipitates abundantly, and iron is oxidized giving the steel slag a yellow colour, with red-brown coloured iron concretions. In the reduced zone, steel slag particles show a white colour, interpreted as portlandite precipitation on the outside of the particles (Comans et al., 1995). The same study shows in laboratory simulations that the thickness of the weathered zone is stable, but depends on the refreshing rate of the water. It is not known how long this discontinuity between core and edge is maintained. It may be expected that as long as exchange of water is possible between the core of the deposit and the surface water (e.g., through water flow), weathering is expected to continue to progress gradually towards the core. In surface waters with a sufficient volume and refreshing rate, application is considered to be environmentally acceptable due to the formation of the weathering layer (Van der Sloot et al., 2007).

4.3. Weathering of recycled concrete aggregate (RCA)

Even though many chemical and/or physical stresses of concrete are known (Hewlett, 2010), carbonation of Portland cement-based concrete is regarded as an important deteriorative chemical weathering reaction (Pade and Guimaraes, 2007; Galan et al., 2010). Similar to MSWI bottom ash and steel slag discussed previously, carbonation of concrete causes alkaline components of concrete, mainly portlandite and CSH, to be transformed to calcite. Much research has been devoted to quantify the uptake of CO₂ over the entire life cycle of concrete, with the purpose of more accurately determining its climate impact (e.g., Pade and Guimaraes, 2007; Galan et al., 2010). For intact concrete in contact with rain and air, the reaction starts at the surface and proceeds gradually inwards with the square root of time (generally 1–5 mm/√year for outdoor exposed surfaces, Lagerblad, 2005). A practical indication of the rate of this process include the carbonation front in a core drilled in the outer wall of a 120 year old fortress in the Netherlands (Pampus fortress), revealing a carbonation front of only 3–4 cm, whereas concrete drilled from a 2000 year old Roman aqueduct was fully carbonated (Van der Sloot et al., 2011). It should be noted that the carbonation rate of present-day concretes may be lower due to more optimized concrete recipes. In addition, some types of modern concretes are less susceptible for carbonation than others, such as high strength/performance concrete (Pade and Guimaraes, 2007). Because modern concrete is only negligibly carbonated when its constructions are demolished (i.e., only the outermost thin layer exposed to the weather), freshly broken RCA exerts a similarly high pH on its environment as new concrete, i.e. around pH 12 (Engelsen et al., 2009; Butera et al., 2014). However, because breaking increases the surface area available for reaction, the CO₂ uptake rate per unit mass of concrete increases dramatically (Engelsen et al., 2005; Lagerblad, 2005). Therefore, fine RCA fractions carbonate more extensively than coarse fractions (Natarajan et al., 2019).

A Norwegian field study (Engelsen et al., 2012, 2017) provides an illustration of the rate of carbonation for RCA in typical construction scenarios as a road base. RCA consisting of 94 % initially uncarbonated concrete was used (1) as a road sub-base under asphalt, and (2) exposed directly to air and rainfall (Engelsen et al., 2012, 2017). After 2 years, the pH of the infiltration water from the asphalt covered RCA decreased from pH 12.6 to pH 11.5, while that of the open RCA application was around pH 8. A phenolphthalein test indicated that in case of the RCA exposed to the atmosphere, the whole surface on each particle was found to be carbonated, but the thickness of the carbonated layer was only 1–4 mm. The observed difference in the effect of carbonation shows the influence of the asphalt covering, which reduces water infiltration and direct air exposure. In the Netherlands, complete sealing (“IBC toepassing”, see Supporting Information) of RCA in road bases is uncommon, because contaminant content and leaching potential are below the limit values above which sealing is prescribed.

Not all lime present in concrete is available for carbonation. Laboratory experiments have suggested that about 75 % of lime present in crushed concrete in the secondary life would be carbonated within a reasonable time span of 20–50 years (Engelsen et al., 2005). Due to progressive carbonation, it may be expected that on the very long term, concrete will ultimately transform into the basic components from which it was produced. To put it in geological terms, “the cement paste turns into marly limestone and the concrete into marly agglomerate; the old Roman concrete structures are basically such a rock” (Lagerblad, 2005).

5. Influence on natural geogenic environment

Studying the geochemical properties of novel anthropogenic materials, their weathering reactions and leaching behavior is positioned in the core of the “grand themes” identified for the field of urban geochemistry (Chambers et al., 2016). Leaching due to the infiltration of rainwater is of interest for the spreading of potential contaminants as well as for the geochemical “imprint” that these materials have on the underlying pristine soil (Comans et al., 2013, 2015).

Studies on potential contaminant leaching from MSWI bottom ash primarily focus on soluble salts, Cu, Sb and Mo (Dijkstra et al., 2006b; Cornelis et al., 2012) and from steel slag on V and Cr (Huijgen and Comans, 2006; Van Zomeren et al., 2011), as these may leach in critical amounts compared to limit values. Within the few decades of experience with the re-use of these materials in engineered constructions, cases of major impact on groundwater have not yet been reported in The Netherlands. This suggests that the (prescribed) application conditions and the legislation are sufficiently protective. International field studies seem to confirm this picture (e.g., Arm et al., 2011; De Windt et al., 2011; Engelsen et al., 2017). Exceptions include the extremely high pH values (pH 12–13) and anoxic conditions in shallow groundwater nearby parking lots constructed using fine-grained LD-steel slag (van der Sloot et al., 2007). These cases have led to a ministerial guideline and recommendations on the technical conditions at which steel slag is more safely applied (Netherlands Ministry of Housing Spatial Planning and the Environment, 2005; Van der Sloot et al., 2007). Although different in scale and conditions, hyperalkaline leachates to groundwater are reported for legacy iron and steel waste deposits elsewhere (e.g., Piatak et al., 2015; Pullin et al., 2019, and references therein).

A frequently used laboratory method to study the leaching mechanisms of (anthropogenic and natural) materials is the pH-dependence leaching test (e.g., EN 14997 (CEN/TC292, 2015), ISO TS 21268-4 (ISO/TC190, 2009), US-EPA method 1313 (US-EPA, 2012)). The method is based on parallel extractions at different pH values, and aims to determine the (near-) equilibrium solubility of elements from a material as a function of the pH (Van der Sloot et al., 1997; Dijkstra et al., 2006a; Ai et al., 2019). One of the extractions is carried out at the “native” pH of the sample, i.e. without acid/base addition. The usefulness of this method to unravel leaching processes is demonstrated in Fig. 5, for the example of calcium leaching from MSWI Bottom ash, LD-steel slag and concrete mortar (representing RCA). Fig. 5 shows data both for fresh material and a weathered and/or carbonated equivalent (Meima and Comans, 1997; Van Zomeren et al., 2011). The MSWI bottom ash had undergone natural weathering in a heap for 1.5 years; the other materials had been artificially carbonated by exposure to a moist, CO₂-rich atmosphere. Even though the example materials are of a widely different origin, the pH-concentration patterns of Ca show striking similarities (Fig. 5 and Supporting Information). The reason is that the pH-dependent leaching behavior of all materials obeys the same solubility constraints and mechanisms, although for different materials and/or samples these occur in different intensities (Dijkstra, 2007). Approximate stability fields of plausible solubility controlling phases are included in Fig. 5, based on several studies (Dijkstra et al., 2008; Engelsen et al., 2009; Huijgen and Comans, 2006; Loncar et al., 2016; Meima and Comans, 1997; Van Zomeren et al., 2011). The leaching curve of calcium follows in all three cases the earlier discussed sequence of carbonation of cementitious phases, i.e. portlandite, CSH, ettringite, and ultimately calcite (Fig. 5). The pH-window most relevant for estimates of dissolved concentrations that enter the underlying natural subsurface is roughly

Table 2

Common applications and grain size classes of the selected materials in civil engineering in The Netherlands (adopted from [van der Sloot et al., 2007](#)). Detailed technical specifications (such as grain size distribution, percentage foreign materials, grain density, bulk density, water absorption, mechanical resistance) for the materials in these applications are regulated in the so-called BRL specifications ([Intron, 2012, 2016](#); [KIWA, 2015](#)) and RAW clauses ([CROW, 2010](#)). N.a. = not applicable.

Material	Application in water works	Application in infrastructure
LD-steel slag	32–90 mm and 45–180 mm: coastal protection in marine and freshwater environments 16–32 mm: base layer / foundation for prefabricated water works elements	0–16 mm and 0–32 mm: road foundation under concrete or asphalt surfaces and paving; layer thickness 0.1–0.4 m. 0–8 mm: base layer / footing under paving, layer thickness a few cm.
LD- steel slag mixtures (LD-steel slag, Blast Furnace Slag, and Granulated Blast Furnace Slag in different proportions)	n.a.	0–5, 0–8, 0–22, 0–45 mm; road foundations (layer thickness 0.1–0.4 m), half-hardening (walkways, bicycle lanes, parking lots). These mixtures harden to some extent after application and compaction.
Recycled concrete aggregate	n.a.	0–22.4, 0–40 mm Road foundations, drainage material.
MSWI Bottom ash	n.a.	0–31.5 mm: large-scale fillings and embankments related to infrastructural works.

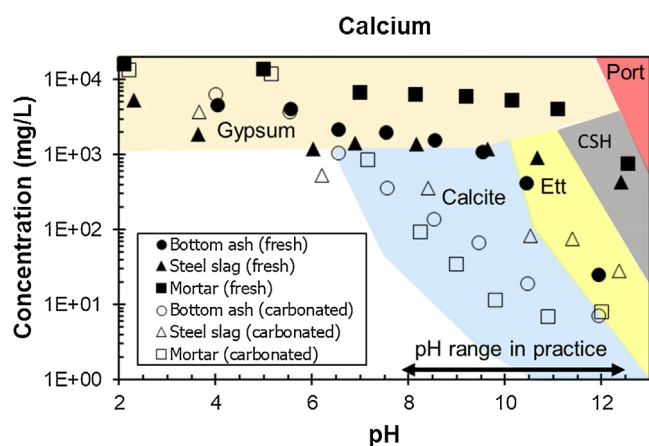


Fig. 5. Leaching of calcium as a function of pH measured using the pH-static leaching test (EN14997; [CEN/TC292, 2015](#)) for fresh and carbonated MSWI Bottom ash, LD-steel slag and cement mortar. Indicated on the X-axis are the approximate start-pH of fresh materials (pH ~12.5) and the pH of weathered/carbonated materials (pH ~8). Shaded areas represent the approximate stability fields for phases commonly reported to control leaching (see main text): “Port” = portlandite, “CSH”: calcium silicate hydrates; “Ett” = ettringite. Data bottom ash from [Meima and Comans \(1997\)](#); steel slag from [Van Zomeren et al. \(2011\)](#), cement mortar from TNO (unpublished, anonymized).

between pH 12.5 and pH 8, corresponding to a pH of the fresh materials and the final pH observed after complete carbonation and weathering ([Kosson et al., 2014](#)). Aided by geochemical models, the pH-concentration pattern over a wider pH-range can be used to obtain information on processes that control leaching of both major and trace elements ([Dijkstra et al., 2008](#); [Van der Sloot and Kosson, 2012](#); [Kosson et al., 2014](#)). Additional examples for other major elements (Si, Al and Fe) are provided in the Supporting Information.

Leaching test results need a mechanistic interpretation before (model- supported) predictions towards field scenarios can be made. Factors such as kinetics, intermittent wetting, and liquid-to-solid ratio need to be taken into account ([Kosson et al., 2002, 2014](#)). Results from leaching tests have been shown to form a functional proxy for field scenarios for MSWI bottom ash, steel slag and concrete aggregate for a range of major and trace elements ([Engelsen et al., 2012, 2017](#); [Kosson et al., 2014](#); [Di Gianfilippo et al., 2018](#)). Noteworthy is that pH dependence test data is available at TNO for a wide range of different natural and anthropogenic materials, both for major- and trace elements (metals, non-metals, metalloids), of which a significant amount is public domain and distributed with the database/expert system LeachXS ([Leaching Environmental Assessment Framework, 2019](#)).

Such information may become essential to assess long-term alterations of the underlying natural subsurface.

6. Synthesis and avenues for further research

An overall finding from our review is that MSWI bottom ash, LD-steel slag and concrete debris are formed during a thermal conversion process resulting in a thermodynamically unstable mix of reaction products. Although their bulk mineralogy is different, they have phase assemblages in common that are characteristic for cementitious systems, that lead to an alkaline character. Steel slag and bottom ash may additionally develop a reducing (i.e., anoxic) character in confined environments. Their extent of weathering depends strongly on the mode of application (open or closed from the atmosphere, wet or dry). Because their widespread re-use in constructions started in the mid-1980s, knowledge on long-term weathering and influence on the natural environment covers presently a few decades and is limited to relatively few, but well documented case studies. Truly long-term mineralogical evolution and influence on the underlying natural rocks and sediments might be derived from rare natural analogue sites ([Clark et al., 1993](#); [Pitty and Alexander, 2011](#); [Martin et al., 2016](#)).

Deposits containing novel anthropogenic materials are becoming volumetrically important in the shallow subsurface of The Netherlands, and likely also in other countries where circular-economy targets apply. Given the growing awareness of risks and opportunities associated with the shallow urban subsurface, improvement of representing these deposits in 3D-geological property models is necessary. Adequate representation of anthropogenic materials in our models requires the development of a more differentiated classification than is presently the case. Whereas the approach for Great Britain undertaken by the British Geological Survey is based on landform and origin ([Ford et al., 2010](#)), the three-dimensional modelling approach in The Netherlands is based on lithological properties as a starting point for classification ([Van der Meulen et al., 2013](#)).

Classification standards primarily developed for natural sediments ([Bosch, 2000](#); [ISO/TC182, 2018a, 2018b](#)) include coded lists for several (field-recognizable) anthropogenic “admixtures” (e.g. brick, sinters, waste), but anthropogenic materials are not differentiated further based on their specific lithological properties. Future research effort is needed on how novel anthropogenic materials can be accommodated in such standards, e.g., by additional distinctive properties (e.g., [Horckmans and Swennen, 2008](#)).

Pilot studies are foreseen on the acquisition of location and geometry of infrastructural works containing novel anthropogenic materials. Such information is expected to be fairly well available at local and/or national authorities. This implies that new sources

of data may be explored, for example, permits and design specifications of infrastructure works. Although spatial heterogeneity is potentially a problematic property of anthropogenic deposits (Price et al., 2011; Ford et al., 2014), modern “engineered” constructions have a certain predictability and homogeneity based on civil-technical requirements. This is illustrated in Table 2, showing applications and grain size classes of the selected materials in civil engineering based on Dutch guidelines (e.g., CROW, 2010; Intron, 2012, 2016; KIWA, 2015).

Finally, based on available lithological and geochemical data, and knowledge on weathering processes of these new materials, decisions can be made regarding the desired degree of differentiation of the lithological class “anthropogenic” in three-dimensional geological property models. Such a differentiation enables a more detailed description of the subsurface in the built environment, in support of informed decision making concerning its use.

Declaration of Competing Interest

None.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.ancene.2019.100229>.

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