DUST – A GEOLOGY-ORIENTATED ATTEMPT TO REAPPRAISE THE NATURAL COMPONENTS, AMOUNTS, INPUTS TO SEDIMENT, AND IMPORTANCE FOR CORRELATION PURPOSES

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ABSTRACT. The paper deals with the recent and present-day natural components of the dust inputs to sediments, and aims to attract the attention of geologists to atmospherically mediated teleconnections among basins across the globe. Similar components as today can also be expected to exist in the pre-human history of the Earth, with capability to affect the quantity and quality of non-carbonate phases in limestones. However, a significant part of atmospheric dust sedimentation is not sufficiently covered by standard measurements. Owing to air quality regulations, most of the present research is conducted to understand the emissions and atmospheric load of PM_{10}, and a lesser number of studies map the possible transport of coarser natural particulate matter over the large distances. With respect to the studied particle sizes and methods of their determination, the present-day aerosol science and geological approach, focused on measured sedimentation or input to sediment, show practically no overlap. We report the first evidence (or at least a well-reasoned hypothesis) that the low numbers of frequently occurring large particles, e.g. with 50-μm d_e or larger, must always represent a substantial mass added to mineral dust budgets. These rare and bulky particles are either transported with the super-storm dust plumes in the troposphere, or with the jet streams near the tropopause. From the geological point of view, it is important to consider all sizes of mineral-lithic particles or grains, particularly from the silt to fine-sand sizes (i.e., 4–250 μm). In atmospheric physics, only the total suspended particulates (TSP) are a partial and often unreliable counterpart. This subject is especially worth of exploring although the emerging discipline, combining the estimates of the total burden of atmosphere by every classes of the natural solid particles and their measurable 'final sedimentary inputs', is still encumbered with much imprecision, and the reported results are more concerned with the principles and rough estimates than all variants of calculations. Classifying the components by their sources, this attempt suggests that the ideal mean airborne inputs to the sediments on the present-day Earth are approximately as follows ($g/m^2/yr$): terrestrial weathering ≈ 3.3; volcanic ash ≈ 0.3; biotic ≈ 0.5; cosmogenic ≈ 0.0002; wildfires ≈ 0.3; solids with ultra fine secondary aerosols ≈ 0.02, giving a total of ≈ 4.4. Therefore, a mean (ideal) input of the natural dust to the present (and possibly also Holocene) sediments of about 4–5 $g/m^2/yr$ should be considered. Naturally, the real inputs vary geographically to a large extent (± a few orders of magnitude).

KEYWORDS: Natural dust, particulate matter, aerosol, sedimentary background, sedimentary inputs, dust teleconnection, limestones, iron contents, Holocene, Present

1. Introduction – aims and constraints

Natural atmospheric dust is a ubiquitous component of our present-day environment. In spite of numerous studies published in this field (ca. 15,000 contributions to related research in 2009), even the broadly accepted reviews (e.g. mineral dust budgets, Zender et al., 2004; Tegen & Schepanski, 2009; Maher et al., 2010) admit that it will be a long and complicated process to achieve the most generalized, accurate and reliable data.

In spite of this, a better understanding of the amounts and components of the present natural dust, including organic and rare particulates, can play a crucial role in the study of transport and deposition of this material in geological history of the Earth. Therefore, this study is primarily orientated at a possible geological reappraisal of this issue. The terms 'recent, present or today' used in this study mean a vaguely defined time interval, during which the estimated inter-annual, decadal and millennial mean values were searched. They refer to the data availability – simply involving geological present, Holocene, the last 12 thousand years and usually less than that. The idea followed in this work is that this step of reassessment of the dust budgets and inputs must rather precede than follow the implications for the geological past.

However, even if we achieve some improvement in the geological value of the data on the 'dust efficiency' today, any future implications for the settings in a deep history of the Earth may remain complicated. The effort to translate results on fluxes and background sedimentary rates to specific situations of the geological past has many limitations (Van der Hoven & Quade, 2002; Stancin et al., 2008; Hladil et al., 2009). These limitations lie in imperfectly known parameters on palaeogeography, atmosphere composition, atmosphere–ocean circulation (i.e., climatic data package) and, not negligibly, also on the density of vegetation cover on land. The presence or absence of vegetation is nowadays a preponderant factor for dust emissions in many areas, but these conditions were strongly varying in the geological history. The first

changes in continental environments from a bare bedrock and unconsolidated sediment blankets towards the complex and diverse alluvial landscapes influenced by the presence of terrestrial plants occurred episodically between the Ordovician and Devonian periods (Davies & Gibling, 2010). However, it is assumed that the scarcity of vegetation cover over vast inland areas was typical for many Palaeozoic and Mesozoic settings (e.g. in the Devonian; Loboziak & Melo, 2002). This made the continent interiors a more efficient dust source than they are today. These rather weakly described factors of the past eras (atmosphere regimes, sedimentary cover, weathering, vegetation cover, etc.) certainly worked in synergy with other environmental parameters, such as major episodes of detritism, volcanism or impacts of large meteorites, which all affected the fluctuating but occasionally very high atmospheric load of these particulates.

Since our topic focuses on truly natural 'dust cycle' today (thought to be later confronted with those of 'pre-human times'), the additional ca. $^1/_3$ of the atmospheric load which relates to the present-day human activities cannot be involved. This very rough calculation of a 'not natural, additional mass of dust' is based on a combination of data on added combustion-derived iron (e.g. Mahowald et al., 2009; Sholkovitz et al., 2009; Maher et al., 2010) and human and urban impact on the landscape and their emission capability (e.g. Geist, 2005). This $^1/_3$ represents only a tentative value, because an accurate separation is almost impossible. The subtraction of the unnatural dust was made individually, respecting the specifics of the individual components. The scarcity and selectivity of data on mineralogy and trace element geochemistry of these particulates is an unfavourable circumstance in general, which introduces a significant but hardly quantifiable bias.

Other problems or uncertainties in the comparisons and recalculations of the measured amounts of particulate matter relate to the understanding of dust size classes. These classes can be expressed by physical (real) equivalent diameters of the particles (d_e), where d_e relates to a tangible object which is handled in the way common in geological and material sciences. However, in the air-pollution monitoring disciplines, the particle diameters have a dynamic meaning (d_a): for example, PM_{10} displays index 10 but this index relates exclusively to the '≤10 μm' in terms of comparable dynamic behaviour. Hence, we can say that such diameter corresponds to the diameter of a sphere with a density of 1 g/cm^3 which has the same settling velocity in air as the particle itself. The different methods and standards used for the determination of the total suspended particulates (TSP) and PM_{10}, $PM_{2.5}$ etc. employ numerous formulae (e.g., for direct or indirect measurements, common or specific emissions, standardized or specific particle sizes, combing aerodynamic and electrical mobility, and also with regard to the normative measures or regulations; Jaecker-Voirol & Pelt, 2000; Khlystov et al., 2004; US EPA, 2010). Hence, many of the PM_{10} determinations cannot be directly used for the 'geological or material' dust budget calculations and must be transformed using the functions, formulae or empirically adjusted estimates. Where appropriate, the basic formula relating the mobility size to the aerodynamic size was used (Hinds, 1999; Khlystov et al., 2004):

$d_p = d_a \cdot (\chi \cdot c_s(d_a)/c_s(d_p) \cdot \varrho_0/\varrho_p)^{1/2}$,

where d_p and d_a are the mobility and the aerodynamic diameters, respectively, χ is the shape factor, c_s is the Cunningham slip correction factor, ϱ_0 is the reference density (1 g/cm^3) and ϱ_p is the density of the particle.

However, the further simplifications to

$d_p = d_a \cdot (\varrho_0/\varrho_p)^{1/2}$, or even $d_e = d_a \cdot (\varrho_0/\varrho_p)^{1/2}$,

or finding the size correction factor

$x = (\varrho_p/\chi \cdot \varrho_0)^{1/2}$,

may introduce large errors (X to XX %; Khlystov et al., 2004) to the calculations of large particles (X to XX μm).

Therefore, simple bridging characterizations of the mineral-lithic particle sizes such as

$d_e = d_0 \cdot (\varrho_0/\varrho_p)^{1/2}$ or $d_e = (\varrho_0/\varrho_p)^{1/2}$

(e.g. Hladil et al., 2008) are very rough. The only advantage of such simplification is that it easily indicates the smallest diameters of heavier solid particles. For example, a 'max.-size PM_{10}' quartz particle may possess a ca. 6 μm d_e, pyrite ca. >4 μm and so forth, in a row from the light to heavy mineral particles. Conversely, the 'very light' hollow particles (pollen grains, some spores, hollow shells or subtle foam fragments) must correspond to larger diameters, but the behaviour of these large, aerodynamically light and complex shape objects is often much beyond that which can be described by the formulae. Therefore, considering specific or collective behaviour of large particles of 'geological' type, we must always rely on observations, experiments and simulations.

Remarkably, in spite of these principal complications, the term PM_{10} is not used exclusively as being directly related to stream flow analysers of air mass but, occasionally, also in the material context (i.e., addressing the chemistry, phases, structures and size & shape of natural dust components; e.g. Cwiertny et al., 2008; Maher et al., 2010).

Other sources of uncertainty (discussed only marginally here) relate to the transport range, residence time in the atmosphere or sedimentation velocity for the individual sorts of materials, as well as sizes and shapes of the particles. This is caused by the fact that the presently used detection and modelling techniques in atmospheric physics relate to the dust (aerosol) particles with the upper size limits of ≤10 or ≤20 μm (Uno et al., 2006) and TSP (≤100 μm), but largely do not apply to the sizes and components typically found in the sampled and scrutinized 'geological, real dust sediments'. Moreover, in many geological settings, emission areas are already depleted in the finest particle ranges (deflation). In such emission areas, the proportion of coarse particulates gradually increases (e.g. Wang et al., 2005). The strongly deflated soil or sediment surfaces 'work' as a source of coarse-

grained suspensions only in cases of strong turbulences with gusts of 30–70 m/s velocities, ca. 110–250 km/h (Pashkevich, 1970). For smaller natural sand grains, the 20±10 times higher wind speed than the relevant terminal fall velocities act with a possible acceleration of 'a mighty tear-up and vigorous emission'. These mechanisms are predominantly related to local and regional aeolian transport.

An often underestimated fact is that large mineral-lithic particles may also be distributed across the world as diluted formations in the jet streams near the stratosphere boundary, i.e., not only in the lower- to middle-tropospheric parts of the turbulent plumes (e.g. Hladil et al., 2008). Hence, although it is generally clear that the sediments closest to the emission areas contain the highest proportions of large, heavy or bulky particles, the dispersal of large grains must be much broader than formerly assumed. We aim at furnishing at least rough outlines of this mass added to the dust budgets and inputs.

The necessity to summarize and reappraise the problem of the 'recent dust efficiency in geological terms of view' seems to be evident. Among other reasons, it relates also to the fact that the concepts, methodologies and the terminology in atmospheric aerosol research disciplines rather follow the requirements of the pollution monitoring (and modelling) than the explanation of geological aspects of the atmospheric transport of particulate matter and its sedimentation. In this context, data on the total amounts of deposited dust have been particularly biased by omission of the added mass from the bulky but not uncommon mineral-lithic grains.

Similar problems are associated with many biogenic components (pollen, spores and microdetritus) due to their small specific density, large sizes, complex shapes and mechanical and aerodynamic characteristics. Data on such components are rather provided by pollen-allergy networks. A common occurrence of aggregated dust grains often escapes the air pollution monitoring scrutiny, being almost exclusively documented by means of the dust sediment analytics (e.g. Diaz-Hernandez & Parraga, 2008). In many aspects, the present atmospheric dust budget modelling on one side and the measurement and analysis of the real dust sediments (or inputs to sediments) on the other side show very little overlap.

In this context, we must observe that the monitoring facilities for measurement of the real dust sediment remain underdeveloped. The network of monitored quadrates for airborne sediment studies surely deserves a future extension both in the area covered and in the number of stations.

Major source components related to this attempt to reappraise the amounts of natural dust in the modern world are as follows: dust from terrestrially (on-land) weathered rocks (section 2), volcanic ash (3), biogenic and organic dust (4), dust from vegetation fires (5), interplanetary dust together with meteorite- and impact-related components, and (6) little contribution to the deposition of minuscule solid particulate phases by secondary aerosols (7).

2. Dust from weathered rocks

As generally accepted, the emitted, transported/dispersed and deposited volumes related to this dust category belong among the main components (Zoller & Faust, 2009). This material is 'loess', in the broadest sense of this term. It represents wind-blown silt together with lesser and variable amounts of fine sand and clay. The present-day atmospheric, marine and geological processes have a full respect to this loess phenomenon, and sufficient data were published and analysed (thousands of papers; e.g. Zender et al., 2004; Tegen & Schepanski, 2009). For the future, this modern world experience needs to be transferred to conditions of the past geological eras, where the number of studies is small (e.g. Palaeozoic with only several tens of papers; Elrick & Hinnov, 1996; Cecil, 2004; Soreghan et al., 2008; Hladil et al., 2009; Koptikova et al., this volume; Machado et al., this volume).

The dust from weathered rocks consists mostly of quartz which contains inclusions of accessory minerals – e.g., amphiboles, chlorites, epidotes, garnets, iron oxides such as hematite, lepidocrocite, goethite or limonite mixtures, three polymorphs of TiO_2 (anatase, brookite and rutile) from Ti-rich metamorphosed rocks, weathered feldspars and micas, or more complex precipitates which occur in grain fractures or in overgrowths. Other typical components are clay minerals and micas, and especially iron oxides which are typical for the weathering products derived from subaerially exposed surfaces. Natural halides, sulphates, carbonates and nitrates are also present (e.g. Tsigaridis et al., 2006). Of course, a wide range of common accessory mineral grains also occur, in spite of the fact that the mean residence times for heavy minerals are often relatively short and connected to low-altitude trajectories.

These quartz-rich natural weathering-product emissions, dispersals and depositions of the most common types gradually pass into 'loess types in the narrower sense of this term'. This loess usually displays elevated contents of feldspars, mica and other minerals, including also small lithic clasts. This loess is often derived from the floodplains of glacial braided rivers or from semiarid areas in the mountain regions or desertified steppes. This material is voluminous in its proximal deposition areas but has also a considerable potential to spread worldwide (e.g. Pye, 1995; Yoshinaga, 1998). The effects of chemical weathering on the composition of these 'mountain' or 'colder' loess particulates are relatively slight; therefore, both the proximal and medium distal deposits fingerprint well the source areas (e.g. Yang et al., 2009; Lisa et al., 2009). Potassium feldspars are contained even in some of the more distal deposits, and Rb/Sr and Na/K ratios may serve as paleoclimate indicators for the originally emitted or accumulated loess material (Chen et al., 1999; Chen et al., 2008).

If we consider several most recently published calculations on the mineral dust budgets (e.g. Luo et al., 2003; Tegen et al., 2004; Zender et al., 2004; Strier et al., 2006), after the possible corrections for not natural components, the present-day annual emission (E) for

these materials must be about 2 Pg (2×10^{15} g). Looking farther to the geological past, including the glacial periods, the E-related estimates for the whole Quaternary is ca. 3 Pg (Andersen & Ditlevsen, 1998). It is worth mentioning that even the most serious estimates for the present-day natural E values fluctuate greatly (±1 Pg; e.g. Tegen & Schepanski, 2009).

However, the state-of-the-art in the disciplines dealing with this sort of mineral dust seems to be still mostly constrained by adhering to the values PM_{10}, because most sampler and remote sensing methods have been adapted to the above mentioned particle class, which may considerably underestimate the abundance and mass of mineral-lithic particles (Hladil et al., 2008). Particularly the last cited case of the Ukrainian dust (spread over the Europe to the northern Atlantic Ocean) provided unquestionable evidence that even giant grains are transported over distances of thousands of km (d_e of 200–250 µm). This, understandably, goes behind the standard models of particle-size distributions (role of jet streams, Section 1). Moreover, we must consider that such a d_e-250-µm grain has roughly the same mass as ca. 1.6×10^4 of d_e-10-µm or 1.6×10^7 of d_e-1-µm particles together. This means, expressed in simple practical terms, that only a few giant grains (e.g., spaced up to hundreds of metres from one another in the air, or several centimetres in the dust sediment) may provide an added mass that is often greater than the mass of the fine (normal) dust particles.

This case of dust from SE Europe suggests a difference of at least one order of magnitude (Hladil et al., 2008) between the mass of the transported particulate matter as indicated by means of measured and calculated PM_{10} or TSP (atmospheric sciences; Birmili et al., 2008) and the transported mass as derived from the measured dust sediment (geology; Hladil et al., 2008). This difference means that the estimates based on real sediment are higher than those achieved by present techniques of air pollution monitoring, e.g., the air stream sampling and infrared-based satellite detection of dust. Furthermore, we averaged these data with all other data measured (e.g., the 'CZ Dust Sediment Data Store', own data on the 2009 Saharan dust and Alaskan volcanic ash events, as yet unpublished). Considering these materials, the ratio between the sedimentologically indicated 'real' mass of the transported dust (*RMD*) and the mass indicated by means of PM_{10} indicates a difference of ca. ×5, and *RMD* compared with *TSP* of ca. ×3. These differences in the calculated amounts of the total mass of the transported and gradually deposited material are primarily explained as an effect of added large-particle mass (Hladil et al.,2008.).

Moreover, wide experience with particles delivered across the world by high-altitude jet streams shows a yet another problem – the presence of large aggregated grains. These large grains with d_e of about 25–50 µm completely originate during the atmospheric transport. Not all are described, but at least one classical example can be mentioned – the particles which have been agglutinated in the air and clouds (iberulites; Diaz-Hernandez & Parraga, 2008). These aggregated grains are still only partly

understood, but may potentially lead to further increase in the estimated mass due to large particles.

The above suggested differences in the total mass of transported 'loess' material (i.e., at least ×3) must also relate to the emissions (E), atmospheric load and the 'final' sedimentary inputs. These relationships were not modelled in detail. Since then, we may speculate that the mean annual emission values of ca. 2 Pg (Tegen & Schepanski, 2009) should be corrected to perhaps ca. 6 Pg (6×10^{15} g) for the whole spectrum of the 'dust', i.e., from very fine particles to the 'uppermost possible limits' of d_e of ca. 250 µm (our estimates).

However, the global generalization for the inter-annual, decadal and millennial dust inputs to sediments is even much more complicated. This is due to the definition of the turnover (mean residence) in the atmosphere, mean atmospheric dust burden, airborne dust sedimentation fluxes and, mainly, the factors which relate to the recycling and 'final' inputs to sediments. Recycling means that the dust already deposited is emitted again and again. However, this is not of the essence in this case. The main role must be ascribed to precipitation, run-off, washing away the dust and its incorporation and chemical-mineralogical transformation in the fluvial muds. This process still awaits global mapping and quantification. However, recycling and alteration of the airborne terrestrially weathered dust has tentatively been assessed as important for the marine systems, where only $^{1}/_{5}$ of the E mass on the Earth may end as the 'final' input to marine sediments (Hladil et al., 2003; 2009). When this mass reduction coefficient ($^{1}/_{5}$) is included in the solution based on the 'geologically-sedimentologically' enhanced E (6 Pg; see above), the desired value can be ca. 1.2×10^{15} g/yr. Dividing this possible input value by the area of oceans and seas (3.6×10^{14} m^2), we can obtain a specific input of about 3.3 g/m^2/yr.

As mentioned above, no reliable data exist to summarize the 'final input' reduction coefficient for terrestrial deposits at a global scale, i.e., including the mass reduction coefficient which may characterize the proportions of the recycled dust deposits (often washed-away and transformed to river muds) and the 'finally' preserved dust deposits (either as 'loess' accumulations or infiltrated into porous soils and rocks; Derry & Chadwick, 2007). Therefore, we can only arbitrarily choose a hypothesis that the ideal specific input of 3.3 g/m^2/yr (3.3 t/m^2/Myr) may also be valid for the whole Earth's surface. These aspects determine the challenge for a further elaboration of these estimates in the future.

This tentatively proposed value is, therefore, only an ideal (and very uncertain) mean value. Moreover, the area-specific values may differ much from this mean, by X % or, in extreme, XX %. Some of the geographic areas have giant aeolian fluxes directly from the arid coastal areas, XX g/m^2/yr (e.g., near W African coasts; Senegal, Mauritania; Itambi et al., 2009), and some areas are really starving for only minimum inputs of 0.X g/m^2/yr or even less (e.g. some polar areas; Delmonte et al., 2008).

However, major areas with reefs, carbonate platforms and their slopes to the ocean usually belong to well fed geographical zones. These areas often show a good teleconnection with the main emission sources (arid areas and trade winds; e.g. Prospero & Lamb, 2003; Garrison et al., 2003; Kiessling, 2005). The predominantly shallow-marine carbonate factories are relatively good traps for the dust, and the areas of carbonate sedimentation which lie along the common trajectories and lateral spreading of the dust plumes show the primary dust inputs of several $0.X$ to XX g/m²/yr today. Marine carbonate surfaces in really dusty conditions (e.g. in the Red Sea or Persian Gulf) show the primary inputs of about 5 to 50 g/m²/yr, and these values are similar to many examples of 'medium dust-fed' limestones of the past (Hladil et al., 2009). Specifically for these conditions, the primary inputs are usually reduced to ¼ due to washing-out and dissolution (i.e., the 'final' input is ca. 1.25–12.5 g/m²/yr).

This similarity allows us to point out some quantities related to limestones of the geological past. When using the latter specific input values, it can be calculated (Hladil et al., 2003; 2009) that the limestones with 20 m/Myr long-term 'rate of sedimentation' (rather non-sedimentation; Adey, 1978) contain 2.5–20 wt. % of the non-carbonate dust impurity component. This shows how strongly the dust component can contribute to the limestone compositions. The proportions of the dust components can be determined using the REE distribution patterns (Hladil et al., 2006).

With respect to iron and possible magnetic signal of the dust from the terrestrially weathered rocks, the importance of the desert 'loess' material and fine fractions rich in Fe is evident (Bosak et al., 2002; Prospero & Lamb, 2003). In coarser varieties of this material, the paramagnetic detrital silicate minerals which contain iron and mainly detrital/diagenetic hematites and magnetites are the main carriers of the magnetic properties. In this context, also the vegetation cover and bacterial activity may have an effect on the primary and secondary mineralogical and magnetic properties of these iron-rich weathering dust components (e.g. Tribovillard et al., 2002; Hladil et al., 2004; Tomescu et al., 2006).

3. Volcanic ash

Compared to emissions of weathering related particulates, the volcanic ash events are much more episodic. The amounts of the relevant particulate load extremely vary in the atmosphere, both for the main plumes and weeks or even months prolonged secondary dispersals. The latter is possible due to spreading around the world by high-altitude winds (Foch et al., 2009; Hladil et al., 2008; Anonymous, 2010).

Considering the averages related to recent volcanic activity on decadal or millennial scales we can assume, considering thousands of available partial reports, that primary particulates and secondary aerosols (including condensed, crystallized and aggregated particles) may correspond, for the Holocene, to an average and very approximately set yearly emission of about 8 Pg (8×10^{15}

g) (our estimates). However, according to the great mass of coarse ash deposited in proximal areas around the volcanoes and along primary volcanic clouds, our very rough and preliminary estimates suggest that only about $^1/_{80}$ of the originally erupted mass have capability to survive in the atmosphere for days or weeks. It is due to the fact that the relatively dense and rapidly energy-dissipating suspensions of volcanic fly ash, particularly those with great amounts of fine-sand-sized and larger grains, have a tendency to fall down very fast, even scavenging the fine particles and early stage volcanic aerosol. Hence, this amount must be much reduced for the case of the volcanic dust teleconnection over the large distances (ca. 1×10^{14} g). The chance of these distal volcanic ash dispersals to be delivered to basins in the world seas is relatively good because the majority of volcanoes which are powerful dust emitters are rimming the continents or ocean arcs, where the explosive eruptions relate to high viscosity and highly gaseous/vaporous andesitic, dacitic and rhyolitic magmas. Particularly the lithocrystalloclastic and glassy particles (see below) are slowly altered in marine environments, although the composition variations may, perhaps, indicate ca. 25 % loss of this material (Weber et al., 1996). If we will suppose about $^3/_4$ of this distal material that is really delivered to seas and has a potential of 'final' embedding in sediments, the calculated specific input to sediments in seas may correspond to ca. 0.3 g/m²/yr. It is more difficult to find the similar estimates for the terrestrial sediments. The mean specific inputs would be much enhanced due to very proximal ash fall but also reduced by soil weathering and washing-away (and transformation of this material). Thus this value for the marine sediments can be only arbitrarily chosen to represent also the global specific input to sediments (the error from this may be ca. ±50 %, but less than from all other corrections used above). Certainly, this series of assumptions or almost intuitively balanced mean values can certainly be considered as very approximate and can be subject for many and much more sophisticated improvements in the future.

The quantitative distributions with time show stochastic behaviour, and usually display well spaced but strong changes in amplitudes. It corresponds to the fact that there are also very strong volcanic explosive events with up to 20–30 km height of the vertical uplift of the dust into the stratosphere. Counts of these events can be made on the centennial or millennial scales. This can be exemplified by the Pinatubo-1991 or Krakatoa-1883 explosive eruptions with $\leq 1 \times 10^{16}$ g of extruded particulate mass, i.e., with the eruption magnitude (M) at ca. M6–7 or volcanic explosivity index (VEI) at ca. 5–6 (see Mason et al., 2004 for relationships between M and VEI and emitted particulates and deposits). The coincidental occurrence of several M6–7 events has certainly a good potential to disturb the time-averaged values of the inputs to sediments, as it depends mainly on the erupted mass and height of the turbulent column containing the volcanic ash particulates.

With an excursion to the past (e.g. interval of 100 kyr or more), we must consider also the supereruptions of about

1×10^{18} g particulate production potential (M8–9 magnitude), e.g. the Indonesian Toba (Mason et al., 2004), a supereruption which is possibly 74,000 of years old. Such supereruptions may enhance the very long-term estimates of the inputs to sediments by 'unbelievable' two or three orders of magnitude. However, the recurrence of such supereruptions should not be overestimated, because the thorough attempts to express the recurrence of such superevents in the terms of probabilistics are referring to a value of ca. 1 Myr (Mason et al., 2004).

Petrologically, the major part of this material is the volcanic ash of tephra compositions and fabrics. The SiO_2-rich glassy matter predominates, but Ca-rich plagioclases are also present, commonly together with amphiboles, pyroxenes, micas, chlorites and variety of accessory minerals. In total-alkali-silica diagrams, the compositions of tephra ash are often between the typical fields of andesite (or dacite, trachyte) and rhyolites, even if the mafic minerals are present. The melt-rich aggregates possess lots of exsolutions and transition crystal lattices. An obvious excess of Na and S in these natural particulates is typical compared to the dust from the weathered rocks. The geochemical difference is usually well recognizable, because the non-volcanic natural mineral dust is richer in K and Th, and has the REE and trace element compositions which are close to the UCC or PAAS standard values (McLennan, 2001).

The volcanic ash REE compositions show positive Eu anomalies; LREE are lower than HREE, and the total REE concentrations are remarkably low. The 'primary volcanic mineral dust' contains also many crystals of iron or iron–titanium oxides (hematite, maghemite, magnetite, titanomagnetite and ilmenite). However, the contents of these highly magnetic minerals vary considerably in number of particles, and the other differentiation pathways (sorting of the fine particles during the aeolian process, recycling, weathering and transformation to other materials) cause that contents of these minerals may not necessarily be very high. Moreover, the details of iron cycle related to fine volcanic ash and the fertilising potential of tephra for the marine environments are not completely understood yet (Duggen et al., 2010). The thicker layers of deposited volcanic ash are gradually altered to bentonite, which is a clay based material that derived from original glassy materials. The diagenetic conversions to bentonites are also described from the Holocene, i.e., not only from the past periods (Naish et al., 1993; Webb et al., 2008).

Although the tephra-related materials may preserve magnetite crystals, it is worth of mention that the total Fe-concentrations in volcanic ash are often a half or equal compared with the common desert loess. It is generally accepted and used that volcanic ash inputs to sediments can be easily detected due to their increased magnetic signal, but it evidently not the case of ultra fine ash fractions below 4 µm (8 Φ), where this signal is similar to that of the desert loess. Only the coarser ash fractions (6 to 2 Φ) may have a very strong signal, which differs from the background by one, two or several orders of magnitude.

This difference is used for the detection of tephras in oceanic sediments (Touchard & Rochette, 2004; Gattacceca et al., 2004) or in lacustrine sediments adjacent to volcanic arcs (e.g. Schiff et al., 2008).

If concluded, the particulates related to the volcanic ash sedimentation have the characteristics which may vary much more than traditionally assumed. We must also consider the question of event- and background-related delivery of volcanic particles. In spite of these uncertainty difficulties in many parameters, the mean specific input of this volcanic component of the present natural dust to sediments can be estimated (≈ 0.3 g/m²/yr). This input is more than ten times lower compared to the normal weathering products (the previous section; 3.3 g/m²/yr). Before the sophisticated monitoring of the volcanic activity worldwide, the 20th century estimates about the mean sedimentary inputs to sediments were much more modest. The significance of the volcanic input to sediments and soils seems to be more and more evaluated in the last years (e.g. Muhs & Budahn, 2009).

4. Biogenic and organic dust

These components, seen in the hierarchy of scales from large particles to minute organic phases or even viruses, significantly contribute to the composition of dust. The assessments across different forms of this biotic and organic matter are still unsettled and they range in wide intervals. It is, as ever, much related to the data available for each of the category and particularly size- and composition-related preferences as applied for the individual studies. For instance, Jaenicke (2005), using meteorological aerosol-related techniques, approximated the annual emissions of cellular material and proteins as 1×10^{15} g (Pg).

Also the selection of techniques and subjects has a great effect on the results obtained, as exemplified by pollen. In present conditions, the pollen grains represent, manifestly, a very substantial part of this material. The numerous data on pollen concentrations in air (e.g. Damialis et al., 2007; Yli-Panula et al., 2009) combined with those which describe the annual deposition fluxes (e.g. Hicks, 1999; Autio & Hicks, 2004) allow us to make some computational attempts to quantify this component. The most complicated moments relate to specific pollen dispersion as well as area or latitudinal characteristics. It is because of the fact that we have, besides the richest areas with visible input to sediments, also the starving areas which are almost exclusively fed by high-altitude pollen transport. The results of any models are dramatically fluctuating according to number of factors which are taken into consideration (or omitted). Although much intuitively, we may prefer one of the solutions which is indicative of the total annual pollen deposition on the Earth about 8×10^{14} g (dry pollen matter). Taking into consideration this value, the ideal primary (not final) depositional inputs may be ca. 1.6 g/m²/yr. The latter value is not more than a quotient between the possible total pollen mass and Earth surface. It is worth of mention that the situation about such estimates for the pollen budgets is still quite unsettled

and the error usually much exceeds the value. On the other hand, the overall experience with the dust sediments which were analyzed in detail confirms that the volume proportions of light pollen grains are often much higher compared with the mineral components. The traditional 'belief' that this light pollen material is only a negligible part of the dust is incorrect, especially if it is seen as the volume proportion. This belief comes from the analyses of the massive (eye-catching) mineral dust deposition events only, and not regards the global annual budgets.

There is also one other possibility to verify roughly these estimates on the primary pollen budgets. We may use a generally accepted estimate about the terrestrial yearly primary production (1.1×10^{17} g of carbon; Gulledge & Pidwirny, 2009), i.e., that photosynthesis by terrestrial plants moves about 110 Pg/yr of carbon from the atmosphere to the biota. Expressed per 1 m^2 of land, this means ca. 750 g C/m^2/yr. By taking the carbon content as ca. ½ of dry biomass and dry biomass as ca. ¼ of the living plant biomass, it is ca. 6×10^3 g of the 'fresh' plant biomass/m^2/yr. The value for pollen and megaspore producing plants must, most likely, be reduced by algae, cyanobacteria etc., but it is likely to remain high (first of $X \times 10^3$ g/m^2/yr). The difference by ca. 3 (or 3–4?) orders of magnitude between the 'fresh' yearly biomass increment and produced pollen or megaspores is, most likely, within the expected limits.

Not only the pollen grains, but also lots of other biogenic particles occur in the airborne dust. These belong to fungal spores, hyphae and plant microdebris that imply an added mass of a few to several tens (or even hundreds?) of Tg (Elbert et al., 2007; Winiwarter et al., 2009). Also bacteria, algal cysts, viruses and other objects of nm–µm sizes are common in the air and contribute to inputs of organic matter to sediments. However, the small d_e determines their smallest contribution to the mass budgets and specific inputs. Besides the above mentioned plethora of objects also animal material is involved. There is common evidence of little amounts of insect-exoskeleton debris, bird-feather barbulae, mites, skin cells or animal hairs. It is also surprising how many cyanobacteria and diatoms are occasionally involved in the present-day dust. These often occur with portions of African dust that comes from dry lakes.

As a rarity among objects transported in air, also unusually big biotic objects can occasionally be sucked up by atmospheric vortices, or other turbulences (cf. reports on raining rags of aquatic biofilms, fishes, or terrestrial plants and animals). More than 50 regional to inter-regional events were recorded in the 20th century, and some took to surprise also the ancient civilizations. The maximum wind speeds measured at the ground level showed gusts 100–140 m/s, and seem to be far more frequent than formerly believed (e.g. Buchan et al., 1999: 113 m/s, 407 km/h, with the 1996 Tropical Cyclone Olivia). The data not approved by World Meteorological Organization may indicate even the gusts of 500 km/h speed (?). Such an air stream has capability to move even the $X \times 10^6$ to $X \times 10^7$ g objects (e.g. giant trees). Of course, these extreme transport events, in present climates, have only a potential to 'throw' such objects over distances of a few km, not more.

The emission of terpens, essential oil related aromatic and other organic compounds from coniferous forests, semi-desert vegetation or many other vegetation formations is, however, a considerable phenomenon. Some examples are known under the common names as 'blue smoke' or 'natural smog'. These volatile compounds released by plants are aerosol particles of submicrometre size but they react with ozone, and the secondary compounds with furandiones, aldehydes and organic acids (from hydrocarbons) condensate and accrete on inorganic aerosol particles. In this way, they can contribute to mass budgets of airborn particulates. In extreme, they may be transformed to 'wax' particles or coatings. The recent measurements (e.g. Christensen et al., 2000) indicate the specific fluxes which, recalculated to surface and time of these and similar vegetation sources globally, may be indicative of a global annual emission of about 2.5×10^{12} g (Tg). The final inputs were not assessed but are rather weak subcomponents.

With an evident predominance of pollen and with addition of all other realizable and includable additional components, the annual amounts of the deposited organic aerosol/dust may, very approximately, be estimated around 1.2×10^{15} g (Pg). It implies an uncorrected value of the possible mean 'primary' input ca. 2.4 g/m^2/yr. Of course, this is only an ideal mean value, because many areas of the world give an evidence about much higher inputs of this matter into sediments, and other, e.g. the Antarctic ice, show very low pollen inputs. However, and it must particularly be considered for the marine basins, an intense transformation of airborn organic particulates precedes their final embedding into sediments. The major problems facing the next interpretation steps are twofold. First, the global budgets related to microbially mediated decay of this organic matter are almost unknown and may considerably vary. Secondly, the same is valid for the food chain. There is lack of solid evidence base to determine the reduction coefficient, so that a very arbitrary (artificial) value was selected, perhaps $^1/_5$. Using this coefficient, we may speculate about the final inputs to (marine) sediments ca. 0.5 g/m^2/yr.

In this specific context, the role of iron was never discussed. As the pollen grains and spores predominate in this airborn particulate matter, several remarks about them are made here. The pollen grains are short-lived, from few days to several hours. With long transport the interior dries up and changes to necrotic matter. In any case, this material is dominated by C, (O and H), and N, P, S. E.g., the latter concentrations in ambrosia dry/moist pollen are usually about 4.7, 0.7, 0.6%, respectively. Microelement and trace element representation is rich and concentrations are relatively high. The concentrations of iron, as commonly determined on fresh-harvested pollen assemblages, are primarily around 10 mg/kg (e.g. Nanda et al., 2003); for pollen ash (0.2% w/w) it is 0.5% of iron, together with trace amounts of zinc, manganese, titanium

and molybdenum. However, the data made on older, altered pollen populations suggest that Fe-content can be increased by one order of magnitude, reaching few percent concentration values. The highest sedimentary inputs of iron mediated by pollen are around $0.X$ $g/m^2/yr$ (e.g. the case of pine pollen in Wisconsin; Doskey & Ugoagwu, 1989). The mean values with this 'final' Fe-input to sediment are, of course, much lower, perhaps about 0.02 $g/m^2/yr$.

There is also little known about the early diagenetic alteration of pollen and spores on the earth surface before they were emitted and, particularly, during their weeks-and-months long transport in air. Although a large literature about selective destruction of exines and their mineral coatings exist (e.g. Rameil et al., 2000), a lot of aspects remain undescribed. The precipitation of gypsum, carbonate, limonite and organic salts leads to origin of coatings, cements or partially permineralized structures. Another typical feature of pollen and spore grains displayed by them, is a trend to stick one to another, adhering to smectite particles of nm-μm sizes, or participation in origin of complex aggregated grains (together with other dust components). With respect to iron, even the bacterial decomposition of these grains on the ground, i.e. 'blackening of pollen', is often connected with origin of magnetite-maghemite or pyrrhotite-pyrite crystallites. Moreover, these ultrafine crystallites may serve as nucleation centres for the origin of Fe-sulphide framboids in the buried pollen grains. This is even further complicated by the changes to this material at the moment of the input to shallow-water limestones or after. This is because of the fact that this input often intersects with oxidation of organics and dispersion (reconcentration) of iron (e.g. Bosak et al., 2002), so that this material can also be nearly obliterated from the record.

5. Wildfires – dust from vegetation fires

The fires that periodically rage in the forest, savannah, peat and other vegetation covers show a certain similarity to the industrial combustion of wood or coal, particularly with the increasing magnitude of the forest fires. The latter can resemble 'power plant combustors'. On the other hand, the wildfires running across sparse dry vegetation have only slight effect on transformation of combustion products from the biomass as well as added mineral particles. The present experience that almost every vegetation fires are caused by some artificial (human) activity is substantiated only in part. It is very interesting that frequencies of wildfires in past are not very different from those of recent periods, i.e. a few or several years for the vegetation covers in relatively dry climatic conditions (Dodson et al., 2005). The wildfires occurred as early as with the expansion of the rhyniophytoid flora on the continents, and were very common with the Carboniferous and Permian 'high-oxygen' atmosphere (Scott & Glasspool, 2006). The fires are relatively well indicated also in the Mesozoic (Finkelstein et al., 2005; Marynowski & Simoneit, 2009).

There are several categories of wildfire markers in stratigraphic geology. Inertinite charcoal particles, carbon coenospheres, also silicate and silicate-magnetite spherules or mullite, melilite and complex cinder-shaped objects in micrometre scales are primarily important (Harvey et al., 2008). Other category relates to chemical markers where we can found internal lipid components in char, or with great magnitude of fires (and burning of some secondary hydrocarbons, not only living or dry organic tissues), the detection of pyrosynthetic polycyclic aromatic hydrocarbons as coronene, benzoperylenes or benzopyrenes may be a meaningful measure for this group of dust materials (e.g. Arinobu et al., 1999).

Concerning the recent data, about three thousands of the papers and conference releases have been focused to wildfires and quantity of the emitted carbon or CO_2. The mean annual total for this carbon globally is generally accepted, and is about 4×10^{15} g (Pg). Almost ¼ originates in Africa (Lehsten et al., 2008). This global wildfire emissions of C can, e.g., be compared with the 2009 industrial gaseous CO_2 emissions (3.2×10^{16} g) which contain ca. 8.7×10^{15} g of C (difference only ×2.2). This provides unequivocal evidence that wildfires belong to several strongest emitters. In comparison with the $PM_{2.5}$ and PM_{10} aerosol categories which are frequently documented (and shows a 'suspect' degree of heterogeneity, e.g. Wua et al., 2006), the PM_{25} or PM_{50} equivalents in lithic-mineral particles, and even larger grains, are less documented and lots of these particles remain to be studied.

With respect to the strong turbulences near the ground, high thermal lifting and great extent of these processes, we can assume that the mass of wildfire-related coarse particulates which is broadly dispersed and ends, after all recycling and reduction of the mass, as a 'final' input to sediments may represent slightly less than $^1/_{10}$ of the values which are assigned to the terrestrially weathered rocks (Section. 2). Hence, this input value must be in magnitude of $X \times 10^{14}$ g, and, perhaps, close to ca. 0.3 $g/m^2/yr$.

The uncertainty of this value is about ± 40 %. This is partly due to poorly elaborated quantitative data on the large sized C-rich particles, i.e., in the categories of coenospheres, charcoals, 'black carbon cinders', aggregated soot, etc. In addition, not all these particles can be unequivocally assigned according to their origin, although some of the applied approaches seem to be very effective, e.g., using the signatures of polycyclic aromatic hydrocarbons which are indicative of various wood-, coal- or 'oil'-derived particles – Oros et al., 2002; Sykorova et al., 2009). However, a great deal of this uncertainty is associated with the mineral particles which are not dominated by C. This relates to the above mentioned 'cinders' and hollowed glassy spherules of high temperature fires, but especially the emitted soil particles (mineral-lithic clasts, smectite-illite-mica flakes, lots minuscule clots, aggregates and peloids, etc.). In this context, also carbonate cystoliths and silica phytoliths (also trichomes) cannot be neglected. Such complicatedly shaped (aerodynamically light) mineral bodies from the plant tissues are several tens of μm large. They have a

good potential to contribute the airborn particulates (e.g. Rovner, 1971; Cary et al., 2005), and this must be much enhanced with the wildfire events.

As far as iron is concerned, the intense forest fires may cause the near-ground T of 750–800 °C (Saharjo, 1999), and T_{max} slightly over 900 °C is expected (NFPA, 2010). With high conditions of volatiles, these high temperature vegetation fires may also produce highly magnetic mineral phases, as a parallel to the power plant particle emission. With the transition of the wildfire dust input into sediment, the wildfire soot with increased concentrations of nutrients may increase microbial activity which often shows a potential to further enhance the concentrations of iron.

Although marginally, we may also mention that the wildfire soot can also lead to melting of polar glaciers (Kim et al., 2005) and this can take effect on exposure of rock surface, weathering and extension of 'lower latitude loess' (Zoller & Faust, 2009), a process which may further complicate the expression of 'cooler–warmer climatic and higher–lower sea level / detrital input patterns' (Ellwood et al., 2000).

6. Cosmic dust and related materials

The interplanetary dust particles (IDPs) provide the major amounts of this material which accretes to the Earth. The 'PM$_{10}$ size equivalents' coming from the dust of the main belt of asteroids have often low geocentric velocities of few km/s. Such 'slow' IDPs are readily attracted to the planet but are accelerated by the gravity force. Deceleration comes at an altitude of ~ 90 km. These well preserved (or least damaged) particles survive because of their delicate dimensions. The highest surface/volume ratio of these fine particles causes that heat is sufficiently radiated away, so that effects of melting and evaporation are slight or negligible. Then the IDPs settle through atmosphere and cumulate in the lowermost stratosphere, at about 20 km altitude. Here, the concentration of these IDPs is much greater than before entering atmosphere, possibly by six orders of magnitude. The chondritic and often hydrated IDPs have slightly different compositions in comparison with meteorites. They contain delicate crystals, e.g. enstatite and forsterite, which are connected by fine-crystalline calcaluminous mass. The crystals are arranged as a framework and form micro-to-nanoporous aggregates (e.g. Flynn, 1994, 1999; Bradley, 2003). Including all related IDPs, from a few µm dimensions to about 50 µm size (i.e., a rough equivalent to atmospheric TSPs limited by the apparent 100 µm size), the recent estimates are kept at about 4×10^{10} g of such a material which may reach the very high atmosphere. However, the bigger dust particles are usually much damaged by melting and evaporation. Therefore, only the remaining amount of this material (a mix of tiny but undamaged and formerly bigger but much transformed particulates) reach the concentration zone in the lowest stratosphere. Then the IDPs travel along the tropopause, fall down and again travel in the troposphere. In the troposphere, these particles are mixed together with other dust components, and follow their further atmospheric development, settling and, finally, incorporation into sediments on the ground or in the seas. The relevant primary (not 'final') input to sediments is possibly not exceeding the value of ca. 8×10^{-5} g/m^2/yr (our estimates).

The coarse IDPs (0.1–1 mm) are pieces of meteoritic material which shows blazing trajectories. They belong to class of smallest but quite common meteors which enter with ablation, melting, and bigger can also explode. This forms upper stratospheric 'meteoric smoke', a source of secondary particles of nm–µm sizes (Curtius et al., 2005). Some of them survive as micrometeorites, the small glassy objects that reach the Earth surface, but the remaining mass contributes to oxide-oxyhydroxide matter of high atmosphere which is, after a long time and in lower altitudes, also inbuilt into more complex structures of the aerosols. This class of the external dust material may represent, very roughly, 3×10^{10} g, and the primary input to sediment is possibly 5–6 $\times 10^{-5}$ g/m^2/yr (our estimates).

The objects of X mm – X m sizes are seen for the night sky observers as large meteors. The meteoroids causing these optical phenomena are the sand to boulder sized interplanetary fragments which usually enter the atmosphere with the velocity of 10–60 km/s. In spite of huge number of observable small meteors during a year, only tens of thousands of meteorites with mass over 10 g can end their 'fiery' fall through the Earth's atmosphere hitting the sea or land surface. The annual primary input of the meteoroids into the atmosphere is estimated to be equal to 1.6×10^{11} g (Curtius et al., 2005). However, only $^1/_5$ of this mass is believed to directly survive in the form of ablated delicate fragments, spray of melt droplets, and relict objects – meteorites (our very rough estimates based on current discussion sources on internet). The $^4/_5$ of primary mass is transformed by hot, friction-driven ablation, where most of the solid mater evaporates with diffusion from the meteoroid's train into ambient air. A part of this ionized mass (but not all) may gradually be collected by aerosols. The relevant specific input of the material related to these 'drastically transformed' meteoroids to the sediment, being corrected down due to the ions which cannot be recollected by secondary particulate condensates, would be, perhaps, about 2×10^{-4} g/m^2/yr.

The large bolide and cometary impacts are rather a separate problem. In this case, the ejecta and settled condensates play the major role. E.g., the Chicxulub impact intensity class may be characterized using the following values (Artmieva & Morgan 2007): the total volume of distal ejecta materials, dust fall particulates and condensates ca. 2×10^{12} m^3; mass of this material ca. 5.4×10^{18} g; ideal global mean input ca. 1×10^4 g/m^2/event; outsides the ring of the crater ejecta ca. 5×10^3 g/m^2/event. These 'super collision' impact events with airborn material deposition magnitude at X kg/m^2 are extremely rare, and it is problematic to integrate them within the short-term estimates of the airborne material budgets. However, there exist a few of well preserved Quaternary craters of ca. km diameters (PASSC, 2010)

and some events possibly occurred also in the Holocene, e.g., Chiemgau?, Araona?, Tunguska explosion? The overall 'dust productivity' of several possible Holocene events is generally unknown and cannot be ascertained to the necessary degree of certainty (not involved here).

If the latter is neglected, the sum of the primary inputs is equal to IDPs + micrometeorites + meteor-ablation debris and meteorite related items, i.e., $8 \times 10^{-5} + 5 \times 10^{-5} + 2 \times 10^{-4} \approx 3.3 \times 10^{-4}$ g/m²/yr. The mass reducing corrections to obtain the 'final' inputs to marine sediments or the sediments on the whole Earth's surface are very problematic. If the same approach as that described above for the volcanogenic material is used (but without any massive local accumulations) and, e.g., the mass reducing coefficient ½ is used, then a tentative value is 1.65×10^{-4} g/m²/yr, i.e. less than ca. 0.0002 g/m²/yr. This contribution of cosmogenic dust is, therefore, by four orders of magnitude smaller than that of the dust from the on-land weathered rocks – compare the section 2.

The enhanced content of iron is typical for this minor component of the airborn particulates, but the visible concentrations of these materials are rare in geological record. The concentration of cosmogenic materials requires the extreme conditions of 'sheltered lag surfaces', extremely low sedimentary rates in calm aqueous environment or 'ice and snow traps' (Rochette et al., 2008; Suavet et al., 2009). In geological past, especially with the rare impact events, the inputs to sediments may abruptly be enhanced, displaying also the well detectable concentrations of iron and the impact related markers in elemental and isotope chemistry.

7. Aerosols in narrower sense

Hence, the atmospheric aerosols in this broader sense are suspensions of solid or liquid particles ('droplets'), or very complex objects retaining the solid-fluid-gas mixtures. With implementation of advanced technical applications in the environmental monitoring systems, meteorology and aerosol microphysics, the term aerosol gained a broad meaning where the geological materials are 'weakly distinguished' according to their nature, shapes, structures and compositions. However, the practice which is more consistent with the differentiation and understanding of all these polygenetic materials is also followed to some extent (e.g. Kondratyev et al., 2006).

Technically, the atmospheric sciences separate the terms according to the origin of particles. There are primary and secondary aerosol particles. The primary aerosol particles are injected into the atmosphere as particles from the surface of the Earth (land and seas), e.g. mineral dust or biogenic dust, while the secondary aerosol formation includes the formation of particle mass out of the gas phase in combination with the mass gain of already existing minuscule particles (during the coagulation).

Naturally, the techniques to discern the individual components are facing many problems. This is particularly due to mixing, mingling and coating of all suspended particulate solids with the atmospheric moisture and drops (or ice crystals). The present overview is, however, specifically focused to the geologically relevant particulate solids. Due to the necessity to categorize these solids for the purposes of this study, the space for aerosols must be artificially reduced only to early-middle stages of secondary aerosols where precipitates or crystallites of the geologically important phases may occur. This means that these phases must be important for the sedimentary inputs to sediments. We categorize these 'aerosols in narrower sense' as an additional carrier of the finest detrital and precipitated solids which is beyond the ranges of other natural dust categories. The precipitation of solids in the secondary aerosol formations brings also many interesting problems, e.g., effects of saccharides (Caseiro et al., 2007).

The secondary aerosol formation is essentially linked to the conditions when gaseous SO_2 combines with water to form aqueous sulphuric acid. However, the natural situations related to the origin of this 'primitive droplet' haze are much more complex that the above mentioned case. The nucleation process starts with the Aitken particles (APs) that are sized below 0.1 μm and continues as a subsequent condensation growth to $PM_{2.5}$. The finest APs are the most numerous among all particles in the air but their total mass is very low (effect of very small d_e). The nucleation and condensation are mostly moisture and thermally or solar energy driven phenomena. For example, in the background coastal/marine areas, the nucleation starts with sub-nanometre iodine molecules, changing with ozone to I_2O_4, I_2O_5; also sulphate ions are trapped. With growth over the 5 nm size, the iodine oxides may be covered by readily condensable organic compounds, e.g. isoprenoids or methyltetrols. The further condensation and coagulation is accompanied by adding of chloride and sulphate compounds and other organic compounds – the particles grow to about 50 nm. The further growth is combined with oxidation of organics, interactions with acids, and mainly embedding of diversified solid/liquid and gaseous, additional components (Vaattovaara et al., 2006). These droplets are in equilibrium with ambient vapour in the air but may have a potentially high rate of evaporation and may often be supersaturated. Fluctuations and reorganizations of the inner structure are typical, and lead to precipitation, dissolution and reactions among the involved compounds. The complexity of possible precipitation of solids increases with the growth of droplets and the mineral or organic particles of nm sizes, collected from ambient air, may serve as crystallization seeds. In present conditions, e.g., the evaporite plains, salt lakes, also marshes, swamps, peat bogs, woodlands or moorlands are producing a lot of secondary aerosols and mixed formations. As already mentioned above, the secondary aerosol formations are not always a 'homogeneous haze'. They may occur also in a mingling of various dust components – e.g., with vaporous and particle loaded gaseous volcanic emissions. This means that lots of combinations exist.

From this reason, the assumptions about budgets are again a quite problematic task. We can only guess, trying

to find proportionality to S, N, C, organic and halide species in the air, how much of this geologically significant matter in the category of 'aerosols in narrower sense' may be delivered to sediments. The preliminary attempts to calculate the mass parameters for these added solids are indicative of the following values: ca. 1 Tg/yr today and not more than 0.1 Pg/yr with imaginable hotter and more humid or stormy settings of the Late Cenozoic. Transforming this to speculative calculations of the specific inputs to sediments, this may imply the values like (?) 0.002 to 0.2 (0.02) g/m^2/yr.

8. Modes and pathways of sedimentation

The modes of sedimentation are basically twofold: dry and wet (or combined) modes. The pathways of predominantly dry dust with its gravitational sinking are complicated by different layers of atmosphere that have different wind velocities, with intense deposition when perturbations in vertical structure occur followed by calm windows. On the other hand, the wet deposition (or wet and dry deposition) driven by precipitation from clouds, is very effective. For example, Japanese studies on the modes of depositions of East Asias dust suggest that about 70 % of the dust mass were brought down in the wet mode (Inomata et al., 2009).

Both extremes of typically dry and/or wet sedimentation occur, e.g., within the collapse stages of huge dust storm-generated tropospheric plumes or downpour from solid-phases-rich frontal, often cumulonimbus clouds, less commonly in the form of gently settling dry dust or dust with drizzle (Hladil et al., 2008). However, these modes often mingle, overlap or transform one to another. This relates, for example, to nucleation and condensation amplified by the presence of fine solid particles in air, incorporation of solid particles into drops, or, opposite, evaporation of solid/liquid particles.

Also the deposition of dust amplified by rain is an interesting phenomenon. Although the general belief is that the presence of dust in clouds must increase the precipitation, it is not true in many cases. Comparisons between dusty and almost sterile clouds suggest that this relation is either ambivalent or even opposite. Numerous condensation nuclei imply a large number of droplets which are not so readily merged into millimetre-size raindrops, and the relationship to surface chemistry of drops also plays a role (Rosenfeld et al., 2001; Yin et al., 2002).

It is also quite common that the flow of mostly dry dust is squeezed under almost pristine clouds, and wetting starts when the rain goes through these underlying dust layers. The hydrophobic or untouched dust particles may arrive at the ground or sea in the form of turbulent trains of drops, i.e., not directly attached to, or jailed inside, the drops. Interactions between several sources of the settling dust (mingling), as well as those between the dust components and various aerosols, influence the contents and speciation of iron and other elements. Iron occurs not only within the typical oxyhydroxide/oxide phases (FeO(OH)–Fe(OH)$_3$, Fe$_2$O$_3$ and Fe$_3$O$_4$) (e.g. Hoffmann et

al., 1996; Wang et al., 2006) but also in silicates, sulphates, carbonates, organic compounds, inorganic-organic amorphous precipitates and 'wet' or dried colloids (e.g. Ozsoy & Saydam, 2001).

Combining the analyses of elemental compositions with backward trajectories (EC-BT) is an effective tool to find the best aerosol provenance solutions (Vach et al., 2009). As for the aerosol inputs to these co-mingled phases in wet deposition, the correlation of Fe and Al concentrations indicates a linkage to terrestrial sources, whereas Na, Mg and Ca are usually indicative of marine aerosols. These two findings are quite expectable (e.g., Nadstazik & Falkowska, 2001). However, the EC-BT approach also points to a considerably good linkage of K, Mn and P concentrations to the subcomponents of biogenic origin. This linkage is significantly positive also with respect to the regional monitoring results (Vach et al., 2009).

Yet another fact of general sedimentological importance is worth mentioning. The delivery of iron-rich dust to seas can cause a certain degree of fertilization with capability to change the productivity of the surface waters. This may further change the conditions of the dust input to marine sediment, with implications for the iron cycle and precipitated mineral phases (e.g., Heisler et al., 2008; Larrasoana et al., 2008; Schroth et al., 2009; Nishioka et al., 2009).

Remarkable sedimentation aspects related to settling of specific dust materials on the surface of seas or lakes also include floating pumice tephra chips or fragments (e.g. observed between Tonga and E Australia, Pacific Ocean; lakes in Nicaragua). This 'silicate foam' keeps floating for months, with experimentally confirmed drowning halftimes of 0.3–2 years. This way, these materials may travel or be secondarily redistributed over thousands of kilometres across the ocean. A similar behaviour is also typical for the pollen grains and some spores, but their halftimes are much shorter: days and weeks only. After becoming waterlogged, pollen sinks to the bottom relatively soon (cf. Section 4 for recycling).

9. Conclusions

The present attempt to reassess the natural dust input to sediments confirmed that the emissions from weathered rocks on the land represent the main proportion of airborne material contributing to 'sedimentation background'. The geological approach based on the direct measurements of the dust deposits (not in the air) suggests that also the mass of large mineral-lithic grains must be involved, because the dispersal – wider than formerly assumed – and large added mass substantiate a reassessment of the budgets. The real mass of the transported dust material (RMD; geologically) compared with the total suspended particles (TSP; atmospherically) differs by ca. ×3. This added mass influences the values of the atmospheric burden and also specific inputs to sediments. The mean values for the transported volumes of the volcanic particles are also higher than usually assumed. This is mainly the effect of porous and rugged tephra particles which show a

considerable capability to be dispersed with the jet streams near the stratosphere boundary over long distances or around the globe. The second most significant airborne input is the biogenic dust. Direct evidence of the measured pollen-grain depositions shows how abundant this sort of material is, in spite of the various but substantial microbially mediated reduction of the mass during the 'final' embedding into sediments. The mean contribution of the cosmogenic dust is of by far lesser magnitude, but cannot be neglected. On the other hand, considerably higher values of the wildfire dust material were set according to burden and sedimentary inputs. This increase corresponds mainly to lithic materials which are simultaneously emitted and incorporated into the structure of these natural particulate burning products. Unlike the other components, the minuscule precipitates of solids coming with 'aerosol in narrower sense' seem to represent only a very small contribution to the whole airborne input to sediments.

The suggested categories of the dust and amounts of the material which substantiate the teleconnection among distant basins on the globe are subject to further investigation and clarification. Furthermore, the dust component characteristics also involve the specification of the availability of iron and its possible reconcentration. The study can be continued mainly in the magnetic susceptibility (MS) aspect, because of the large airborne input of paramagnetic and ferri-/ferromagnetic minerals from the weathered rocks on continents, as well as the presence of volcanogenic, wildfire and other materials. The combination of these inputs may be of crucial importance in the assessment of interregional MS stratigraphic links, particularly in limestones.

Presented data are tentative, and refer to ideal mean global values. The first estimates of these geologically reappraised values for the natural dust inputs to the present-day sediments are as follows:

Component (source & material type)	Input (g/m²/yr); 'equivalent to (t/m²/Myr)'
terrestrial (on land) weathering – 'loess'	*3.3*
volcanic ash	*0.3*
biotic components	*0.5*
wildfires	*0.3*
cosmogenic material	*0.0002*
precipitated solids – from 'aerosols n.s.'	*0.02*
Σ (dust; airborne input to sediments)	*(4.4202)* = ca. 4–5 *(?)*

Two principal aspects emerged with this geology-orientated reappraisal of the natural dust issues: First, we found that the dust budgets and inputs to sediment are several times higher when the whole spectrum of the particles and grains from silt to fine-sand sizes (4–250 μm) is taken into consideration. This finding is based on comparison of the sedimentological and atmospheric-science data. Second, we found that the direct measurements (monitoring) of the dust deposition provides a very realistic and promising picture of dust inputs to sediments. This emerging discipline in the field of current geological processes can help to change the traditional mindset of many researchers that these sedimentary background inputs are 'near negligible'. An advantage of the particle deposition monitoring facilities is that the material can be accurately studied by sedimentological and petrological methods before the infiltration of the dust into soils and sediments, blowing or washing away (to dust traps), or mingling with or transformation to other sedimentary materials.

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11. References

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