nature physics

Why do particle clouds generate electric charges?

T. Pähtz¹, H. J. Herrmann¹ and T. Shinbrot^{1,2}*

Grains in desert sandstorms spontaneously generate strong electrical charges; likewise volcanic dust plumes produce spectacular lightning displays. Charged particle clouds also cause devastating explosions in food, drug and coal processing industries. Despite the wide-ranging importance of granular charging in both nature and industry, even the simplest aspects of its causes remain elusive, because it is difficult to understand how inert grains in contact with little more than other inert grains can generate the large charges observed. Here, we present a simple yet predictive explanation for the charging of granular materials in collisional flows. We argue from very basic considerations that charge transfer can be expected in collisions of identical dielectric grains in the presence of an electric field, and we confirm the model's predictions using discrete-element simulations and a tabletop granular experiment.

s long ago as 1850, Michael Faraday commented on the peculiarities of the production and discharge of electric 2 charges during sandstorms¹, a phenomenon repeatedly 3 rediscovered over the intervening century and a half 2-7. Similarly, 4 sand is known to become strongly electrified by helicopters 5 travelling in desert environments, producing spark and explosion 6 hazards⁸, and the issue even has implications for missions to the 7 Moon and to Mars9,10, where charged dust degrades solar-cell 8 viability and clings to spacesuits, limiting the lifetime of their ٩ joints¹¹. Several research groups have investigated mechanisms 10 by which similar particles may charge one another, for example 11 because of non-uniform heating¹², differences in contact area¹³ 12 or particle size^{14,15}, inductive charging of isolated particles¹⁶ or 13 aqueous ion transfer at particle surfaces¹⁷. Recent work has also 14 revealed that identical water droplets can acquire and transfer net 15 charge at minute points of contact¹⁸. 16

Notwithstanding these developments, the phenomenon of 17 granular charging remains poorly understood for want of adequate 18 explanations for two very basic and well-documented facts. First, 19 insulators-which by definition have no free charge carriers to carry 20 out the task-transfer large amounts of charge^{8,12,19}, and second, 21 identical materials-such as grains of sand in the desert-are 22 known to charge one another on contact^{12,13,15,20}. Here, we propose 23 a mechanism to address these twin conundrums. To do so, we note 24 that granular charging predominates for insulating materials under 25 dry conditions, and indeed, first-hand reports state that charges 26 in sandstorms dissipate rapidly on the onset of rain²¹. Under such 27 insulating conditions, charge should not be transported either by 28 the insulating grains or by the dry and insulating environment; on 29 the contrary, charged insulators should be expected to neutralize 30 at points of contact. We therefore propose a mechanism by which 31 neutralization of particles near their points of contact can generate 32 the seemingly paradoxical increase¹⁵ in granular charges. 33

We begin by considering a caricature of a collision of two grains 34 within a strong electric field-as is documented to be ubiquitous 35 within charged dust clouds²²⁻²⁴. As shown in Fig. 1, if the grains are 36 initially electrically neutral and both grains and their environment 37 are sufficiently insulating, the effect of an electric field, E, will 38 be to polarize the grains. We depict this in Fig. 1 as producing 39 negatively charged upper and positively charged lower hemispheres. 40 The simplest case occurs when the grains collide and respective 41



Figure 1 | **Proposed charging mechanism of colliding particles in an electric field.** Initially (left panel) a pair of particles polarized by an external electric field collide (centre panel) to neutralize adjoining hemispheres. Once separated (right panel), the particles again become polarized by the external field. In this way, initially neutral but polarized particles gain one unit of charge following every collision. Blue denotes negative and red positive charge, as indicated by the numbers beside each hemisphere, and the arrows indicate representative particle velocities.

hemispheres become neutralized, as indicated in the centre panel of Fig. 1. Real collisions, between non-spherical particles containing complex charge distributions²⁵, would certainly be much more complicated than this diagram can capture; however, this simplified model has the merits that it can be fully analysed, and as we will show, it provides experimentally testable predictions.

The result of the caricatured collision shown in Fig. 1 is that the top- and bottom-most hemispheres of the granular assembly retain a charge, whereas the contacting hemispheres become neutralized. After the collision, as shown in the right panel of Fig. 1, each individual grain is again exposed to the pre-existing electric field, causing the grains to be repolarized with additional unit charges top and bottom. As the right panel indicates, the result of this process is to increase the negative charge by one unit on the upper particle, and the positive charge on the lower one by the same amount.

This charge transfer occurs for every collision, and so in this scenario, collisional granular flows should pump positive charges downward to ground and negative charges upward to the top of an agitated bed at a predictable rate, proportional to the collision frequency in the dust cloud. We can therefore estimate the rate of particle charging for this model from kinetic

42

43

44

45

46

47

48

49

50

51

52

53

54

55

56

¹Institute für Baustoffe, ETH-Zürich, CH-8093 Zürich, Switzerland, ²Department of Biomedical Engineering, Rutgers University, Piscataway, New Jersey 08854, USA. *e-mail: shinbrot@soemail.rutgers.edu.



Figure 2 | **Simulation results. a**, Granular bed at three representative depths after 2×10^7 computational timesteps, colour-coded by charge. Qualitatively, shallow and deep beds produce less charge than intermediate depth beds. **b**, Quantitative granular charge versus bed depth. Each data point is an average of 750 measurements taken between 5×10^6 and 20×10^6 iterations. The dashed curve is a fit to equation (3) using $\varepsilon = 0.7$, about that of glass. **c**, Solids fraction versus height, fitted with cubic splines. Lower parts of deeper beds (solid) attain a solidified state between the random-loose and random close-packed densities shown. Shallower beds (dashed) are nowhere solidified; thus, grains remain in motion everywhere. The error bars are smaller than the plot symbols throughout.

theory, which provides that the collision rate, R, for moving particles is simply²⁶:

3

$$R = Cn^2 V_{\rm rms} \tag{1}$$

⁴ Here *C* is a constant proportional to the particle cross-section, ⁵ *n* is the number density of particles, $V_{\rm rms}$ is the mean particle ⁶ velocity and *R* has units of collisions per unit time. For ⁷ granular flows, $V_{\rm rms}$ at steady state is achieved by a balance ⁸ between the rate of energy input and the rate of dissipation. ⁹ We consider here the situation in which energy is input from ¹⁰ below—as is documented to occur when windblown particles ¹¹ strike the ground²⁷—and in which dissipation occurs during ¹² inelastic particle collisions.

To make the problem analytically tractable, we specify that whenever a grain strikes the ground, it is ejected upward with velocity, V_o , that is diminished by a fixed restitution coefficient, ε , by each overlying layer of particles that the grain passes through. We assume that the velocity is diminished up to a maximum number of layers, L_{max} , beyond which no further impulse is transmitted. In this case, we can write:

$$V_{\rm rms}(L) = \sqrt{\frac{\int_{1}^{L} (V_0 \varepsilon^{\ell})^2 d\ell - \nu^2}{L}}$$
(2)

where *L* is the number of layers of particles through which an ejected particle may pass and ν is a constant that ensures that $V_{\rm rms} = 0$ when $L = L_{\rm max}$. If we make the first-order approximation (which we validate with computations shortly) that the particle density grows linearly with *L*, then after insertion of equation (2) into equation (1), we obtain: 26

$$R = \alpha L^{3/2} \sqrt{\varepsilon^{2L} - \beta} \tag{3}$$

42

43

where the constant $\alpha = CV_o/2\sqrt{|\ln \varepsilon|}$ is determined by the particle 28 cross-section C, the velocity $V_{\rm o}$ and the coefficient of restitution ε , 29 while the constant β depends on gravity and ε . In practical terms, 30 equation (3) predicts that the charging rate should be small for both 31 very shallow and very deep agitated beds: for shallow beds (that 32 is, small L), the charging rate will be small because the number 33 density will be small and hence particle collisions will be infrequent, 34 whereas for deep beds (large L), the charging rate will be small 35 because collisions will be numerous, and so the finite coefficient 36 of restitution will cause the bed to collapse. We remark that the 37 rapidity of the dropoff in charge at large L is regulated by the 38 parameter β : for large β the dropoff is abrupt; for smaller β the 39 dropoff is more gradual, and that the dimensional charging rate is 40 *R* multiplied by the unit charge imparted per collision. 41

To test this model, we carry out simulations and experiments in which inelastic particles are agitated from below and we evaluate

20

ARTICLES

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

62

63

64

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

83

84

85



Figure 3 | **Experiment.** a-d, Snapshots for 4 (a,b) and 10 (c,d) layers of particles of spouted bed fed from below by an airstream through a porous plenum at the centre of the bottom plate. The plenum and plate are conductive and grounded. The metal plate above the glass jar is connected to a 30 kV generator, and the container itself (about 30 cm in diameter) is sealed except for a <1 mm gap around the bottom edge to let air escape. e, Some grains adhere to the top (circle) or sides (arrow) of the container, after the airflow has been halted. The particles are coloured glass beads of diameter $1.6 \pm 0.1 \text{ mm}$, and the relative humidity is measured by a sling psychrometer to be $51 \pm 2\%$.

the accumulation of charge in the presence of an externallyapplied electric field.

3 Simulation

The simulation we use is modelled after Walton and Braun²⁸, and 4 tracks the motions of polydisperse spherical particles that collide 5 inelastically in three dimensions under the influence of gravity. 6 After each collision, the net charge on each particle is recalculated 7 and a vertical force is applied that is proportional to the product 8 of that charge and an external, vertically oriented, electric field 9 of fixed strength. As described in Fig. 1, charges on upper and 10 lower hemispheres of each particle neutralize during every collision, 11 and each particle is repolarized by adding opposite unit charges 12 to its top and bottom following the collision. To keep upper and 13 lower charged hemispheres aligned vertically, collisions are taken 14 to be frictionless though inelastic with coefficient of restitution 15 0.94 (a value that generates a fluidized bed similar to that used in 16 comparison experiments discussed shortly). Whenever a particle 17 strikes the bottom of the simulated volume, both hemispheres of 18 the particle are neutralized, and to mimic the so-called 'splash' 19 that particles impacting on a sand bed produce during aeolian 20 transport²⁷, the particle is ejected vertically with velocity $V_0 =$ 21 $2.7\sqrt{gd}$, a value that empirically produces granular fluidization over 22 a wide range of parameter values. 23

The simulated volume is periodic in the horizontal directions. 24 We have also carried out simulations using fixed walls; however, 25 the results of these simulations do not differ noticeably from those 26 shown here and we omit them from our discussions. Likewise 27 separate simulations using horizontal dimensions of 8×8 , 11×11 28 and 13×13 mean particle diameters yield indistinguishable results 29 provided that the depth of the bed (discussed shortly) is held fixed, 30 so the data shown are for 8×8 diameter periodic domains. Particles 31 that acquire charge greater than $m \cdot g/E$, where m is the particle 32 mass, g is gravity and E is the applied electric field strength, are 33 removed from the simulation once they are out of contact with all 34 other particles. These particles are then replaced by particles of zero 35 charge beneath the simulation with upward speed V_0 . The nominal 36 depth of the bed is counted in number of layers, L, where one layer 37 consists of the number of particles (about 62) that can be placed in 38 a monolayer in this domain. We assume that this nominal depth 39 defines the number of layers of particles through which a grain 40 ejected at the bottom of the bed must pass (L in equation (2)). /1

Typical results of simulations are shown in Fig. 2a for 288 particles (4.6 layers), 540 particles (8.7 layers) and 828 particles (13.4 layers). Particles are coded depending on their net charge as defined in the colour bar. The mean charge per particle reaches a steady asymptotic state within about 5×10^6 time iterations: we have extended simulations in several representative cases out to 10^9 iterations, and we find no detectable differences in the spatial distributions of particles or their charges. Qualitatively, it is apparent from Fig. 2a that the most strongly charged particles are near the top of the bed, and that many more particles are highly charged (red) for intermediate numbers of layers than for either high or low numbers, as predicted by equation (3). A quantitative comparison between the mean charge per particle from the simulation (blue solid line) and the fit predicted from equation (3) using $\beta = 1$ is shown as a dashed line in Fig. 2b, which confirms the qualitative impression of Fig. 2a.

As we have described, the derivation of equation (3) depends on two essential assumptions. First, the number density is assumed to grow approximately linearly with L, which implies that the depth of the agitated bed should not grow as L is increased. This is what leads to low density and hence weak charging at small L. We remark that there is no analytic framework to predict how the depth of an agitated and charged bed will depend on L; however, our computational results shown in Fig. 2a indicate qualitatively that the depth of the agitated bed remains comparable as L is increased. This can be quantitatively confirmed by evaluating the number density, n, of the bed as a function of height. This is plotted in Fig. 2c, where we have calculated n by dividing the computational volume into horizontal slices and counting the number of particles within each slice. Consistent with equation (3), grains apparently extend to about 13 or 14 mean grain diameters irrespective of L; thus, the volume of the agitated bed does not depend significantly on L and it seems justifiable to set the particle density proportional to L. The second assumption underlying equation (3) is that $V_{\rm rms}$ diminishes with L because of inelasticity of particle collisions. This is what produces weak charging at large L. Figure 2c shows that beds 11 layers or deeper attain a solidified, nearly random close-packed, state, whereas shallower beds maintain a number density below 50%. Thus, as predicted by equation (3), our simulations confirm that shallow beds remain fluid-like and so charge weakly because their number density diminishes with L, whereas deep beds solidify and so charge weakly because their collisional velocities are suppressed.

Experiment

Our model and simulation hinge on simplifications for which the validity remains to be demonstrated. To test our theoretical and computational results, we constructed a 'spouted bed', in which

ARTICLES



Figure 4 | Numbers of levitated grains in experiments and simulations. The computational number of grains (inset) is an average over 9,500 independent realizations; the experimental number (main plot) is summed over 20 successive snapshots taken at 2 s intervals. The experiments were carried out over the course of several days, with the relative humidity ranging between 45% and 53 \pm 2%. The dashed lines are plots of equation (3) from Fig. 2b, rescaled and offset by three layers as described in the text.

coloured glass beads of mean diameter 1.6 mm are fluidized by air 1 blown from below through a porous plenum 6 cm in diameter. 2 As shown in Fig. 3a, the experiment is contained in a 5-mm-thick 3 glass jar about 25 cm diameter at its base, that is separated by 4 a small distance, to allow for air egress, from a grounded metal 5 supporting plate. In each experiment, the airflow is set to the 6 lowest pressure at which the grains above the plenum just become 7 fluidized, so that by design grains charge only by contact with one 8 another or with the grounded plenum. An external electric field is applied by placing a second metal plate that is connected to a 10 30 kV van de Graaff generator above the apparatus and outside the 11 jar. As shown in the enlargements in Fig. 3b,d, shallow beds only 12 weakly fluidize, whereas deeper beds become energetically agitated. 13 In both cases, grains float spontaneously within the chamber and 14 hover or bounce against the upper surface. Movies are included 15 in Supplementary Information. When the generator is turned off, 16 grains remain adhered both to the top surface and to the side of 17 the glass jar (Fig. 3e). We emphasize that because the upper plate 18 was at a high positive potential, only negatively charged grains 19 could remain adhered to the nearby glass, yet the bottom plate 20 is grounded, and there is no source of negative charge anywhere 21 within the glass jar. 22

In this experiment, measuring actual particle charge is prob-23 lematic because particles are deliberately isolated inside a glass 24 enclosure and the entire experiment is exposed to a strong electric 25 field that would interfere with any sensitive charge measurement. As 26 a surrogate for particle charge, we evaluate the number of levitated 27 particles within a fixed window between the granular bed and 28 the top of the glass jar. This number is manually counted in 20 29 successive snapshots taken at 2 s intervals, and the resulting average 30 is plotted in Fig. 4 as a function of the number of layers, L, of 31 grains. To obtain this plot, L is determined by weighing the number 32 of grains that will fit in a monolayer on the bottom plate of the 33 experiment. Successive multiples of this weight of grains are then 34 loaded and levelled in the apparatus, so that one monolayer gives 35 L = 1, two monolayers give L = 2 and so on. This is the identical 36

37

38

39

40

41

42

43

44

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

62

63

64

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

83

84

85

86

87

88

89

90

91

92

93

94

95

96

97

98

99

100

procedure used to define numbers of layers in the simulations that we have discussed. Once a fixed number of layers is loaded into the apparatus, the pressure is then adjusted, as we have described, to the minimum value at which grains remain fluidized, and then the van de Graaff generator is turned on, snapshots are taken and numbers of levitated grains are counted and averaged.

In the inset of Fig. 4, we show for comparison the number of levitated grains from the simulations previously described. For the simulation, we count the number of particles in a fixed-size window between 15 and 21 mean particle diameters from the bottom of the bed—a distance above any solidified substrate as shown in Fig. 2c. In both the main plot and the inset, we overlay the dashed curve from Fig. 2b, rescaled and offset to account for the fact that charges below a fixed threshold cannot be expected to levitate finite-weight particles.

We have introduced a simplified model that seems to accurately predict the charging of granular materials in collisional flows in the presence of an electric field such as are encountered in particle clouds such as sandstorms, volcanic plumes or industrial fluidized beds. Our simulations and experiment confirm the essential features of the model, namely that identical grains in the presence of an applied electric field can pump charge upward through repeated collisions in the absence of any conductive mechanism of charge transfer either in the particles or their environment. We find as predicted that shallow agitated beds—as could be expected in weak winds or for heavy grains—charge weakly, as do very deep agitated beds—as would be expected for highly dissipative materials. Under intermediate conditions, however, we observe dramatic charging, with the most highly charged particles found preferentially near the top of the agitated bed.

We emphasize that this charging mechanism has nothing to do with electrochemical differences in surface states, or variations in sizes or types of contact. Such differences do unequivocally lead to charging, however not for identical materials under consideration in this study. All that is needed is repeated collisions between dielectric particles in the presence of a sufficiently strong electric field. The charging effect reported here seems to be robust: indeed, the experiments shown were carried out at moderate humidity, between 45 and 53% relative humidity, but similar effects have also been seen in our laboratory at relative humidities down to about 20%. Moreover, despite the fact that we used beads large enough to facilitate counting in the experiments described, we have reproduced the vigorous charging and levitation of grains using smaller and irregular particles as well.

In closing, we stress that although this work explains how grains in an electric field can acquire strong charges, it does not define mechanisms that may generate the required electric field. Such fields are well documented to exist^{22–24}, yet their cause in natural sandstorms is poorly understood. In some cases, the source may be external, as in reports that nearby thunderstorms⁵ or charged bodies²⁹ can provoke granular charging. In other cases, it remains to be determined how a sandstorm might both generate strong charges and produce the electric field that engenders the charging to begin with. We hope that larger scale studies can both probe the accuracy of the simple model presented here and identify mechanisms by which a self-sustaining electric field may be established.

Received 17 November 2009; accepted 26 February 2010; published online XX Month XXXX

References

- Baddeley, P. F. H. Whirlwinds and Dust-Storms of India 3–4 (Bell & Daldy, 1860).
- Shaw, P. E. The electrical charges from like charges. *Nature* 118, 659–660 (1926).
- 3. Gill, E. W. B. Frictional electrification of sand. *Nature* **162**, 568–569 (1948).

NATURE PHYSICS DOI: 10.1038/NPHYS1631

2

4

5

6

8

9

10

11

12

13

14

15

16

17

18

19

20

21

22

23

24

25

26

27

28

29

- 4. Anderson, R. et al. Electricity in volcanic clouds. Science 148, 1179-1189 (1965).
- Kamra, A. K. Visual observation of electric sparks on gypsum dunes. *Nature* 240, 143–144 (1972).
- Brook, M. & Moore, C. B. Lightning in volcanic clouds. J. Geophys. Res. 79, 472–475 (1974).
- Tomas, R. J. *et al.* Electrical activity during the 2006 Mount St Augustine volcanic eruptions. *Science* 315, 1097 (2007).
- 8. Palmer, K. N. Dust Explosions and Fires (Chapman & Hall, 1973).
- Eden, H. F. & Vonnegut, B. Electrical breakdown caused by dust motion in low-pressure atmospheres: Considerations for Mars. *Science* 180, 962–963 (1973).
- Forward, K. M., Lacks, D. J. & Sankaran, R. M. Particle-size dependent bipolar charging of Martian regolith simulant. *Geophys. Res. Lett.* 36, L13201 (2009).
- Mills, A. A. Dust clouds and frictional generation of glow discharges on Mars. Nature 268, 614 (1977).
- Lowell, J. & Truscott, W. S. Triboelectrification of identical insulators. J. Phys. D 19, 1273–1280 (1986).
- Shaw, P. E. Electrical separation between identical solid surfaces. *Proc. Phys. Soc.* 39, 449–452 (1927).
- Lacks, D. J. & Levandovsky, A. Effect of particle size distribution on the polarity of triboelectric charging in granular insulator systems. *J. Electrostatics* 65, 107–112 (2007).
- Kok, J. F. & Lacks, D. J. Electrification of granular systems of identical insulators. *Phys. Rev. E* 79, 051304 (2009).
- Wu, Y., Castle, G. S. P., Inculet, I., Petigny, S. & Swei, G. Induction charge on freely levitating particles. *Powder Technol.* 135–136, 59–64 (2003).
- Whitesides, G. M. & McCarty, L. S. Electrostatic charging due to separation of ions at interfaces: Contact electrification of ionic electrets. *Angew. Chem. Int. Ed.* 47, 2188–2207 (2008).
- Ristenpart, W. D., Bird, J. C., Belmonte, A., Dollar, F. & Stone, H. A.
 Non-coalescence of oppositely charged drops. *Nature* 461, 377–380 (2009).
- 19. Harper, W. R. Contact and Frictional Electrification (Clarendon, 1967).
- 20. Shinbrot, T., Komatsu, T. S. & Zhao, Q. Spontaneous tribocharging of similar
 materials. *Europhys. Lett.* 83, 24004 1-4 (2008).
- Baddeley, P. F. H. Whirlwinds and Dust-Storms of India 11
 (Bell & Daldy, 1860).

- Zheng, X. J., He, L. H. & Zhou, Y. H. Theoretical model of the electric field produced by charged particles in windblown sand flux. *J. Geophys. Res.* 109, D15208 1-9 (2004).
- Rasmussen, K. R., Kok, J. F. & Merrison, J. P. Enhancement in wind-driven sand transport by electric fields. *Planet. Space Sci.* 57, 804–808 (2009).
- 24. Latham, J. The electrification of snowstorms and sandstorms. Q. J. R. Meteorol. Soc. **90**, 91–95 (1964).
- 25. Ireland, P. M. The role of changing contact in sliding triboelectrification. *J. Phys. D* **41**, 025305 1-11 (2008).
- Maxwell, J. C. On the dynamical theory of gases. *Phil. Trans. R. Soc. Lond.* 157, 49–88 (1867).
- 27. Anderson, R. S. & Haff, P. K. Simulation of Eolian saltation. *Science* 241, 820–823 (1988).
- Walton, O. R. & Braun, R. L. Viscosity, granular-temperature, and stress calculations for shearing assemblies of inelastic, frictional disks. *J. Rheology* 30, 949–980 (1986).
- 29. Shinbrot, T., LaMarche, K. & Glasser, B. J. Triboelectrification and razorbacks: Geophysical patterns produced in dry grains. *Phys. Rev. Lett.* **96**, 178002 1-4 (2006).

Acknowledgements

We thank E. Strombom for her dedicated experimental work, and we thank the National Science Foundation, Division of Chemical and Transport Systems and the Eidgenössische Technische Hochschule, project ETH-10 09-2 for financial support.

Author contributions

T.P. carried out the simulations. H.J.H. directed the simulations and provided geophysical expertise. T.S. conceived the project, constructed the experiment, carried out the analysis and prepared the initial manuscript. All authors discussed the results and implications and commented on the manuscript at all stages.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturephysics. Reprints and permissions information is available online at http://npg.nature.com/reprintsandpermissions. Correspondence and requests for materials should be addressed to T.S.

NATURE PHYSICS | ADVANCE ONLINE PUBLICATION | www.nature.com/naturephysics

Page 1

Query 1: Line no. 12

'due to' changed to 'because of' here. OK?

Query 2: Line no. 18 Can the idiom 'for want of' be changed to 'because of the lack of' here?

Query 3: Line no. 34

Can 'a caricature of' be deleted here, or should 'caricature' be changed to, for example, 'schematic diagram' or 'representation'?

Query 4: Line no. 48

Can 'caricatured' be deleted here, for consistency with the query above.

Query 5: Line no. 50 'while' changed to 'whereas' here. OK?

Query 6: Line no. 54

According to style, 'additional' should be changed to 'extra', 'further' or 'more'. Which is most appropriate here?

Query 7: Line no. 59 Should 'the' be inserted before 'ground' here?

Page 2

Query 8: Line no. 1

In the second last sentence of figure 2c's caption, can 'Shallower beds (dashed) are nowhere solidified' be changed to, for example, 'Shallower beds (dashed) are not solidified anywhere'?

Query 9: Line no. 30

According to style, 'while' can be used only in reference to time. Is it best changed to 'whereas' or 'and' here?

Page 3

Query 10: Line no. 4

The style guide states: 'In general, avoid using authors' names and phrases such as 'A. N. Author has shown that' in the text. For example, say 'Water flows downhill2' rather than, 'Garwin has shown2 that water flows downhill.' There are occasions when this rule can be broken, for example when two authors' results are being compared or when the significance of an author's contribution needs emphasis.' Please check the use of 'Walton and Braun' here.

Query 11: Line no. 10

Should the text here be 'the charges on the upper..'?

Query 12: Line no. 15 Is 'though' best changed to 'although' or 'but' here?

Query 13: Line no. 20 Is 'impacting on' best changed to 'landing on' or 'striking'?

Query 14: Line no. 77

'due to' changed to 'because of' here. OK?

Query 15: Line no. 87

'whose' changed to 'for which the' here, to avoid the use of the former according to style. OK?

Page 4

Query 16: Line no. 16

The web address has been removed from this sentences. There will be links to your Supplementary info movie files on the nature website.

Query 17: Line no. 65

According to style, 'dramatic' should be changed to 'marked' or 'pronounced'. Which is most appropriate here? (If neither is appropriate, please provide an alternative word.)

Page 5

Query 18: Line no. 7

Please provide page range for ref. 7.

Query 19: Line no. 36

Does the page number provided in reference 21 represent the total number of pages in the book or the first page of the relevant page range? If the latter, please provide the final page number of that range.