Are geopolymers environmentally friendly?

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Abstract

The increase in environmental awareness in recent years across the world and in Turkey has led to the evaluation of the environmental effects of construction materials, in addition to their technical properties. Many binder systems are being discussed as alternatives to portland cement (classic) concrete. Geopolymers are a prominent such alternative. Geopolymers form structures through the partial or complete dissolution of a powder binder that does not react with water only. The powder binder is often an aluminosilicate such as ground blast furnace slag, fly ash, or metakaolin, and the activator is commonly an alkaline solution such as sodium hydroxide or sodium silicate. As geopolymer concretes don't contain portland cement and the powder binder used is typically an industrial waste or a minimally-processed natural material, they can have lower carbon dioxide emissions than classic concrete and be presented as evironmentally friendly. However, despite carbon emission being the primary criterion used in environmental impact evaluations, many others exist such as fresh water and marine water ecotoxicity, human toxicity, ozone layer depletion, acidification, and eutrophication. An economical, technical, and environmental evaluation of geopolymeric systems reveals that they are not currently a complete alternative to classic concrete and that they can be preferred for special applications. Considering the irreplaceability of cement and concrete in the near future, it would be wise to use greater

amounts of mineral admixtures in the production of cement and concrete, while searching for alternative materials.

Introduction

Despite being centuries old, the environmental movement (revolution) has spread with the observation of the first Earth Day (April 22nd) in 1970, the establishment of the Environmental Protection Agency (EPA) in the same year, and numerous international conferences held under the auspices of the UN in the following years. Through the efforts of several non-governmental organizations, legislation concerning topics like waste management, soil and water conservation was made, and the presently most widely used definition of "sustainable development" originated in the Brundlandt Commission Report [1] published in 1987: "Development that meets the needs of the present without compromising the ability of future generations to meet their own needs".

In the 90s, environmental awareness started affecting the construction sector, with the creation of green building documents such as LEED (Leadership in Energy & Environmental Design) in the US, BREEAM (Building Research Establishment Environmental Assessment Methodology) in England, and HQE (Haute Qualité Environnementale) in France, following the World Summit held in Rio de Janeiro with the participation of over 100 heads of state [2].

Portland cement (classic) concrete is the most widely used material in the construction sector. Portland cement, which lends its binding capability to concrete, is produced by burning low-cost and widely-available raw materials at very high temperatures, and is an exceptional material with its ability to react with tap water, produces a water-resistant reaction product with mechanical properties sufficient for its use in building. Despite its positive properties, its production at ~1500 °C and the need for its grinding from clinker require a high amount of energy. It also causes the emission of high amounts of carbon dioxide (CO_2) and lower amounts of other harmful gases, due to the reactions which take place in the kiln and its need to be ground from the clinker produced. While there can be some differences depending on its type and method of production, the manufacture of one ton of cement is reported to be responsible for an average of about 0.9 tons of CO_2 emitted [3]. The fact that the amount of cement produced annually worldwide surpasses 3.5 billion tons [4] and that this sector is responsible for 5% to 8% of anthropogenic CO_2 [3, 5], has led researchers to look for materials with lower ecological (environmental) impact. Among potential alternatives to portland cement systems are calcium sulfoaluminate cement, magnesium cement, magnesium phosphate-containing, alkali-activated, and geopolymeric systems [6]. Nowadays, geopolymers are perhaps the most popular among researchers. So, are geopolymers environmentally friendly?

What is a Geopolymer?

Geopolymers are made by partially or completely dissolving powder binders that won't react with water alone, using chemical activators. The powder is typically an aluminosilicate such as ground blast furnace slag, fly ash, or metakaolin, and the chemical activator is commonly an alkaline solution like sodium hydroxide or sodium silicate. As such, the name alkali-activated (slag, fly ash, etc.) is also used. Siliceous and aluminous typical geopolymers are compared with some other binders according to the framework of RILEM Technical Committee 224 (Alkali-Activated Materials) in Figure 1.

Figure 1. Comparison of geopolymers with other binders in terms of chemical composition [7].

As seen in Fig. 1, while the name geopolymer is often preferred for low-calcium systems, alkali-activated is used to encompass high-calcium systems such as slag, as well. However, there exist geopolymers made using slags of other metals, as well as acid solutions or other activators.

Although the name geopolymer was coined in the 1970s, due to typical examples being mostly amorphous, containing elements abundant in the earth's crust, and having 1-, 2-, and 3-dimensional structures similar to those of organic polymers, systems with similar chemistry have been known at least since the 1930s [8]. In calcium-rich systems like slag, calcium silicate hydrate (C-S-H) gel similar to that formed during cement hydration also forms. Depending on the powder binder and activator used, and on curing conditions, geopolymers with better resistance to acids and other chemicals than portland cement paste, able to resist temperatures of 1000-1200 °C without suffering damage to their internal structure [9], showing low alkali-aggregate expansion, and high freeze-thaw, sulfate, and corrosion resistance [10] can be made. Geopolymers can have lower or higher strengths and elastic moduli depending on their Si:Al ratio. Examples with strength close to 100 MPa [11], and 20 MPa strength gain in 4 h at room temperature [8] exist. On the other hand, while some geopolymeric mixtures set and gain strength at room temperature, a great many of the mixtures in the literature require curing at 40 to 80 °C for 6 hours or longer to attain strengths comparable to those of portland cement systems [12, 13]. The use of geopolymers for heavy metal [14], toxic material [15], and radioactive waste [16] containment has been suggested. Detailed information on different geopolymeric systems and their properties is available in various books [8, 17, 18].

Comparison of the Carbon Emissions of Classic Concrete and Geopolymeric Systems

It is known that the production of classic concrete results in large amounts of carbon being emitted. According to some studies in Australia, of the green house gas emission from concrete 70 %-75 % is associated with cement, 17 %-25 % with aggregate production and procurement, and 3 %-5 % of the remainder with the transport of concrete [19, 20]. As geopolymers don't contain portland cement and the powder binder used is typically an industrial waste or a minimally-processed natural material, they can have lower carbon dioxide emissions than classic concrete. Some factors influencing the amount of emitted equivalent CO_2 by geopolymers are given in Table 1.

Table 1. Some factors influencing the amount of emitted equivalent CO_2 by cement concrete and by geopolymers.

Differences in the known or assumed values used in different studies for these factors can affect the calculated equivalent carbon emission of geopolymers. Most studies compare geopolymers with concretes containing cement only (no admixtures) where as fly ash- or slag-incorporated cements concretes have been reported to emit 13 %-22 % less equivalent CO_2 than cements with no admixtures [21]. While it has been reported that the CO_2 emission of geopolymeric systems can be up to 80 % less than portland cement systems [22, 23], in calculations using different mixture ingredients, proportions, etc., this drop can be much less. Some studies have reported this drop in equivalent CO_2 emissions as 26 %-64 % [24, 25], and even 9 % [19]. Despite comprising a small part of the total volume or mass, activators contribute the most to the carbon emission of geopolymers (Table 2).

Table 2. Effect of portland cement and different geopolymer ingredients on global warming potential.

Carbon Emissions and their Role in Environmental Impact Evaluations

It is a widely-accepted scientific opinion that anthropogenic (man-made) CO_2 and other (CH_4, N_2O, fluorinated gases) green house gases cause climate change [26]. As it constitutes 84% of man-made green house gases (about 6.7 billion tons per year), CO_2 is defined as the primary green house gas and in calculations the harming impact of other gases is converted to CO_2 impact (equivalent CO_2). Nevertheless, the harming impacts per ton of some of the other gases are much greater. For example, the polluting power of CH_4 is reported as ~25xCO_2, and that of N_2O as ~300xCO_2. There also exist important differences between the different gases such as the amount of time they remain in the atmosphere [27]. The fact that CO_2 is known as the primary green house gas can cause it to be considered as the only criterion for evaluating the environmental impact of a material among construction materials researchers and among the general public. It can thus be deemed sufficient to reduce the carbon emission of a material or product in order to reduce its environmental impact.

Nowadays, the environmental impact of a product (e.g. concrete) is assessed using the life cycle analysis (LCA) method and considering the materials it is made of (e.g. cement), all the industrial processes related to the production of these materials (e.g. concrete), and taking into account all stages of its use (“cradle to grave”), its production, use, and after-life (recycling, disposal, etc.), as well as the related resource consumption, energy consumption, and emissions to air, to water, and to soil. This method is used to assess numerous different global and local environmental impacts together [28]. While these impact categories can vary between analyses, the principal ones are presented in Table 3.

Table 3. Life cycle analysis impact categories [28, 29].

CO_2 emission only affects global warming potential among these categories. Fluorinated gases, however, cause ozone depletion, and N_2O causes eutrophication (essentially the reduction of oxygen in a body of water due to an increase in nutrient amount). Factors other than CO_2 emissions and related impacts need to be taken into account when evaluating the environmental impact of classic concrete and of geopolymers.

Life Cycle Analysis of Geopolymer Concretes

Although there are many studies on the carbon emission of geopolymers, very few studies report on their life cycle analyses. Among these, some only focus on one or two categories in which geopolymers are typically advantageous such as carbon emission, energy consumption, and resource consumption [30, 31, 32].

In the most thorough existing study, Habert et al. [24] took 79 geopolymer paste and concrete mixtures containing fly ash, ground blast furnace slag (GBFS), and/or metakaolin as their powder binder, and sodium hydroxide and/or sodium silicate as their activator, from 15 articles in the literature, and evaluated the environmental impact of producing 1 m^3 of concrete.

One average mixture was selected for each type of the 49 fly ash-, 13 slag-, and 17-metakaolin-containing geopolymer mixtures, and the proportions of these three mixtures were used in LCA. Results were compared with the environmental impacts of producing 1 m^3 of classic concrete with identical 28-day strength, and with 1 m^3 of concrete containing 30% mineral admixture. The cement content of the classic concrete was chosen to yield the desired strength [24]. The aggregate/paste ratio of the geopolymer concrete was used for the classic concrete as well. As an example, the fly ash-containing mixture had 408 kg/m^3 of fly ash, 17 kg/m^3 of solid NaOH, 103 kg/m^3 of sodium silicate solution, 1848 kg/m^3 of aggregates and a compressive strength of 36 MPa. Classic concrete with the same strength was calculated to contain 354 kg/m^3 of cement.

The data used to calculate the environmental impact of 1 kg of the materials in the concrete mixtures were taken from Chen [33], and Chen et al. [34]. The LCA methodology is explained in the ISO standard 14040 [35]. The specific method used in Habert et al. [24] was CML01 [28] which takes into account 10 different environmental impact categories. Data about the emissions and energy of the primary raw materials and secondary materials were taken from the Ecoinvent [36] database. The LCA results of geopolymeric, 100% portland cement-containing, and 30% mineral admixture-containing concretes are compared in Figure 2.

Figure 2. Comparison of LCA results for geopolymeric, classic, and slag-containing concretes (modified from Habert et al. [24]).

As seen in Fig. 2, the global warming potential caused by fly-ash or slag geopolymers, or 30% mineral admixture-containing blended cement concretes is lower than that of classic concrete. That of the metakaloin geopolymer is higher. Since metakaolin is produced by burning kaolin at 600-800 °C and the amount of sodium silicate needed to achieve high strengths is greater than with slag or fly-ash geopolymers, the associated green house gas emission is relatively high.

All three types of geopolymers appear to have greater impacts than classic concrete in the impact categories other than global warming potential. Their (negative) impact is 8 to 15 times greater in terms of fresh water ecotoxicity, 7 to 10 times greater in terms of marine water ecotoxicity. Differences in impact in other categories can be up to several times.

The environmental impact of geopolymers depends heavily on activator production, particularly the production of sodium silicate. Sodium hydroxide can be produced via electrolysis in the chlor-alkali process, which requires a large amount of electrical energy. Sodium silicate is produced from silica sand and sodium carbonate at around 1400 °C and under pressure. Sodium carbonate is produced from ammonia, limestone, and salt water in the Solvay Process or as a product of the chlor-alkali process. In the Solvay Process, limestone is calcined at around 1000 °C to produce CO_2. Ammonia, in turn, is produced under pressure and at high temperatures in the Haber Process [37, 38]. It can easily be seen that each activator in geopolymer concretes is itself the product of a chemical process not simpler than the production of portland cement and as such the related environmental impact cannot be overlooked.

When the three types of geopolymers are compared, it is seen that the slag-containing ones have the lowest environmental impact. However, even that type has a more negative environmental impact than the blended cement concrete in all categories but global warming potential. As it is possible to use even greater amounts than 30 % of a mineral admixture in concrete, blended cements and concretes appear to be a good alternative.

Important Provisions/Factors Infuencing the Results of Life Cycle Analyses

Results obtained with LCA are only as reliable as the data used and assumptions made. Hence, analysis results can deviate from the truth if the assumptions about the production, acquisition, etc. of the materials that classic concrete or the geopolymers contain (e.g. the cement or chemical activators) do not reflect the actual situation. For example, while the amount of energy required for the production of one ton of clinker is theoretically 1.6 GJ [39], the actual average consumption is 2-3 GJ. This consumption can exceed 5 GJ in companies employing old technologies [40]. 100-150 kg of medium-quality coal is needed for this extra consumption, resulting in 250-600 kg extra emitted CO_2 per ton of cement.

It is not easy to compare classic concrete and geopolymer concretes in terms of environmental impact. Just as there can be examples of classic concrete containing larger or smaller amounts of cement, there are geopolymers containing more or less chemical activator. One method of comparison can then be to compare mixtures with similar ultimate strengths, as in the above-mentioned studies. However, as there can be differences between the rates of strength gain, fluidity, and durability of these two products, mixture proportions need to be considered taking into account the properties desired of the concrete. For example, typical geopolymer concretes reported on in the literature are cured at temperatures slightly above room temperature, consequently their strength gain rates are high. However, many geopolymers gain strength slower than classic concrete at room temperature, and some not at all. Conversely, the resistance to high temperatures, acids, and sulfate solutions of geopolymers is greater than that of classic concretes. Hence it may not be reasonable to compare mixtures with equal strength. Also, if a chemical admixture is used to obtain properties like fluidity, etc., the type and amount needed may differ for these two chemical systems.

As mentioned above, activator chemicals are primarily responsible for the negative environmental impacts of geopolymers. Sodium hydroxide is sold as a solid, at different purities, and does not show much variation. Its effectiveness (activity) is easy to evaluate by looking at past studies since it is typically used as a concentrated solution. On the other hand, sodium silicates are available in very many classes depending on their SiO_2/Na_2O ratios, and can be solids or solutions of varying fluidities. Hence both their activities and their environmental impacts can vary. For a given amount of powder binder, lower quantities of a more effective binder can yield the desired strength. This makes it difficult to evaluate the environmental impact of the production of sodium silicates used in different studies. Also, these two activators can be used alone or together to achieve a target strength. Their activator effectiveness at room temperature and at elevated temperatures also vary [41]. It should not be forgotten that, while less common, activators such as potassium hydroxide, potassium silicate, alkali sulfates, carbonates, and aluminates also exist [18]. There can be substantial differences between the effectiveness, economics, and environmental impact of activators.

Another factor complicating the LCA of geopolymers is the reactivity of the powder binder used. Metakaolin and slag used in studies can be of different structure and reactivity depending on the burning temperatures and cooling rates during their production. To equal the strength achieved in one study using a very reactive slag will require greater amounts of activator with a less reactive slag under the same curing conditions, resulting in greater environmental impact. As fly ashes are typically classified according to their calcium contents, burning conditions such as temperature, etc. of the coal, which affects their reactivities, are not reported in studies. Some of the fly ashes in Turkey do not form geopolymeric structures even with very concentrated sodium hydroxide solutions and using oven curing. The possibility of using powder binders other than the ones mentioned here, such as natural pozzolans or other wastes [42, 43] is worth mentioning. The use of materials not able to be used for cement replacement such as ferronickel slag [44] or tungsten mine waste mud [45] can make geopolymer systems more meaningful in terms of environmental impact. Of course, the distance of the powder to its site of use, and its abundance, are also important.

There are several factors that may indirectly influence LCA and are difficult to assess. For instance, as there is little information about the durability of geopolymers, their service lives can only be predicted. Materials with long service lives and that don't require maintenance could have very low environmental impact. It is possible that non-cement mixtures containing high doses of fly ash pose a threat to their surroundings due to fly ash being obtained from coal burning and its radiocativity increasing, or that their pulverisation at the end of their service lives causes problems due to the heavy metals they may contain. There is an insufficient amount of research and information on such topics.

There also exist several methods that can be followed for LCA, and they can yield different results. Even the decision of whether a material is to be considered an industrial waste or a by-product can ve complicated [24, 46] and can affect the calculation.

Conclusion

It is possible to produce geopolymer concretes with lower net carbon emissions than classic concrete. In return, the environmental impact of geopolymers can be more negative than that of classic concrete in many other categories. The environmental impact of construction materials should be evaluated by taking into account not only their net carbon emissions but all related processes, using life cycle analysis. The materials that geopolymers contain, the amounts used, and their production methods can show more variation than portland cement and cement production methods. Therefore the assumptions made and values used for the geopolymer for which LCA is performed should be realistic with regard to local and special conditions.

Natural, or waste/by-product powder binders are used in large amounts in blended cements. Hence the carbon emission and other negative impacts of using cement can be reduced significantly. Fly ash and slag can compete with portland cement in terms of their contribution to carbon emission even when transported long distances [20]. It is estimated that by 2030, through the use of admixtures and other methods, the average CO_2 emitted for 1 ton of cement produced will be lowered to 0.7 tons [5]. The activators used in geopolymers also affect the economics of the concrete produced. The price of geopolymer concrete can vary for different mixtures, from slightly lower than classic concrete, to as much as twice as high, per cubic meter [25].

In conclusion, while the question in the title of this article does not have a simple and definitive answer, the choice of geopolymers to replace classic concrete does not appear to be rational from an environmental point of view, although it can be for certain limited applications in which they provide technical advantages.

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Figures

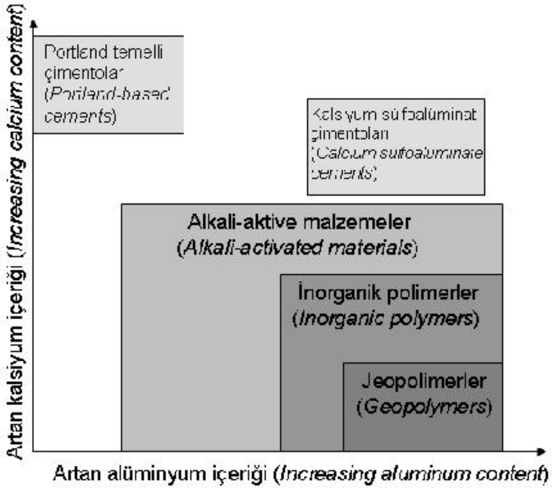

Figure 1. Comparison of geopolymers with other binders in terms of chemical composition [7].

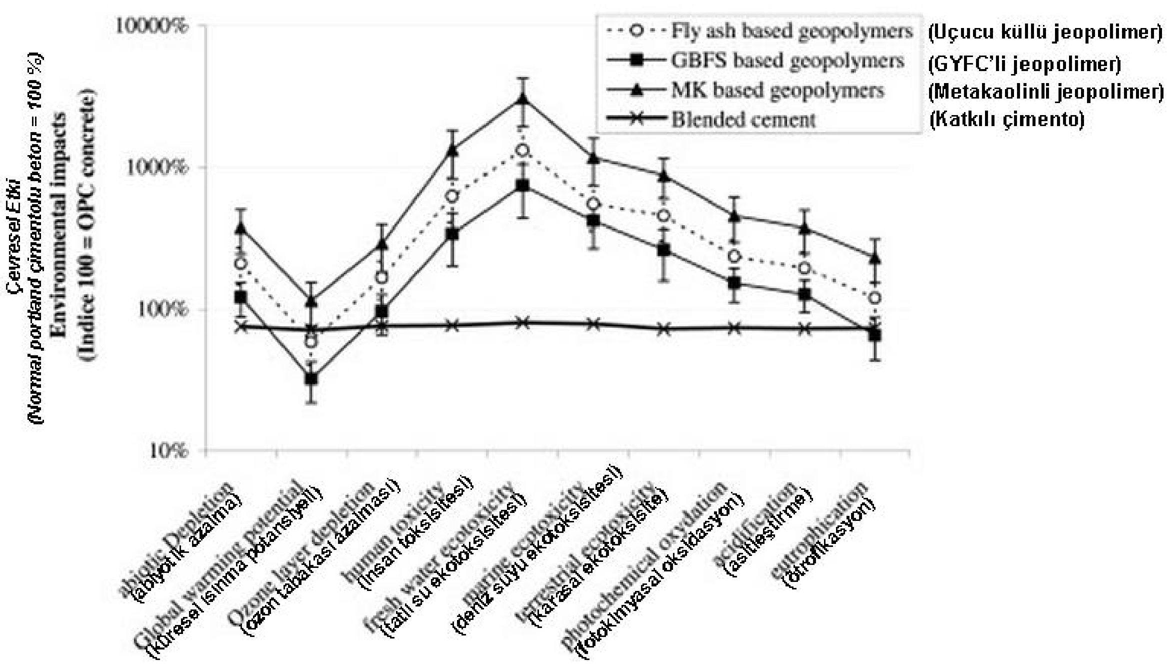

Figure 2. Comparison of LCA results for geopolymeric, classic, and slag-containing concretes (modified from Habert et al. [24]).

Tables

Table 1. Some factors influencing the amount of emitted equivalent CO_2 by cement concrete and by geopolymers.

Çimentolu (Cementitious)	**Faktör (Factor)**	**Jeopolimerik (Geopolymeric)**
✓	malzeme temini, işlemesi, taşınması (material procurement, processing, transport)	✓
	aktivatör üretimi için gereken enerji (energy required for activator production)	✓
✓	toz bağlayıcı miktarı (powder binder content)	✓
✓	enerji için kullanılan yakıt türü (type of fuel used for energy)	✓
	kullanılıyorsa sıcak kür için gereken enerji (energy required for heat curing, if being used)	✓
✓	toz bağlayıcı kaynağının kullanım yerine uzaklığı (distance of powder binder source to place of use)	✓
✓	hava koşulları (weather conditions)	✓
✓	tesis tipi (type of plant)	

Table 2. Effect of portland cement and different geopolymer ingredients on global warming potential.

İçerik Malzemesi (*Ingredient*)	**Etki (*Effect*)**
çimento (*cement*)	yüksek (*high*)
uçucu kül (*fly ash*)	düşük (*low*)
yüksek fırın cürufu (*blast furnace slag*)	düşük - orta (*low-intermediate*)
agrega (*aggregate*)	çok düşük (*very low*)
su (*water*)	çok düşük (*very low*)
metakaolin (*metakaolin*)	orta (*intermediate*)
sodyum silikat (*sodium silicate*)	yüksek (*high*)
sodyum hidroksit (*sodium hydroxide*)	orta - yüksek (*intermediate-high*)

Table 3. Life cycle analysis impact categories [28, 29].

Etki Kategorisi (Impact category)	Birim (unit)
abiyotik azalma (*abiotic depletion*)	kg Sb (antimon) eşdeğeri (*kg Sb [antimony] equivalent*)
küresel ısınma potansiyeli (*global warming potential*)	kg CO_2 (karbondioksit) eşdeğeri (*kg CO_2 [carbon dioxide] equivalent*)
ozon tabakası azalması (*ozone depletion*)	kg CFC-11 (kloroflоro karbon 11) eşdeğeri (*kg CFC-11 [chlorofloro carbon] equivalent*)
tatlı su ekotoksisitesi (*fresh water ecotoxicity*)	kg 1,4 DCB (diklorobenzen) eşdeğeri (*kg 1,4 DCB [dichlorobenzene] equivalent*)
deniz/sucul ekotoksisitesi (*marine ecotoxicity*)	kg 1,4 DCB (diklorobenzen) eşdeğeri (*kg 1,4 DCB [dichlorobenzene] equivalent*)
karasal ekotoksisitesi (*terrestrial ecotoxicity*)	kg 1,4 DCB (diklorobenzen) eşdeğeri (*kg 1,4 DCB [dichlorobenzene] equivalent*)
insan sağlığı ve ekotoksisitesi (*human health and ecotoxicity*)	kg 1,4 DCB (diklorobenzen) eşdeğeri (*kg 1,4 DCB [dichlorobenzene] equivalent*)
Asitleştirme (*acidification*)	kg SO_2 (kükürt dioksit) eşdeğeri (*kg SO_2 [sulfur dioxide] equivalent*)
Ötrofikasyon (*eutrophication*)	kg PO_4^- (fosfat iyonu) eşdeğeri (*kg PO_4^- [phosphate ion] equivalent*)
fotokimyasal oksidasyon/hava kirliliği (*photochemical oxidation / smog*)	kg C_2H_4 (etilen) eşdeğeri (*kg C_2H_4 [ethylene] equivalent*)